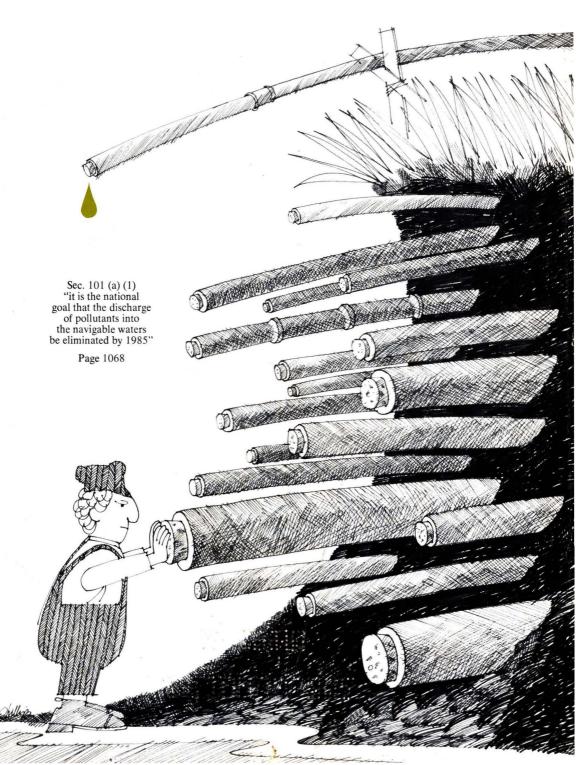
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Volume 6, Number 13 December 1972 ESTHAG 6(13) 1051-1148 (1972)

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contents

Feature

1078 Tackling resource recovery on a national scale by Richard Lesher, NCRR, Inc.

Interview

1065 Melvin Evans, governor of the Virgin Islands, talks about environmental priorities in an American paradise

Outlook

- 1068 Congress gets its way with new water legislation
- 1071 Computer model predicts air quality over highways
- 1072 Dow's latex plant completely recycles process water
- 1074 ICPRB lacks power to enforce Potomac cleanup
- 1076 Cleaning up the environment—British style

Departments

Editorial 1057	Bookshelf 1129
Currents 1059	Meeting guide 1131
Industry trends 1125	Professional consulting services 1133
New products 1126	Classified section 1136

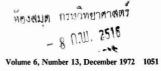
New literature 1128

Current research contents 1052

Annual Index, Vol 6, 1972

Names 1137

Subject 1140



Solid waste—plastics composites: Physical properties and feasibility for production 1085

L. T. Fan, D. G. Retzloff, and W. O. Vanderpool

A plastic composite of solid waste and monomer is another answer for today's mounting solid waste disposal problems. Formed by radiation-induced polymerization of the monomer, the composite could be used as a substitute for wood-based materials. Composites have been formed from waste paper (which make up approximately 50% of urban solid waste), and the possibility of forming plastic composites with other solid waste materials is discussed.

Influence of Coriolis forces on airflow in conifuges 1091

W. Stober and T. T. Mercer

Conifuges are used to measure aerodynamic particle sizes; the first operational design came in 1950. Particles of equal aerodynamic size are located on concentric rings on the envelope of the cone. An updated mathematical model of the airflow in such particle size spectrometers is presented.

Growth rate determinations of the macrophyte Ulva in continuous culture 1096

T. D. Waite, L. A. Spielman, and R. Mitchell

Continuous culture experiments now reveal that estimates of biomass synthesis may be in error when the algal mass is assumed to be carbohydrate. This method may prove useful for assessing the low growth rates of phytoplankton.

Aerosol filtration with slip flow

R. D. Chmielewski and S. L. Goren

A model is proposed for determining the effects of slip on low-speed filtration of an aerosol at fibrous filter surfaces. New measurements of the parameters—filter efficiency and pressure drop—confirmed data predicted by some earlier investigators but failed to predict certain data of others. With slip, the filter efficiency increases as the pressure drop decreases.

X-ray spectrometric analysis of air pollution dust

C. L. Luke, T. Y. Kometani, J. E. Kessler, T. C. Loomis, J. L. Bove, and B. Nathanson

The presence of metals in atmospheric particulate matter can now be measured by the direct nondestructive technique of X-ray fluorescence spectrometry. The major advantage of this technique is the elimination of the time-consuming and expensive ashing step. Data obtained with this technique compare favorably with the same samples after ashing.

Removal of heavy metals from water and brine using silicon alloys 1109

J. P. McKaveney, W. P. Fassinger, and D. A. Stivers

Silicon alloys of calcium and magnesium can be used to remove heavy metals from waste waters and brine solutions. What happens is that the heavy metal exchanges with the calcium and magnesium in the alloy; the latter enters the water. The alloy can be used in column or batch-type operations and will remove Ag, As, Au, Cd, Cr, Fe, Hg, Pb, V, and Zn.

Notes

Improving linear relations to obtain kinetic parameters 1114

D. M. Di Toro

Graphical techniques are used to analyze kinetic data that are routinely applied to water quality studies. Statistical analysis of resulting kinetic parameters becomes an important adjunct to any curve-fitting technique such as the direct least-squares error minimization.

Concentration of arsenic from water samples by distillation 1116

E. J. Farkas, R. C. Griesbach, D. Schachter, and M. Hutton

The experimental technique for the determination of arsenic in drinking water is improved by first concentrating the sample into a smaller volume by distillation. Optimum values are presented.

Particle counting by glow discharge perturbation 1117

N. A. Fedrick, A. T. Bell, and D. N. Hanson

A device for counting and sizing airborne particulate matter makes use of a dc glow discharge maintained between two electrodes separated by a disc containing an orifice of approximately the same size as the particles to be counted. Passage of a particle through the orifice causes a perturbation in the discharge current; the magnitude of the perturbation is proportional to the particle diameter.

Prediction of heat of combustion of solid wastes from ultimate analysis 1119

D. L. Wilson

A formula has been developed for calculating the heat release per pound of solid waste material. Values obtained with this formula are adequate for assessing incinerator efficiency and design calculations.

Particles containing lead, chlorine, and bromine detected on trees with an electron microprobe 1121

G. H. Heichel and L. Hankin

There is further evidence that the lead burden of plants near highways is increased from vehicle exhausts. It has now been found that at least part of the lead on or embedded in the bark of trees growing near roads is combined with chlorine and bromine in particles similar in size and composition to particles in automobile exhausts.

Distribution of mercury in the sediments of New Haven (Conn.) Harbor 1123

M. D. Applequist, A. Katz, and K. K. Turekian

Core samples in the Long Wharf area on the west side of the harbor show that there does not appear to be any evidence of an additional source of supply of mercury to the New Haven Harbor sediments other than municipal sewage treatment plants.

Correction

Recent Sedimentary History of Lake Mendota, Wis. 1124

G. C. Bortleson and G. F. Lee

This article appeared in ES&T, September 1972, p 799.

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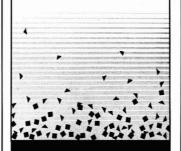
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editorial

In support of environmental R&D

Existing technical information is an inadequate basis for the cheap and efficient technology of the future

e don't know how many of our readers are familiar with ES&T's stablemate, *Chemical Technology*, but we do know that those who don't get to see it every month are missing out on some stimulating reading. One of the things they would miss is "The Industrial Chymist" column, written in thought-provoking, breezy style by *Chemtech* editor Ben Luberoff.

The purpose of this plug for a sister publication stems from more than nepotism; it is to bring our readers' attention to some comments Luberoff makes in his October 1972 column that are highly appropriate to the environmental field. Luberoff's contention, in essence, is that we too often rush off after new facts before we have even tried to do anything with the facts we already have in hand. In our opinion, this is what has happened all too often in the environmental arena and it specifically characterizes the strong penchant of scientists to want to study every aspect of a subject, whether or not it has been done previously. Fat government grants have in the past added immeasurably to the supply of technical reports which, once issued, seem to vanish forever into some remote, inaccessible vault.

The reckless abandoning of old information in favor of the search for new information, we are happy to say, has been recognized by the federal EPA for what it is—a vast waste of time and money. The agency is now taking the view that just about everything that needs to be known to solve environmental problems is lying around somewhere, if only we can put our hands on it!

Our imputing this view to EPA is based on observation of the change in emphasis in the agency's programs, away from new technical research and development and toward consolidation of information and mechanisms designed to transmit information to those who can use it. The reduced visibility of the old FWQA industrial pollution control demonstration grant program and the rise in emphasis on the new technology transfer program is a perfect case in point. Indeed, we applaud a good deal of this change as a step in the right direction (see "Federal demonstration projects: what has been achieved?", ES&T, June 1971, p 498). We can heartily agree that entirely too much effort has been spent "rediscovering the wheel" and too little in putting information that has been known for years to work in the cause of environmental protection.

However, we cannot agree with those who say that because there exist large amounts of scientific and technical data which are largely untapped, there is therefore no further need for scientific and technical research and development. We can concede that even if no more environmental R&D were ever performed, we could nevertheless use existing technology to clean up all the grosser forms of pollution that plague the country. But the cost would be horrendous, because primitive technology-and that's what we're talking about-is expensive technology. History shows that R&D effort has to be expended to make technology both more efficient and cheaper. Since, as must have been said a million times, we will all pay for the costs of environmental cleanup, we all have a very personal stake in ensuring that the technology which does the cleaning up is as efficient and as inexpensive as we can possibly make it. It is difficult to see how that technology can be discovered merely by examining what has been done in the past.

By all means let's learn what we can from the existing body of environmental information. But let's not at the same time kid ourselves that that's all we need to know.

) H. Micrael Bowen



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WASHINGTON ICC freight ruling irks CEQ Chairman Train

The Interstate Commerce Commission recently approved railroad freight rates which would appear to be oblivious to the U.S. commitment to resource recovery. It has been generally felt that freight rates discriminate against secondary materials, (such as ferrous scrap, for example, in favor of iron ore) and the recent rate increase for transportation of secondary materials offers no relief. Charged with the responsibility of receiving and commenting on environmental impact statements required by NEPA, Train maintains that analyses made by ICC are inadequate from which to draw conclusions concerning the impact of freight rates on recycling and environmental quality. At press time, the rate increase had been set aside until next June.

EPA construction grants and nondegradation clause

Construction grants awards

(all figures in millions of dollars)

Ala.	\$24.8	Neb.	\$10.8
Alaska	5.0	Nev.	4.4
Ariz.	2.6	N.H.	6.1
Ark.	2.4	N.J.	31.1
Calif.	116.1	N.M.	4.6
Colo.	14.5	N.Y.	64.1
Conn.	14.4	N.C.	20.7
Del.	4.4	N.D.	1.2
D.C.	31.7	Ohio	82.2
Fla.	26.4	Okla.	21.3
Ga.	25.6	Ore.	13.3
Hawaii	5.8	Pa.	75.3
Idaho	4.1	R.I.	2.6
Ш.	84.9	S.C.	11.5
Ind.	30.5	S.D.	2.3
lowa	23.6	Tenn.	13.7
Kan.	17.6	Tex.	95.1
Ken.	27.8	Utah	0.8
La.	15.1	Vt.	5.1
Me.	12	Va.	32
Md.	40.2	Wash.	25.1
Mass.	27.4	W.Va.	1.3
Mich.	87.2	Wis.	35.8
Minn.	18.2	Wyo.	0.4
Miss.	5.1	Guam	3.6
Mo.	21.5	Puerto Rico	7.2
Mont.	7.1		

Since December 1970 when it was formed through the end of fiscal year 1972, the Environmental Protection Agency has provided \$1.3 billion in construction funds to help cities and states build \$3.5 billion worth of waste treatment facilities. This translates to 2000 projects serving 70 million Americans. During the same period, EPA made awards totaling \$571 million for projects that were begun before EPA was formed. Meanwhile, on the air front, language for a "nondegradation" clause is being prepared. A recent appeals court ruling (November 3) upheld an earlier June decision that air cannot be degraded. EPA had till the end of last month to promulgate regulations against significant air deterioration in any state. EPA's general counsel John Quarles had warned earlier that the EPA would not be in a position to meet the deadline and that the appeals court ruling might be referred to the Supreme Court.

New noise and pesticides laws are on the books

These two environmental bills were sent to President Nixon in the raft of bills to be signed before last month's general election; both of them were signed into law after Congress had adjourned. P.L. 92-516, the pesticides law, for the first time addresses the problem of misusing properly registered pesticides; it also controls pesticides that move solely in intrastate commerce. P.L. 92-574, the noise law, authorizes the EPA administrator to set noise standards for numerous classes of products including consumer items such as lawnmowers, can openers, and the like. Awaiting the 93rd Congress when it opens for business next month are a couple of environmentally related issues including a national energy policy and a national land use policy.

Commerce gears up for international trade study

What effect cleanup measures mandated by recent air and water laws will have on domestic industry and its competitive position internationally is a subject of prime concern to the U.S. industries. Now, a Department of Commerce study will attempt • to document the effect of such cleanup programs on production cost and the market price of goods on an industry-by-industry basis, and • to identify the probable extent of competitive advantage any foreign article may have and whether competing foreign articles are produced under clean laws or not. To be prepared in cooperation with other federal agencies, industries, and public centers, the study concentrates on nine countries including Japan, Germany, France, Spain, Canada, and the U.K. Due April 18, 1973, their report stems from a requirement in the new water law, P.L. 92-500 (this issue, p 1068).



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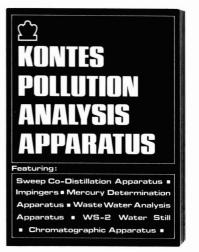


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PROMPT SHIPMENT FROM STOCK

STATES Federal facilities disregarding state laws in Pa., Ky.

The Pennsylvania Department of Environmental Resources has filed suit against the Secretary of the Army, the operating contractor for the Army's ammunition plant at Scranton, and the commanding officer of the plant for violating the state's Clean Streams Law. Approximately 1.5 million gal. of oily industrial wastes are discharged daily by the plant.
Faced with refusal by many of the 169 federal facilities in Kentucky to obtain permits required from all potential air pollution sources, the state Air Pollution Control Commission has asked assistance from the Attorney General and the administrator of the federal EPA.

Delaware solid waste recovery plant finally financed

The state of Delaware has received a \$9 million grant from the federal EPA for a resource recovery system that will convert solid wastes and sewage sludge into compost and will recycle metals and glass. To be operated by Hercules, Inc. (Wilmington, Del.), the plant will handle 485 tons of domestic solid waste, 15 tons of light industrial solid waste, and up to 55,000 gal. of sewage sludge daily. Earlier plans which were developed two years ago and abandoned owing to a lack of funds, are now being financed. The state's share is \$3.8 million.

Hawaii, Fla., N.J. crack down on water discharges

• The U.S. Justice Department is suing five Hawaiian sugar companies— Hilo Coast Processing Co., Paauhau Sugar Co., Laupahoehoe Sugar Co., Honokaa Sugar Co., and Hamakua Mill Co.—for polluting coastal waters (December 1971, p 1174).

• The Florida Department of Pollution Control has issued orders to 42 municipalities prohibiting additional connections to sewage treatment facilities. Allegedly, their plants will not meet Florida's January 1, 1973 deadline for secondary treatment.

• Major industries along the New Jersey side of the lower Delaware River were given deadlines by the N.J. Department of Environmental Protection to halt pollution of the Delaware River. The orders followed abandonment of plans by the Delaware River Basin Commission to build a multimillion dollar regional treatment plant in southern New Jersey.

Autos tested in Calif.; air polluters registered in Mo.

• General Motors Corp. and the state of California are testing and evaluating advanced emission control systems on a limited number of GM cars in state fleets. Using platinum and other noble metals, the catalytic systems will be periodically tested to evaluate the emission control systems as mileage accumulates.

• Permits are now required in Missouri for the construction of all new air contaminant sources in the state and for modifying existing sources. Ohio took similar action earlier this year (July 1972, p 586).

Pennsylvania moves ahead with environmental protection

Effective October 1972, state erosion and sedimentation control regulations require that all earth-moving activities in Pennsylvania be conducted with approved erosion and sediment control measures to prevent accelerated erosion and sediment runoff. A recently enacted law gives the Pennsylvania Environmental Quality Board the power to adopt rules and regulations for air pollution abatement and establish maximum quantities of air contaminants permitted under certain conditions.



Protecting Hawaii's coast

Deadlines for New Jer industries	sey
Shell Oil Ço.	3/31/74
Du Pont Chambers Works	12/1/75
Du Pont Charney's Point Works	6/15/74
Du Pont Repauno Works	3/31/75
Texaco Inc.	9/30/74
Monsanto Chemical Co.	8/1/75
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Olin Corp.	12/3/73

TECHNOLOGY SO₂ removal processes continue apace

Union Carbide Corp. and J. F. Pritchard & Co. (Kansas City, Mo.) are touting a new process for removing sulfur dioxide from sulfuric acid manufacturing plant off-gases. Key to the process is a unique regenerable organic solvent manufactured by Carbide and licensed by Pritchard. The solvent absorbs SO₂ from the off-gases and may be regenerated by a combination of heat and steam stripping. The SO₂ removed can be recovered and used to make more sulfuric acid, boosting the efficiency of the acid plant to about 99.8% Pritchard says the process reduces SO₂ concentration in the emission gases to 150 ppm, about half the level currently permitted by the federal EPA.

Device curbs emissions using leaded fuels

Questor Corp. (Toledo, Ohio) has unwrapped an automobile emission control device which the company says is effective with leaded or unleaded fuel. The device is a 3-stage package with a thermal exhaust reaction, followed by catalytic reduction, and a final thermal treatment. A proprietary catalyst is mounted in a stainless steel grid-type housing. Questor says the device will bring emissions below the 1975–76 standards and works best with fuels which contain less than 0.5 gram of lead/gal. The thermal reactors are run rich with a 15% fuel penalty, but the fuel consumption can be reduced. Currently, 50,000 mile tests are under way, but testing won't be complete soon enough to allow production runs for installation in '75 or '76 models.

RESEARCH Recovering materials from phosphate sludge

Experimental work which could lead to the practical recovery of phosphates and metals from phosphate sludge has been conducted by the U.S. Bureau of Mines. Taking waste sludge generated by phosphate coating processes, BuMines scientists were able to recover trisodium phosphate, zinc, and iron as separate products. The sludge is first dissolved in hydrochloric acid. The iron is extracted as ferric chloride using isopropyl ether; zinc is extracted by 2-diethylhexyl phosphoric acid in kerosine, and phosphate is recovered by crystallization from the raffinate. The value of the by-products recovered far exceeds reagent costs for extraction, according to BuMines.

Developing sensitive tests for tritium, Hg

Scientists at the University of California, Berkeley, have devised a simple, inexpensive method for analysis of tritium in the air. Tritiated water vapor from the air is exchanged with nontritiated water in a silica gel column. When the gel is half spent, it is stirred in water until tritium equilibrium is reached, diluted into a dioxane solution, and counted for 20 min by liquid scintillation. Sensitivity is about 1 nCi/m³.
Ultratrace detection of mercury at levels of about 10 picograms in samples of 1–10 mg is possible with a technique developed at the Ames Laboratory of Iowa State University. In the method, a liquid sample is dropped on a tantalum strip in a furnace. When the strip is heated, the mercury vapor released is adsorbed by a gold strip which is then heated, releasing the mercury produces fluorescence which is directly measurable by a spectrophotometer.



Ultratrace mercury detection

INDUSTRY

Paper company turns its mill into recycling center

Crown Zellerbach will convert its Antioch, Calif., paper mill into one of the largest waste paper recycling centers in the U.S. The company already recycles some waste paper at the mill-about 150 tpd-but is planning to spend about \$9.2 million to install new equipment in a two-phase plan that would boost the mill's recycle capacity to roughly 600 tpd. Waste paper from the mill will consist largely of corrugated shipping containers discarded in the San Francisco Bay Area. Crown Zellerbach says the two-step conversion process will be complete about the middle of 1974. Phase-out of the papermaking operations will coincide with phase-in of the recycling operation.

Glass reclamation plant to go on-stream

Construction has begun on the country's first mechanized glass reclamation plant adjacent to the demonstration solid waste processing plant at Franklin, Ohio (October 1971, p 998). Funded jointly by the Glass Container Manufacturers Institute and the EPA, the reclamation center will sort waste glass from the glass-rich residue at the Franklin project. Techniques for sorting glass into three colors-clear, green, and amberinclude high-density magnetism, air currents, screening, and optical sorting. Expected to be on-stream early in 1973, the plant will process about 4 tons of glass daily which will be shipped to nearby glass container plants for recycling. The plant will cost about \$330,000.

Steel companies collaborate on coking cleanup

Four of the nation's top steel and coal producers have joined forces to develop a better coking process which will control gas and dust operations presently associated with conventional slot-type coke ovens. Bethlehem Steel Corp., National Steel Corp., Republic Steel Corp., and Consolidated Coal Co. have let a contract for a 500-tpd pilot coking plant to Dravo Corp. and the Lurgi Mineralotechnik Co. (Germany). The process is designed to produce a strong coke pellet from a single highvolatility coal followed by calcining. Because the process does not use conventional briquetting machines, a higher proportion of binder can be used—an important consideration because more binder strengthens the final product. Costs for the project and a related full-scale blast furnace demonstration program are estimated to be about \$37 million.

Firms get federal money for environmental studies GRANTS

• EPA has named two consulting engineering firms-Dames & Moore (Los Angeles, Calif.), and Ryckman, Edgerly, Tomlinson and Associates (St. Louis, Mo.)-to make joint emergency investigations of oil, chemical, or dangerous materials spills that may occur on the Great Lakes or other midwestern waterways. The team would assemble a field unit for on-sight inspection and make a full report to EPA on mishaps in Regions V, VII, and VIII.

• The Navy will conduct studies with the fluidized bed incineration system developed by AWT Systems, Inc. (Wilmington, Del.) with a view toward destruction of hazardous wastes arising from explosives or fuels manufacture, pesticides, herbicides, and the like.

• Westinghouse Environmental Systems, (Pittsburgh, Pa.) has received a contract from the Atomic Energy Commission for a state-of-the-art study of the use of salt water in power plant cooling towers.



Coking pilot plant in Md.

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Governor Melvin Evans

Melvin Evans is governor of the U.S. Virgin Islands. He was appointed governor by President Nixon in July 1969 and then duly elected in the general elections of 1970. A native of the islands, a medical doctor by profession, Evans tells ES&T's Stan Miller of his concern for what is truly an American paradise —the islands of St. Croix, St. John, and St. Thomas. The Virgin Islands offers an example where concern for further development focuses on an area limited in size and where growth of social services must keep pace with additional physical and economic growth.

Governor Evans has done something unique by creating a Governor's Citizens Advisory Council. It was established by Executive Order and signed by the Governor on May 19, 1972. The Council involves the citizens in a constant interplay with the Governor so that he is aware of their feelings on matters such as recreation, environmental quality, and community development.

Priorities

What are the main priorities of the island government?

Put broadly, as we see them now, the main priority is to improve the quality of life, to allow us to catch up with the many things that we had fallen behind on as we grew. Specifically, this means providing more and better roads, adequate waste water treatment facilities, schools, and expanding hospital services to meet the expanded population. In addition, it includes bringing in industry and business to provide employment for a constantly expanding work force and then in a broader way, improving the interhuman relationships which in any growing community tend to have stresses and strains. Obviously, another priority which, in a way has to do with the interpersonal relationships, would be improving law and order. I don't like to use the term because it means different things to different people, but we want to get out of the mainstream of the rising tide of crime.

How fast is the Virgin Islands growing?

In our opinion, actual growth since 1960 is almost 300%. Despite the 1970 Census figure of 64,000, every indication, every index points to a population in the neighborhood of 95,000–100,000. The 1960 Census, which we think was much more accurate, listed a population of 33,000. We feel we have almost tripled in this 12-year period. Our school population, for example, has gone up 120% in three years. In September 1969, our public school population was approximately 10,000 students; this year, it's 22,000. This is public schools alone.

Overall, the islands have grown and we expect growth to continue. We have no intention of stopping growth but we must control the growth. In the past, in the uncontrolled growth, the social services and amenities of life did not keep pace with the physical and economic growth, both of which



have operated to produce many disadvantages. Basically, we need now to take stock, to consolidate, and improve the quality of life; to emphasize quality more than quantity. Before, we were just content to grow, grow, and grow.

On the priorities that we mentioned, we are pretty well along in seeing them through. I think that this is true for roads, sewage, schools, hospitals, and housing. We still need to do a great deal on the interpersonal, interhuman relationships, and the matter of delinquency, crime, and drug abuse. We are attacking these latter things now but have not made quite the same headway as we have made on some of the earlier things.

Recreational development

Surely tourism is the No. 1 business in the islands, but what assurance is there that residents will have adequate recreational facilities?

For the first time in the history of the Virgin Islands and largely because of the work of William Barber, the island government has come up with a 5-year recreational plan. It is based on an inch-by-inch study of the islands to relate, by accepted formulas, the population of any area to its population density, so that we can tell in terms of numbers of people, the need for the number of softball diamonds, for example, and where they should be put. The study revealed the need for a certain number of multipurpose courts and where they should be put. We needed so many swimming pools, tennis courts, and so on, and the study showed where they should be located. We were trying to get away from the hit or miss business of putting a little playground here or there because someone demanded it.

To what extent are some of these things being accomplished?

We are working. First, we had to have approval of the plans, which we got. Then, we had to have authorization of the funding for legislation, which we got. At the present time, we are actually implementing many of these with the Bureau of Outdoor Recreation of the Department of the Interior. We are putting in major recreational facilities at Tutu, where we have a large population center of 8000–9000 people. We are about to put something into a section called Savaan. Of course, there are certain financial constraints in how rapidly we can implement the plan. Particularly in St. Thomas, we have a problem of land acquisition because flat land is not easy to come by.

What opportunities are available for citizens of the islands?

At the present time, there are several things I might touch on. One is an intensive reevaluation of our public school system including, but not limited to, such things as building additional modern facilities, providing adequate recreation, and getting ready to build a skill center and a vocational training center in St. Thomas. We are seriously considering adoption of a 12-month school year in order to provide still better opportunities. Our college has been zeroing in on teacher education, specifically, providing training in the communities. We have a very active nursing education program, a fully accredited program, that leads to an associate degree. The colleges are also taking steps to provide training in police work, accounting, construction work, hotel work, and in other areas where the need in the communities exists. There is also a very active territorial scholarship program to assist people to go away and get training which we need here but which might not be available here.

What is the status of environmental protection measures taken by the island government?

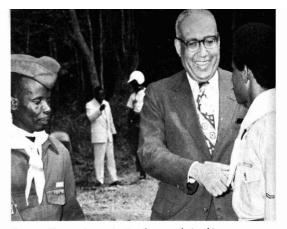
As far as environmental protection is concerned, federal legislation and this administration (in Washington, D.C.) have shown much care for environmental protection. For example, the Virgin Islands is now part of an air quality control region. We have switched fuel for electric power production; we now use low-sulfur oil. We check and monitor the emissions from the Hess (oil) and Harvey (aluminum) plants on St. Croix, and not only air emissions but heavy metals in water and so on. In addition, we have passed laws requiring very careful control of land clearance for building purposes to reduce erosion, sedimentation, and other forms of water pollution. Also, we have taken a very strong point of view on inshore dredging to protect bays and harbors.

Water treatment plants

Ocean disposal was an item of concern when the President's Water Pollution Control Advisory Board met here in 1970. What is the policy on waste water treatment?

As of this moment, there is still more discharge than we want because our treatment plants are not yet finished. The treatment plant in St. Croix is finished and in operation, but all of the interceptors and collectors are not completed. So, in that sense, there is still some untreated effluent going to the ocean. However, when these lines, which are under construction, are finished, St. Croix will be completely free of any ocean discharge of untreated sewage. Sometime within the next 12–18 months we will reach that goal.

Water treatment for St. Thomas is coming along, but they are a little bit behind for many reasons. Their plans were late in being developed; the work is much harder in the terrains (more rock) on St. Thomas, and congested areas also add to the problem. Probably within the next 18 months the entire matter of untreated sewage will be a thing of the past. The plant in St. Thomas is not yet in operation; it's near completion. Because of the small size of St. Thomas (32 sq miles), it's quite possible that they may finish the same time as St. Croix (84 sq miles) even though at the present time they are behind.



Governor Evans. Improving interhuman relationships

Actually many of the settlements or developments have been made to put in their own treatment plants. For example, St. Croix had a treatment plant at Estate Scion Farm; we have one at Camporico; hotels have put in their own. On St. Thomas, there is a treatment plant in the developing area of Tutu. The master plan calls for them all to tie into the sewer system eventually.

In an effort to conserve water, we are engaging in a pilot study of ground recharge where we will subject our waste water to tertiary treatment and then pump it underground for reuse later. We have started a pilot program in that regard and are in the process of doing the recharge.

What are the statistics on plants? For example, how many are on line, how many are planned, how much did they cost?

We have two major treatment plants, one on St. Croix and one on St. Thomas. The consulting engineering firm of Camp Dresser and McKee (Boston, Mass.) did the major design of these plants. We also had some work with Black, Crowe and Eidsness (Wilmington, Del.). The federal share of these plants was 55%, the local government share, 45%.

When was the first treatment plant built?

Our first plant on St. Croix was finished this August. It's a 4-mgd plant but it is expandable. It could easily be expanded to 16 mgd. Originally, the entire system of two plants was estimated in the neighborhood of \$15 million. The bids have been coming in higher than estimated, and cost has been going up so I suspect the total cost to be close to \$20 million.

Dredging activities

Is there any environmental conflict between dredging activities under Department of the Interior permits and goals of the island government?

More apparent than real. The problem here is that many of the permits issued by Interior were issued before we knew as much about environmental control and ecology as we do now. Many of the permits were old. It was my recommendation that permits under a certain age be recalled and restudied. Some of the criteria used in deciding whether to grant permits four years ago have changed. Many of the permits granted four years ago would not be granted under the same conditions now.

Basically, there is no conflict. Interior sees the need for controlling; so do we. However, there was a problem in that under the federal law we did not have the outright jurisidiction to police dredging. The policing was left to Interior, and not being on the spot, they couldn't do a good job. We are working on arrangements where it is in our interest to police dredging, even in the absence of specific federal law. It's working out quite well, so we feel that this particular hurdle has been overcome.

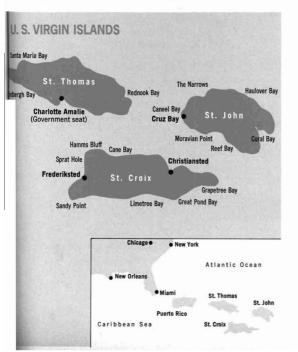
How often are permits reviewed and how many have been issued?

Most permits are not more than 4 or 5 years old. They usually have expiration dates on them. We would like to see all permits called in for review and resubmission every two years. We don't know if the recall procedure is feasible because in some cases it involves a tremendous expenditure of money.

I haven't the faintest idea on the number of dredging permits. As a matter of fact, some of them we never know about. In the beginning, the permits were handled through Interior. After I took office (July 1969) we set up a procedure whereby no permit would be granted by Interior unless it came through the Governor of the Virgin Islands with recommendations. So in the last two years, we have been able to know what is going on and can control it. We have no record of permits issued before then. Until we see somebody doing something and check on it, we never know about that permit. Of course, this type of permit is getting more infrequent.

Can you elaborate on the working relationship of the island government and the Interior Office of Territorial Affairs:

I think it's a very good relationship. The Interior deputy assistant secretary for territorial affairs is the Hon. Stanley Carpenter. The minute-by-minute, day-by-day control that existed at one time no longer does. With the election of the Governor here (in the election of November 1970), the Governor is no longer an employee of the Department of the Interior. The Department of the Interior has, in keeping with the Congress and others in general, attempted to give the government of the Virgin Islands more autonomy in running its own affairs. They still keep a watch on several things. For example, they maintain an office of controller to check on fiscal management; they also retain control over the matching funds for federal construction grants.



Economic development

What industries are being attracted to the area?

A few months ago, I signed the Investment Incentive Act into law, and we are now trying to attract industries of many types. Our requirements, of course, are for industries that do not pollute or disturb the environment. Unless these industries produce their own electricity and water, then they would be industries that do not make large demands for water and electricity because these items are commodities which are not present in large supply. We also want industries that tend to be permanent, not just ones that come here for the duration of the tax benefits. Specifically, we are trying to attract light chemicals, pharmaceuticals, certain types of paper industry. We would also like to expand some industries that we already have, such as watch assembly and textiles, industries which would require the employment of small numbers of relatively skilled workers. We feel that we have a work force here which, even if it isn't trained in a particular area, has the potential of being trained because we have compulsory education, a very literate population and a lot going for the islands.

Is the island government tied up in federal red tape or can it operate autonomously, or what?

In some cases the territorial government is tied up in federal red tape. When we are dealing with many of the federal grants we get involved in federal red tape. Many of the programs would be much further along had they not been slowed up because of the needs of getting approval from the various federal government agencies. We have about the same autonomy that a state or territory has in terms of federal grants. We do not have that much authority as far as laws are concerned because the Congress passes laws specifically for the Virgin Islands, something it does not do for the states.

New environmental offices

How do the various environmental offices of the island government fit together to make an environmental team? What is the responsibility of each—the Department of Conservation and Cultural Affairs, the Division of Environmental Sanitation, and the Caribbean Research Institute?

The Department of Conservation and Cultural Affairs has a very broad spectrum of responsibility. It's responsible for recreational facilities, cultural development such as libraries, protection of the environment such as dredging and land clearance; it is also responsible for the development of parks, other green areas, and preservation of historical monuments.

The Division of Environmental Sanitation touches some of the above but it is involved with checking of eating and drinking establishments for sanitary purposes, the eradication of mosquito breeding places, checking milk and water supplies, including drinking water. The two groups work well together but, of course, they have different spheres. On the other hand, when it comes to protection of beaches, the problems of thermal pollution, water pollution, and bacteria pollution come to the fore. So, on these issues, the two offices do overlap a bit.

The Caribbean Research Institute offers the technical and scientific expertise that we need to attack and find solutions to certain problems that can be done on the islands. Why, for example, has the spiney lobster, which was once plentiful and a mainstay of our diet, become very scarce? Is it due to excessive and careless fishing and handling? Thus, as one example of its activities, the Institute is looking at what can be done to bring these lobsters back.

outlook

Water pollution law — 1972 style

Congress commits the nation to continuing water cleanup; the public must adopt a wait-and-see attitude

After being vetoed by the President, the Federal Water Pollution Control Act Amendments of 1972 were passed into law by an overwhelming majority in both houses of Congress on the very last day of the 92nd session. It's a farreaching law, and the most significant piece of environmental water legislation ever to come out of the U.S. Congress.

Without question, the new law is tough, complicated, and costly. The big question now is whether the financial backing for cleanup will be forthcoming. In his letter to Congress, President Nixon made the point that "despite its laudable intent," he was vetoing the bill because of "its unconscionable cost." But without the new law the U.S. machinery for clean water would have come to an abrupt and screeching halt. Now, it's the only blueprint that the U.S. has for water cleanup, one for which the public has been waiting since the 92nd session of Congress convened. Bottled up in conference since May, P.L. 92-500 extends the Federal Water Pollution Control Act of 1965, as amended in 1966 and 1970. (The 1970 amendments are known as the Water Quality Improvement Act.)

The U.S. now is wed to a new cleanup schedule. Whether the marriage will be successful, only time, money, and commitment can assure. There is something

Minicalendar for standards watchers, P.L. 92-500

Water quality standards & implementation	 Must submit plans for any intrastate water quality standard Must determine that wqs are consistent with new act, if not notify states Must adopt, submit intrastate wqs Must approve or reject state intrastate wqs plan Must promulgate or substitute plan Must have held at least one public hearing on wqs by Oct. 18, 1975 	
National standards of performance	 Must publish degree of effluent reduction attainable by secondary treatment Must publish list of industries for which new source standards apply Must publish informa - tion on alternative waste treatment management Must propose and publish standards Must publish non-point source guidelines Must promulgate standards for new industrial sources 	
Toxic effluent standards	 Must publish list for which effluent standards are to be set Must propose effluent standards Must promulgate standard with annual updates 	
Pretreatment standards	Must publish proposed regulations for introduction of pollutants to works Must promulgate standard Achievement of the pretreatment standard by Jul. 18, 1976	State of the state
	EPA administrator States A Industries This calendar is not to be interpreted as a complete list of legal requirements for the EPA administrator Convright 1972. The American Chemical Society	La LE T N LE LE

old, something new, something borrowed, and something blue in the new law—old in the sense of continuation of the 1965 law; new with regard to the state permit program and the National Study Commission; borrowed (from previous federal environmental legislation) with regard to the numerous deadlines; and blue since it was passed over the President's veto.

It's not too surprising that the new law contains deadlines, nor that it is modeled closely after the clean air bill, since the water law was given to us basically by the same legislators who gave us the 1970 amendments to the Clean Air Act (Es&T, February 1971, p 106).

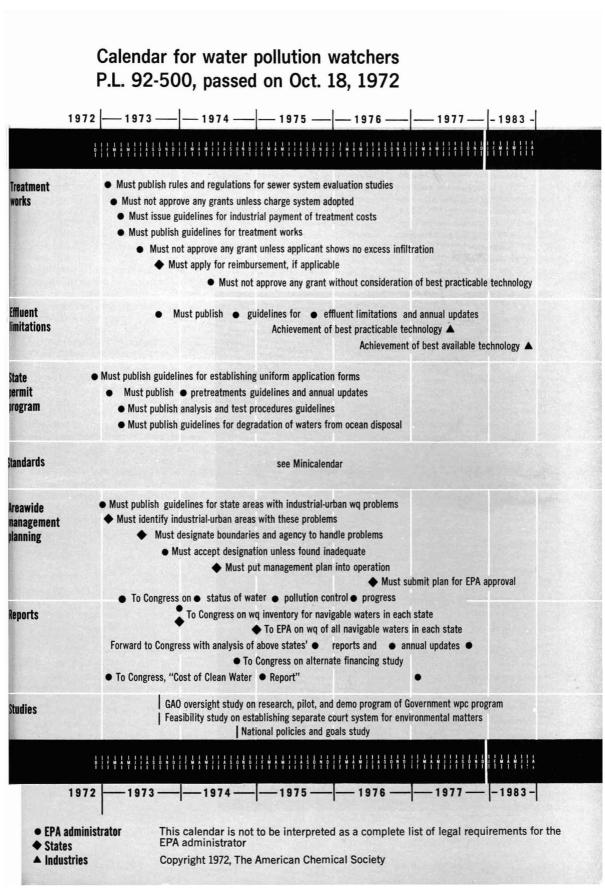
Biggest expenditure

To be sure, the largest authorization goes for publicly owned waste treatment works, a whopping \$18 billion over the next three years—three times more than the President had requested for the same time period. What is more, there are \$2.75 billion authorized for reimbursements for treatment works already constructed.

In the new law, the definition of "treatment works" has been expanded to include interceptor sewers, sewage collection systems, pumping, power, and other equipment, as well as land site acquisition for either the plant or the site for ultimate disposal of sludges.

The good news for the recipient of "treatment works" grants is the fact that the federal share toward construction funds is now 75%. "Construction" means everything from preliminary planning to actual construction. Another piece of good news is that this share can be paid for treatment works from funds authorized for any federal fiscal year beginning after June 30, 1971.

On the other hand, the cautious hopefully not bad—news is that certain limitations are spelled out. For example, after June 30, 1974, the EFA administrator cannot make such "treatment works" grants unless the applicant has studied alternative waste management techniques and taken into account the reclaiming and recycling, or otherwise discharging of pollutants. These so-



called areawide management plans would spell out the needs of the area for a 20year period, with annual updates.

Keyword

Although terms like effluent limitations do not carry the label "standards" when published within a year after enactment, they will put existing industries on notice, so to speak, that certain technology is being used by some leaders in their industries. The notice, in effect, says that the industries must have "best practicable technology" by July 1, 1977, and "best available technology" by July 1, 1983, with the goal being no discharge of pollutants by 1985.

Standards are the keyword in the new water law, just as they were in the now

2-year-old air law (Es&T, February 1971, p 106); national standards of performance; toxic effluents standards; pretreatment standards; as well as water quality standards and implementation plans for their achievement in intrastate waters.

Of course, the public surely knows by now that standards alone have no effect whatsoever on the achievement of water quality. Only when standards and control technologies are applied will water quality be enhanced.

Under the earlier provisions of the water amendments, states have had most responsibility for water pollution control, but now they have an additional authority to establish a state permit program (under federal guidelines, of

CONSTRUCTION GRANTS FOR TREATM	ENT WOR	KS Subtoto	1 \$21,350	
Treatment works construction	\$5000	\$6000	\$7000	
Reimbursements		2750	>	
Basin planning		200		
Areawide waste treatment management	50	100	150	
Technical service to agencies	50	50	No.	
RESEARCH AND RELATED PROGRAMS		Subi	total \$608	
Research, investigations, training,				
and information	100	100	••	
R&D grants	75	75	••	
State pollution control grants	60	75		
Mine water demonstration program				
Training grants and contracts	25	25	10.00	
Great Lakes cleanup demo project	-	25		
In-place toxic pollutants removal contracts	1.19	15		
Alaska village demo project	-			
Lake Tahoe study	4	0.4	5	
STANDARDS AND ENFORCEMENT	al and a second		al \$1,451	
Standards' development	100	100		
Clean lakes grants to states	50	100	150	
Revolving oil fund		35		
National Study Commission		15		
Alternate financing methods study	+	1_		
Other than the above sections	250	300	350	
ADMINISTRATION AND STUDIES		Subtotal \$10		
Environmental financing funding		100		
National policy and goals study		5		

course, to ensure some uniformity). The state permit is for the construction and operation of any activity that may result in any discharge into the navigable waters. (Basically, it is a plan similar in concept to that the Corps of Engineers announced two years ago, just prior to the establishment of the EPA.) The permit also applies to effluent limitations, national standards of performance, as well as toxic and pretreatment effluent standards. (See Minicalendar.)

How reasonable?

While it is true that the deadline for "best available technology" for point sources is 1983, the National Study Commission, another new creation of this law, must report (within three years) to the Congress specifically on all the social, economic, and environmental effects of achieving or not achieving the effluent limitations specified for 1983. The 15-member commission—comprising senators, congressmen, and members of the public—can contract for studies with the National Academy of Sciences and the National Institute of Ecology, among others.

For the scientist, there will be created an advisory board selected from the scientific community for its expertise on scientific and technical information. The board will advise the EPA administrator on effluent limitations, water quality information and other matters. Eight members are to be appointed by the administrator for a 4-year term and can be reappointed.

Without question, none of the plans is any good without enforcement. Again, modeled after the air legislation, penalties for violation of the new water law are tougher under P.L. 92-500 than under earlier water laws. Conviction for a knowing violation is subject to a penalty of \$25,000 per day or imprisonment for one year, or both. For a second knowing violation, the penalty increases to \$50,-000 per day of violation, imprisonment for two years, or both. What is more, any person who violates a provision of the state permit in regard to standards is subject to a civil penalty of \$10,000.

The public's role in water pollution control, which stems from a then unique feature in the air law, has been carried over into the water field: Citizen action suits can be brought against polluters and government officials, but such suits cannot be brought against the EPA administrator in cases where he is given discretion to act.

Computer model simulates air pollution over roads

Sophisticated modeling technique which predicts pollutant movement accurately will aid in design of new freeways

Freeway designers may soon be able to design roadways which minimize the effects of pollution from automobile emissions with the help of a new computer modeling technique called EXPLOR.

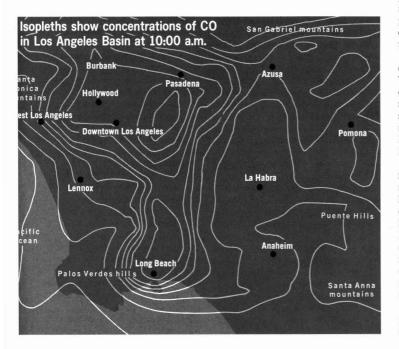
EXPLOR (Examination of Pollution Levels of Roadways) is an offshoot of another computer modeling technique called NEXUS (Numerical Examination of Urban Smog) developed under contract with the Environmental Protection Agency by Systems, Science and Software, Inc. (S³), La Jolla, Calif.

What sets NEXUS and EXPLOR apart from most other air pollution modeling exercises is a greater degree of sophistication, according to Lynn Teuscher, group leader of S³'s Air Quality Section. The computer simulation techniques take into account topography surrounding the road bed, weather conditions, and other variables that could be expected to influence levels of pollution.

Regional air quality monitoring requires accurate numerical simulation of the effects of advection (horizontal air movement) and eddy dispersion processes on pollutants emitted to the air both from point sources and mobile sources, Teuscher explains. The techniques developed for NEXUS use the computer to track the movement of discrete parcels of polluted air through a region divided into grid cells.

The application of NEXUS techniques to EXPLOR similarly allows the tracking of small volumes of air over roadways. The computer programs have been adjusted to provide information for the specific pollutants—carbon monoxide, unburned hydrocarbons, nitrogen oxide, nitrogen dioxide, and particulate lead which may be directly attributed to automobile emissions.

EXPLOR was developed specifically to predict pollutant concentrations in a milewide corridor traversed by a roadway. The air overlying a roadway is divided into cells, and the particles of pollutants in the air are tracked from one cell to the next. The number of cells varies with the configuration of the roadbed—whether it is elevated, depressed, or at grade (level with the



surrounding terrain)—but a typical grid network might consist of approximately 10 cells overlying the width of the highway, 30 cells in a vertical direction extending up to about 300 ft, and cells downwind and upwind as far along the roadbed as one wishes to track.

Actual amounts of emitted pollutants, topography of the surrounding terrain, and corrections for temperature and wind-shear forces are taken into account. EXPLOR uses a Univac 1108 computer with the large configuration to predict where the pollutants will go, where they will concentrate and in what amounts, and how long they are likely to remain.

Teuscher emphasizes that the EXPLOR program does not use Gaussian distribution models which assume constant wind and diffusion, bounded in a region limited by the land surface and a possible inversion layer. Instead, EXPLOR is a numerical integration model which takes the basic differential equations of diffusion and solves them from grid to grid.

EXPLOR can be used to predict:

• what type of land development is most acceptable near freeways

• the difference in pollution levels generated by stop-and-go driving vs. constant high-speed driving

• how pollutant levels would differ with freeway design at grade, below, or aboveground.

Data from the computer are displayed as dot pictures of the parcels of polluted air. Alternatively, the output may be obtained in the form of computer-contoured isopleths (sets of curves, each at a constant pollutant concentration).

NEXUS was developed to study photochemical smog in the Los Angeles Basin. The EXPLOR computer model was validated by use of data obtained from a multifaceted study of a 42-mile loop of superhighways in the Los Angeles area, made up of the Harbor Freeway, San Diego Freeway and the Santa Monica Freeway.

The major objective of the loop study, funded by the Federal Highway Commission, was to help solve problems related to traffic flow, according to Teuscher. In conjunction with the study, however, the California Department of Highways made some air pollution measurements. Validation of the EXPLOR model was done with data measured for carbon monoxide. "We have very good results," Teuscher said. "The correlation coefficient is greater than 0.9."

A coefficient of 1.0 would have shown perfect correlation between the model and the actual data, he said.

Putting the closed loop into practice

Dow Chemical Co.'s latex plant in Dalton, Ga., was built with complete water reuse in mind; for the past five years it has completely recycled its process water

Waste water discharges are becoming more and more limited by federal law (see story on the new water bill, p 1068), and increasing emphasis is being placed on a closed-loop system of water recycling both to conserve water and to prevent pollution. Few manufacturing plants can yet boast of complete water reuse. However, a Dow Chemical Co. plant in Dalton, Ga., is successfully operating such a system.

The Dow plant manufactures styrenebutadiene latex for carpet backing and carpet construction and latex foam for carpet backing. The plant is ideally located, since at least 240 carpet manufacturers producing tufted carpeting (only 5% of the market accounts for woven carpet) are located in the Dalton, Ga., area. In fact, Dalton is known as "the carpet capital of the world."

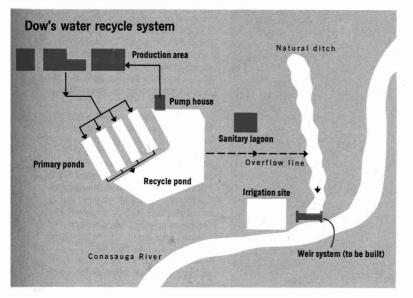
Carpet manufacturing itself explains the need for latex. Carpetmaking ma-

chines with up to 12 needles per inch continuously punch loops (or tufts) of yarn into a jute or synthetic scrim (backing). Since there is no "lock" stitch, any tufted carpet could be unraveled by pulling one of the yarn loops. Latex applied to the back of the scrim locks the tuft. When water evaporates from latex—an industrial adhesive—a synthetic rubber film is formed that keeps the carpet from unraveling. Latex also affixes another scrim to the carpet as secondary backing.

The Dow plant

In view of the importance of latex to the carpet industry, Dow officials decided to construct a latex plant in Dalton rather than ship large amounts of latex to its carpet customers. The plant site was chosen in 1965, explains Jim Kaye, plant manager at Dalton.

In the early planning stages, Dow



1072 Environmental Science & Technology

officials considered an intake water station at the Conasauga River which flows along one side of Dow's 420 acres of property. However, the river water would need to be treated and cleaned to remove clay silt before use in the plant. Also, effluent discharged back into the river would have to be treated in this once-through system.

Water from the municipal system was available, but would have been too expensive to buy for once-through use. Also, the tremendous volume of water needed for latex production could seriously affect local supplies in times of drought.

Dow officials then decided, with the approval of the Georgia Water Quality Control Board, to use municipal water for makeup and sanitary water, and to provide a closed-loop system for reusing process, wash, and coolant waters. The goal was to eliminate discharge to the river and to benefit economically by recycling water. The plant was then built in 1966 and began operation in 1967.

Latex produced at the Dow plant is a water suspension of very minute synthetic rubber particles formed by a proprietary process for emulsion polymerization of styrene and butadiene. An initiator starts the reaction in a water phase, with soaps added to emulsify the oil-water interface. The latex particles produced are very tiny (1500-2500Å) but with a tremendous surface area per unit volume. The final latex product is approximately at a 50% solids level.

The large surface area of the latex particles accounts for the fact that only trace quantities of them will give water a cloudy appearance.

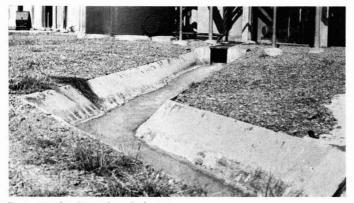
Besides visual pollution, effluent from the manufacture of latex contains bits of latex, trace quantities of unreacted monomers, and ammonia. The effluent has a high chemical oxygen demand (COD), but is "essentially nil in BOD," emphasizes plant manager Kaye. Latex particles are chemically stable and will remain in suspension. Coagulation and removal are essential for COD reduction as well as for appearance.

Large amounts of wash water are used in the Dow plant. Latex is easy to wash away while it's still wet, but after it dries, it forms a skin that is difficult to remove. Thus, equipment, tank cars, and tank trucks are repeatedly washed to prevent buildup of dried layers of latex.

Closed loop

Here's what Dow does with its effluent: About a million gallons of water are used daily for processing, cooling, and washing. All drips, spills, wash water, and process water end up in the outflow stream from the plant. The plant's effluent stream flows through a series of concrete trenches leading to the primary ponds. Metered amounts of alum (aluminum sulfate) are added to the waste stream to destabilize latex and to coagulate it. The coagulating effluent is then neutralized by either sulfuric acid or caustic soda to maintain a narrow pH range, preferrably around 7. Optimum coagulation occurs below pH 8. Also, neutrality protects the process and fire system piping when the water is later reused. Alum, and acid or caustic when needed, are added in the trench to promote mixing by natural turbulence.

The trench system leads to four primary ponds. Each one and one-half



Reuse. Another latex plant discharges wastes directly into a stream (below); however, Dow's latex effluent is treated in a trough (above) and pond system and then is ready for recycling (above left)



million gallon pond is used alternately. Any three can be shut off while the remaining one is filling with effluent. Under optimum conditions, floccing occurs in the pond within 2–3 hr; however, each primary pond of effluent usually undergoes a settling period of one to one and one-half days. About a million gallons of water daily flow to the primary ponds for settling; each pond is thus filled once every four to six days.

After settling, the clarified water drains by gravity into the $10^{1/2}$ million gallon "recycle" pond. On the edge of this large pond is a pump house where vertical turbine pumps recycle the clean water continuously back to the plant through buried pipelines. Also, two 2500 gpm fire pumps, one diesel and one electric, feed the sprinkler, deluge, and hydrant system. The recycle reservoir serves as a fire protection reserve since both major raw materials (styrene and butadiene) are very flammable, and the plant is located in a rural area. "We keep a minimum of four million gallons in the pond," says Kaye, "for fire protection purposes."

The Dow plant also has two other water systems. In one, a cooling tower recirculates two million gallons of cooling water each day. In the other, sanitary wastes from the plant are treated in a state-approved biological oxidation pond. In periods of flood or heavy rains, the sanitary pond does sometimes overflow; however, the capacity of this lagoon is such that the discharge is of acceptable quality.

Overflow

In past years during the rainy winter months, some overflow from the recycle pond flowed through a drainage ditch which eventually led to the Conasauga River. Last year, Dow installed a system that it hopes will prevent any overflow from entering the river.

Land around the plant and pond system was graded so that rainwater would not run into the ponds. Rain runoff, in the winter months, has in the past contributed to filling the ponds to capacity and causing overflow. To boost capacity, the recycle pond was dredged from its original nine million gallon limit to its present 10¹/₂ million gallons.

An irrigation system was set up to handle surpluses and permit pond-level management in anticipation of the wet winter season. Underground pipes carry water to an adjacent area for spray irrigation. The high nitrogen content of the recycled water (400–500 ppm) contributes to vegetative growth, and this system avoids direct discharge to the river.

A single, natural low-line leading to the river takes any outflow from the active plant site, the ponding systems, and rainfall runoff from the irrigation area. Dow also plans to install a weir dam where this drainage line meets the river. A V-notch weir will measure the flow of water. Water samples taken at the dam site will be regularly tested for quality. "The real test will come this winter during the rainy season," says Kaye, but he is confident that "we'll have no discharge all the way around."

The ponds are monitored at least once per shift (the plant operates 24 hr/day, 365 days/year) and up to five or six times per shift. The primary ponds are monitored for pH and clarity, and a log is kept to record whether each pond is filling, settling, draining, or empty.

Last year, two of the primary ponds were dredged to remove the accumulation of sludge which had reduced pond capacities to about 750,000 gallons. The latex sludge presents some problems in landfilling, largely connected with drying. CKL The Interstate Commission on the Potomac River Basin, hampered by a lack of enforcement authority, tries to help formulate the master plan for . . .

Cleaning up the Nation's River

According to usually reliable sources in the Nation's Capitol, George Washington once threw a silver dollar across the Potomac River. Today, there are places where the Potomac is so thick, he might as easily have walked across with it.

Silt, agricultural runoff, sewage, acid mine drainage, and inadequately treated industrial effluents continue to pollute the Potomac.

Yet some progress is being made. Parts of the Potomac are relatively clean. There is hope, at least, that beefed-up federal laws and new agreements between the jurisdictions that make up the Interstate Commission on the Potomac River Basin (ICPRB) will provide the muscle needed to clean up the river.

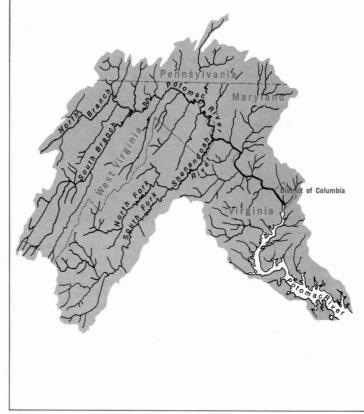
The ICPRB was established by an act of Congress in 1940 with passage of the Potomac River Sanitation Compact and charged in a general way with helping to abate pollution along the Potomac. Membership in the ICPRB is made up of delegations from Virginia, West Virginia Pennsylvania, Maryland, the District of Columbia, and the federal government (see box, p 1075).

The depth of the commitment to pollution abatement during the 1940's and 50's is perhaps best judged by the fact that during those years the ICPRB had no full-time staff. The sole employee of the commission was a part-time executive director.

In fact, the Commission did not have a full-time executive director until 1967 when Carl J. Johnson, a former Director of Conservation with the state of West Virginia, was named to the post. Johnson held the job until his death early this year and is generally credited with greatly enhancing the effectiveness of the ICPRB.

The present Executive Director, Paul Eastman, was named to his post in July 1972.

In the early days of the ICPRB, virtually all of the Commission's efforts were directed toward generation of public support for pollution abatement. Even today, much of the ICPRB's activity Drainage area served by Interstate Commission on the Potomac River Basin



centers around grass-roots education and the marshalling of public support for basinwide programs the Commission sees as necessary to carry out its mandate.

Scope of compact

The Commission in its present form began to take shape in 1970 following a 10-year tug-of-war among the signatory powers which came to an end late in 1970 when the federal government inked an amended Potomac River Basin Compact.

The new Compact widened the sphere of influence of the ICPRB to include water quality and supply management and land use planning for the basin as a whole. Prior to the new compact, the Commission's authority had been limited solely to questions of water pollution.

Even newer proposed amendments to the Compact—formulated by a Potomac

River Basin Advisory Committee would once again broaden the scope of ICPRB's activities to include enforcement powers and financing authority. The proposed amendments have been ratified by two states—Virginia and Maryland—and require signatures from each of the remaining four jurisdictional bodies before they become effective.

Under P.L. 91-407, approved by the Congress in September 1970, several broad powers are given to the ICPRB. Briefly, they include instructions:

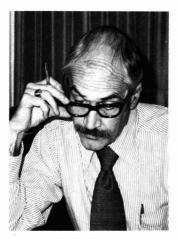
• to collect, analyze, interpret, and otherwise prepare data bearing on the problems of pollution or water quality in the Potomac Valley Conservancy District—the area designated by Congress to be under ICPRB's jurisdiction

 to cooperate with state agencies and other interested groups or persons in promoting uniform laws for abatement of pollution or development of water resources

• to conduct public information and education campaigns, provide liaison with agencies dealing with pollution and related water problems in the Potomac River Basin

• to review or comment on proposed plans by any agency within the Conservancy District relating to water pollution or related water quality and supply matters.

Among the goals of the ICPRB are full compliance with currently approved water quality standards, in accordance with the implementation plans of the signatory agencies; a Basinwide program



ICPRB's Eastman reasonably optimistic

for water management (to be prepared by the end of 1973) which would assure adequate water supply through the year 2000; and development of comprehensive land use plans by the end of 1974 for areas adjacent to the major streams draining the basin.

Lacks enforcement

ICPRB, unlike several other river basin commissions of which the Ohio River Valley Sanitation Commission (ORSAN-CO) is more or less typical, does not have even limited enforcement authority ("ORSANCO: Pioneer with a new mission," ES&T, January 1971, p 22) according to Eastman. Under the new amendments to the compact yet to be ratified,

Commissioners	
District of Columbia Lee F. Dante Paul V. Freese Malcolm Hope Virginia James J. Corbalis, Jr. John A. K. Donovan Maryland Oscar W. Meier Marvin Mandel A. T. Brust, Jr.	U.S. government Lawrence M. Fisher Eugene T. Jensen Arnold Sternberg Pennsylvania Maurice K. Goddard West Virginia N. H. Dyer Cliff Umscheid William A. Powers
Officers	
A. T. Brust, Jr., Chairman Maurice Goddard, Vice Chairman Lee F. Dante, General Counsel	Oscar Meier, Treasurer Paul W. Eastman, Executive Director G. L. Carley, Jr., Asst. Executive Directo

ICPRB would finally have some power to enforce its recommendations and longrange plans.

The new amendments to the compact would permit ICPRB to hold hearings, issue subpoenas and compel testimony under oath. ICPRB could sue and be sued in its own name and could impose penal sanctions up to \$500 for "petty" offenses.

Perhaps as important as the power to set and enforce regulations would be the power granted under the new amendments to enter into financing agreements for projects in the Potomac Basin Conservancy District. The Commission could negotiate for loans, grants, or other types of revenues to carry out any of the projects necessary for pollution abatement or water resource management. A special article in the new Compact would establish procedures for issuing bonds or other instruments of long-term financing.

The Commission could condemn property through eminent domain and could establish the amounts payable by each member government for projects to upgrade water quality in the basin.

The new amendments to the compact would place responsibility for the affairs of the ICPRB closer to the top of state governmental affairs. The new compact would make the Governors of Virginia, West Virginia, Maryland, and Pennsylvania, and a presidential appointee representing the federal government voting Commission members.

Eastman feels reasonably optimistic that the states which have not yet ratified the amendments to the compact will do so in the future, perhaps even in the coming year.

He is not so optimistic about the recently enacted water legislation however. "The widely advertised promise of over \$24 billion of federal funds and uniform nationwide federal controls to eliminate all pollutants by 1983 will bear scrutiny and perhaps some skepticism by the Potomac Basin states as well as other areas of the nation," Eastman comments.

Eastman terms the new legislation "complex beyond belief," and questions the wisdom of massive federal control over state and local pollution abatement efforts. "Geographical and other differences between the upper Potomac mountain creeks and the giant Mississippi in the lowlands of the south have been eliminated by federal statute," Eastman charges. And he sums up his misgivings by quoting T. S. Eliot: "Between the idea and reality, between the motion and the act, falls the shadow." HMM

U.K. environmental boss carries clout

British minister has what it takes to make a real impression on pollution problemssweeping powers and lots of the green stuff

On a recent visit to Washington, D.C., Peter Walker, Great Britain's Minister of the Environment, paid some courtesy calls to his opposite numbers in the U.S. So pervasive are the responsibilities of Walker's 2-year-old department that he visited not only William Ruckelshaus at the U.S. Environmental Protection Agency, but John Volpe at the Department of Transportation, George Romney at the Department of Housing and Urban Development, and Christian Herter, Jr., at the State Department

Walker's responsibilities as "Secretary of State for the Environment" (his official title) are more than just skindeep. When the department was formed in 1971, it absorbed the functions of the previous Ministries of Housing and Local Government, Transport, and Works. Walker hints, with perhaps just a touch of complacency, that U.S. environmental protection officials are a mite jealous of his powers. And well they might be. Whereas the EPA administrator, for all his very considerable powers of persuasion and purse-string handling, has no real power over environmentally pertinent activities at the local level, Walker, in contrast, controls local government in Britain. He also controls the siting and construction of highways and other major land-use projects (something that Ruckelshaus would probably dearly love to be able to do).

Some of Walker's views emerged during a short press conference arranged by the British Embassy in the course of his Washington visit. Chief among them is a thinly veiled skepticism over the ways in which the U.S. has sought to legislate an end to pollution. Attempts to "force" technology along, such as are embodied in the most recent versions of the Clean Air Act and Federal Water Pollution Control Act (this issue, p 1068), are regarded by Walker as

ards should be based," he explains. Walker has the power under the British Clean Air and Alkali Acts to direct all companies within a given industry

well-meaning

the standards

to install similar cleanup technology, but he will not do so, he says, unless he is convinced that the technology will work. He would not, for instance, require the British auto industry to make cars (for home consumption) subject to the very strict emission requirements of the U.S. Clean Air Act, because "No one knows how to meet them." Nevertheless, "We are very glad that you in this country have taken the lead!"

All in all, Walker takes a rather amused view of the haste with which the U.S. has sought to clean up. He wonders in fact, if the resulting legal snarls aren't just making the U.S. "a land fit for lawyers to live in." The reason for Britain's calmer view of the situation, he suggests, is that "In the nineteenth century, we were so much dirtier than you were." Consequently, he goes on, Britain was forced to recognize and start working on problems long before the U.S. got around to them.

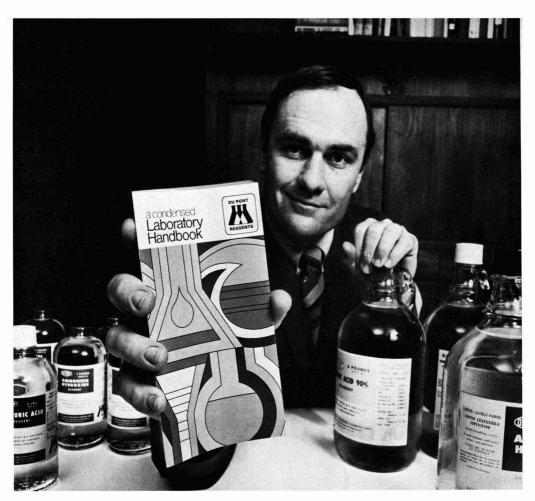
Walker refuses to acknowledge that there might be any antipathy between British industry and the Department of the Environment, or indeed between his department and other branches of the British government (such as the Department of Trade and Industry) that might be expected to be hostile to the economy-depressing aspects of a full-blooded drive for environmental protection. Government and industry, he says, are partners in cooperation,



something which U.S.-based observers find difficult to believe, but which may nevertheless be somewhere near the truth in Britain. Nevertheless. Walker's low-keyed description of the great strides Britain has made in cleaning up pollution left some reporters unconvinced, steeped as they are in the publicly displayed hostilities in the U.S.

Paradoxically, perhaps, Walker is a member of Britain's ruling Conservative (or Tory) party, which has traditionally taken an anti-big government view. He is, though, like all British cabinet ministers, an elected member of parliament; his constituents live in the well-heeled English city of Worcester. And the commitment of the British electorate to a reversal in the environmental trend of things is obviously as strong as that of the U.S. population. Furthermore, Walker has been pegged by the British press as an ambitious politician. (As if to support that assessment, it has coincidentally just been announced that Walker will move up to head the ministry of trade and economic affairs!)

Besides Walker's considerable powers, sufficient in fact for him singlehandedly to prevent the construction of a new shopping center in the Northern England city of Sheffield and to block the expansion of Leeds airport, his annual budget is enough to make U.S. environmental officials drool with envy: this year it's over \$9 billion; EPA's is \$3 billion. DHMR



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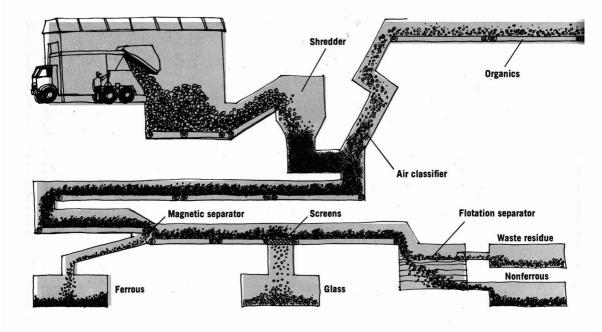
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Recovery of resources from mixed municipal solid waste



Tackling resource recovery

Richard L. Lesher

National Center for Resource Recovery, Inc. Washington, D.C. 20036

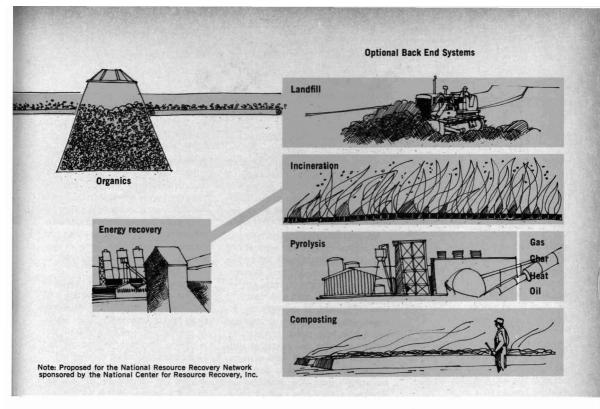
Solid waste recycling has in the past been too costly to compete with the dump, but technology may in the future provide solutions at a profit

ontinually spiraling costs of municipal solid waste collection and disposal combined with the rapid depletion of vital natural raw materials have spurred increased interest in resource recovery. One organization which has accepted the challenge of solving this growing problem is the National Center for Resource Recovery, Inc. The Washington-based nonprofit, nonlobbying corporation is unique both in concept and mission. The National Center was founded jointly by industry and labor in October 1970 with the primary purpose of seeking man-made solutions to man-made solid waste disposal problems.

The first major legislation to deal directly with the nation's solid waste problem was the Solid Waste Disposal Act passed in 1965. This act provided for a national research, development, and demonstration base for better waste management. In 1970, after extensive congressional hearings, this initial law was amended to become the Resource Recovery Act. Since 1970, federal attention has been focused on recovery as an alternative, in part or in total, to traditional refuse disposal techniques. While there has been substantial activity, little progress has been made toward actual installation of full-size recovery facilities.

In fact, recovery, rather than associated with mechanical separation or refuse processing in any form, is thought of as voluntary separation in the home, separate pickup of garbage and trash, can redemption centers, Boy Scout paper drives, etc. Attempts to develop a resource recovery industry have been beset with technical and, more often, economic difficulties. Resource recovery has simply been too costly to compete with the dump.

Technology has been applied so sparingly to resource recovery that the potential return from efforts of scientists and engineers is great. In a free enterprise system such as the U.S. enjoys, there must be open and total exchange of information and experience within industry. This exchange,



on a national scale

feature

accompanied by a total systems development approach, appears to offer the best prospect for achieving solutions to U.S. solid waste management problems.

The systems approach embraces many areas of interest: consideration of reclamation and reuse of products, changes in distribution and marketing patterns and procedures, consumer education, and other areas. But to be truly effective, it must define the manner in which the present waste collection and disposal systems function within the total environment, isolate weaknesses and develop corrections for them, and propose systems compatible with the physical requirements of the American life style.

Since this broad systems approach was an undertaking too vast to be assumed by any single agency, company, or industry, 12 founding industries (since joined by three other industries) together with founding labor unions created the National Center for Resource Recovery, Inc. Industries sponsoring and supporting the National Center represent aluminum, appliance, automotive, brewing, distilled spirits, food processing, food retailing, glass, metal can, paper and paperboard, plastics, rubber, soft drink, and steel. Labor representation includes United Steelworkers of America, and the Glass Bottle Blowers Association unions.

The creation of the Center is, in itself, unique, with competing industries joined by American labor to form an organization to help solve this country's solid waste disposal problems problems that have plagued mankind for nearly 2000 years. Actually, the National Center is a catalyst, the focal point for American industry and labor to work with government at all levels and to find answers to the national problem of solid waste.

Basically, the Center has two divisions. One, Research, is headed by James G. Abert, who handles research, analysis, engineering, and demonstration programs. He is also primarily responsible for the formulation of the National Resource Recovery Network program designed to evaluate the technical and economic feasibility of resource recovery.

The other division is headed by C. Wade St. Clair, who is Director of Information. The National Center disseminates information through its public awareness and education programs with the main objective of providing the public with factual information about the complex subject of municipal solid waste.

The problem

Let's now break down solid waste management into three basic elements: collection and transportation, disposal, and resource recovery systems. Collection and transportation involves primarily an economic problem. Collection of municipal solid waste has, in recent years, become a major national industry, both in cost and size. Some 80% of a city's solid waste management costs are expended to collect and trans-

port municipal refuse. It ranks with education, streets and highways, and police and fire protection as one of the largest municipal expenditures.

The majority of these funds are allocated for labor. And, as pay scales rise and the volume of waste generated increases, total collection costs will climb proportionally.

In many instances, a significant amount of this expense can be traced directly to outmoded equipment and systems. This is unfortunate, as new technology and methods are available today to both reduce costs and increase efficiency of collection operations (ES&T, May 1972, p 412). For example, at an "intermediate disposal site" or transfer station, refuse from smaller collection trucks is transferred to larger vehicles for more efficient long-distance hauling. Mathematical models can be used to determine the most efficient and least expensive collection routing for a specific city.

New collection equipment and systems include compactor trucks that grind, crush, or chop refuse as it is loaded, increasing the vehicle's hauling capacity and reducing the number of trips required. Another compactor truck innovation incorporates a "mechanical arm" operated by the driver from the truck's cab, netting major savings in both personnel salaries and time.

One of the more promising recent advancements in waste collection has been the "vacuum pipeline." Pioneered in Sweden, this system has been installed in apartment buildings, hospitals, and amusement parks to whisk away trash and garbage to remote collection and transfer sites.

As with collection, so have disposal methods remained essentially the same over the years. Presently, a majority of U.S. communities still practice some form of open dumping-the depositing of waste in one selected location-that pollutes the air as well as surface and groundwater. Such dumps, though now illegal in many areas, are not only unsightly and odoriferous, they are a breeding ground for rodents and disease-carrying organisms. Ocean dumping, another long used disposal practice, has also been recognized as environmentally hazardous under most circumstances.

sites

Incineration, another step in the refuse disposal cycle, might seem at first to be the solution to many solid waste disposal problems. Without question, improved incinerator systems can reduce volume of municipal waste by some 80%; and, if appropriately designed, can convert the heat generated into salable heat, steam, or electricity, But construction and operating costs are high, especially in light of recent federal and state pollution control requirements-most large municipal incinerators cannot meet rigid air pollution standards.

Practical alternatives

The most practical alternative to

open dumping is sanitary landfilling,

where solid waste is compacted in layers

and covered with soil. Properly en-

gineered, constructed, and operated,

sanitary landfills not only eliminate

health hazards, but are easily converted

into useful land for recreational facil-

ities, community parks, and building

While sanitary landfilling and incineration can be employed effectively in municipal solid waste management programs, neither provides for the recovery-the third element of solid waste management-of the increasingly valuable materials so abundant in each

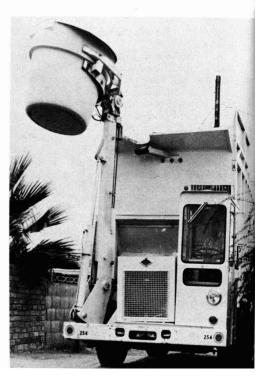
city's waste stream. The concept of resource recovery is not a new one. For centuries, scavengers have sorted through discards to recover items for reuse. The junk and salvage industry is performing an essential service in buying and selling scrap materials. More copper and zinc, for example, come from scrap efforts than from mining. For years, industry has been recycling waste products from the manufacturing process along with the materials bought from scrap dealers. Citizens groups collect materials for recycling.

A new concept in resource recovery is that of mechanical extraction and utilization of materials and energy from mixed municipal solid waste. In application, it offers a positive and promising approach to present and future municipal refuse management.

Not too many years ago, the U.S. enjoyed an abundance of essential natural resources. Such is not the case today, as testified to by our growing dependence on imported raw materials. These and other materials-iron, steel, aluminum, zinc, and glass, for example-can be recovered and returned to the "new product stream."

While these inorganics make up only some 15-20% of municipal waste,

Innovative. This experimental garbage truck, operated efficiently by one man, has saved Scottsdale, Ariz., \$200,000 annually





Resources. Technology is available to convert unsightly trash into useful products

they do represent the largest potential values for immediate recycling, which today are being transported—at no small cost—to dumps or landfills for burial.

Recovery and reuse of the remaining organic waste is equally promising. Paper and paper products have long been reused, to some degree. Other organics, such as food scraps, grass clippings, plastic containers, etc., have also been successfully converted into fuels and energy through high-temperature heating processes and biological decomposition. Composting the conversion of organic waste into a soil conditioner—has been practiced for centuries.

Resource recovery, therefore, is not new. Why, then, has it not had broader application? For one reason, not until recently has it been able to compete economically with conventional means of disposal. That, however, is changing with increased costs of landfill operations, incinerator construction and operation, and the increasing values of materials found in the waste stream. Also, recent emergence of new techniques and equipment is making resource recovery a more attractive alternative to basic disposal systems. For these reasons, the National Center is convinced that resource recovery from municipal solid waste is not only feasible, but that it can be developed into a major, new American industry.

Plan for action

Therefore, the National Center has developed a "plan for action," aimed at hastening the application of resource recovery from municipal waste by: conducting thorough and systematic studies of available operating systems, surveying the quantity and content of municipal refuse in representative U.S. cities, identifying possible markets for recovered materials, and analyzing the economic suitability of resource recovery, both from the standpoint of municipalities and private operators.

Favorable results from these efforts could lead to the development of a National Resource Recovery Network, conceived by the National Center as a program for demonstrating the feasibility of resource recovery in selected locations across the country. The Network is specifically designed to:

 Provide a demonstration program for displaying and evaluating new technology—or application of technology used in other industries—to recover resources from mixed municipal refuse

• Serve as a catalyst in the development of a new industry, consisting of self-sustaining resource recovery facilities for extracting raw materials and other economically valuable resources from what today are waste products

Technology is complicated, not so much in hardware or equipment, but in management systems; for example, organizations and organizational arrangements which must be included in the undertaking. Implementation of a technological concept progresses through three steps-innovation, development, and implementation. During the development step, the operational mechanisms which support the concept are selected. Installations made during this time are surrounded by a set of "special circumstances." The Network provides supportive framework for cooperative effort within which interested parties can, by playing the roles dictated by their proper self-interests, ensure the best possible conditions to develop this industry.

The Network, through its conditions

for membership, also provides uniformity and comparability required for objective evaluation to take place in order that the future of this industry will be based on a sound understanding of the economics of resource recovery. The Center's public awareness program will also provide the widest dissemination possible for information obtained from Network sites.

The National Center is, at the present time, going through a research and development stage to select the best opportunities from among the available options. The Network will begin with one site selected from four "planning" cities in about a year and build to a point where there are 12 sites with facilities for extracting all the economically valuable materials.

This multisite approach will provide a broader base than could be achieved with a single site. In terms of geography, markets, and public awareness, this base is needed as the foundation of the resource recovery industry. In addition, the Network is a time-phased program—lessons learned from early sites can be applied to those which follow.

City sites will be chosen according to strict criteria covering availability of local markets for recovered materials, alternative cost of disposal (pegged at \$6 a ton or higher), and daily availability of at least 500 tons of mixed municipal solid waste, approximately the size of a city with a 200,000 population. For back-up, the latter should represent approximately 50% of the available waste. In other words, Network sites will be part—not all—of an existing disposal system.

Two-phase effort

Each Network site or facility will be implemented into a two-phase effort, each divided by technology. During Phase I, or "front-end" separation, a facility will segregate collected refuse into light (organic) and heavy (inorganic) fractions. Work in the first phase, however, will concentrate on reducing waste mass and processing only the inorganic portion.

Each Network facility will accomplish this through a series of dry separation techniques. Incoming refuse will first be reduced by shredding and further processed by air classification (dividing

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the organic and inorganic fractions) before a selective process of separation begins. Iron and steel will be extracted by magnets. Density screening will remove glass, and a color-sort mechanism will additionally segregate the recovered glass material by color. Heavy media flotation devices will then recover aluminum and other nonferrous metals. Some paper will also be recovered by handpicking.

Phase I will employ off-the-shelf technology to reclaim the valuable inorganic materials. The remainder or the bulk of the light, combustible organic fraction, will be disposed of by incineration or landfill.

Phase II, or "back-end" recovery, will be later incorporated in each facility to include technological advances energy recovery, fiber reclamation, or production of compost—in recovering goods from the organic fraction. Other possibilities include direct reclamation of plastics and recovery of a greater amount of paper.

The National Center will provide, in the beginning, the planning, implementation, and management services necessary to initiate each recovery facility. While the Center will also contribute a certain amount of "seed money," each site will be owned by private or public concerns, such as refuse collectors, salvagers, refuse disposal operators, or secondary materials dealers.

In all cases, individual facilities will operate on an enterprise basis. The enterprise nature is essential, whether privately or publicly held, to evaluate goals of the demonstration. The basic requirement is that each site be a separate, independent cost center so that its performance can be measured.

The local owner/operator will have responsibility for initial development of the facility and its day-to-day operation. The role of the National Center will be supportive rather than managerial, ensuring that the owner/operator has the required technical information and that he applies it systematically to all Network elements. The Center's contribution will be concentrated in the technical and economic evaluation of operations.

There are several reasons why resource recovery should, at this time, be undertaken wherever feasible. First, "alternate costs" of solid waste disposal are increasing—a result of air and water pollution laws coupled with rising land costs. Second, the mood of the citizenry is changing toward ecology and conservation. In many locations, traditional methods are no longer acceptable.

One further reason why it can be said that the future of resource recovery looks promising is that the National Center has taken the systems approach to the solid waste management problem. By considering all facets of the problem, and working with governmental and private organizations, it has developed an approach to the solution through the concept of resource recovery. And, the Center has developed a plan for implementing this concept in a logical, comprehensive, and selfreinforcing manner.

The advantage of resource recovery in general and of the Network in particular is that it provides a positive and constructive answer to the problems of solid waste management. Further, it provides immediate evidence of working toward a solution which already has public acceptance, under the name of recycling. The alternative is restrictive legislation of unproved merit which is often passed in haste. The Network approach, therefore, strikes at the root of the problem, treating the disease and not just the symptoms.

This is the exciting challenge accepted by the National Center: to serve as a catalyst in furthering the development of a new industry...an industry which, in the words of President Nixon, "... treats many types of solid wastes not as pollutants but as recoverable and reusable 'resources out of place.'"



Richard L. Lesher is president and chief executive officer of the National Center for Resource Recovery, Inc. With his education in business and economics, Dr. Lesher, prior to his present position, directed a management consultant firm. He also has served as assistant administrator for technology utilization in the National Aeronautics and Space Administration.



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Solid Waste—Plastics Composites: Physical Properties and Feasibility for Production

Liang T. Fan,¹ David G. Retzloff, and Wayne O. Vanderpool

Department of Chemical Engineering, Kansas State University, Manhattan, Kan. 66506

The feasibility of producing a composite by radiation-induced polymerization using solid waste and methyl methacrylate as starting materials was investigated in this work. Composites were prepared from the following solid wastes: used computer printout paper containing green dye and ink printing, newsprint, sawdust from fir lumber, cardboard, bagasse, wheat straw, dried sediment from the primary treatment of municipal sewage, sludge from secondary sewage treatment, cotton cloth, clothes lint, and human hair. These preliminary composites machined easily and their water absorption rates were inversely related to their polymer contents. The compression and tensile strengths as well as Rockwell hardness of the waste paper composites were found to be comparable or superior to those of standard construction materials such as concrete, aluminum, and wood. Samples prepared under anerobic conditions exhibited greater tensile and compression strength than those prepared under ambient conditions. The Rockwell hardness values of the samples were insensitive to the presence of oxygen during preparation. From their observed physical properties it is concluded that such composites are suitable for use as wood substitute materials.

he U.S. is currently faced with an ever-increasing quantity of solid waste and the problem of disposing of or recycling this material in a manner acceptable to society. These wastes are increasing at the rate of 6% a year as contrasted with a 1% increase in our population (Grinstead, 1969). It has been predicted that by 1980 every person in the U.S. will produce 8 lb of solid waste each day (Environ. SCI. TECHNOL., 1970). In the past this problem was dealt with by disposing of the solid waste in an open dump but because of the increasing scarcity of land and associated environmental problems this method provides only a temporary solution at best. Several alternative solutions to the solid waste problem have been proposed (Grinstead, 1969; Citizen Advisory Committee on Environmental Quality, 1969; Spilhaus, 1969; ASEE, 1969) and a general summary of present disposal methods is given by Witt (Witt, 1971). These solutions fall into three categories: incineration, compacting, and reuse. Most incinerators in current use pollute the air, and the problem of what to do with the unsalvageable residue

remains. Compacting, on the other hand, reduces the waste to sufficiently dense blocks for use as building blocks or disposal in the ocean. These blocks are claimed to be sterile. However, the amount of compressed wastes that can be utilized in construction is unknown and the stability of this material to the various weather conditions to which construction material is subjected remains to be determined. The most attractive alternative is the recycle and reuse of waste materials. Because paper constitutes over 50% of the total waste, most recycle efforts have been directed to this component (Ad Hoc Group for the Office of Science and Technology, 1969). While there are several processes which recycle wastes (Witt, 1971; Felton, 1971; Freed et al., 1971; Engdahl, 1969), the unusable residue from such processes still poses a mounting solid waste problem (Slatin, 1971; Gellman, 1971). By formation of a composite of solid waste and monomer, the residue problem can be avoided and the products of the process should be a good substitute for wood-based materials. This method, if feasible, has three distinct advantages over other recycle methods, namely: the products of such a process can be used as starting materials once they have served their purpose and finally end up as solid waste, the process appears suitable for handling the solid waste residue from other recycle processes, and the process produces no residue. From the studies on plastic-impregnated woods (Chem. Eng., 1965; Ballantine, 1965; Kent et al., 1963a,b; Bliss et al., 1963) as well as bamboo-plastic and bagasse-plastic (National

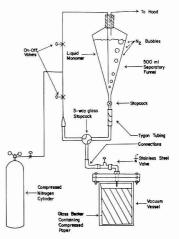


Figure 1. Monomer introduction system

¹ To whom correspondence should be addressed.

Science Council, 1970), it appears possible to form plastic composites with solid waste. This paper describes the properties of such composites when formed from paper waste and the results of the preliminary experiments which were conducted to test the possibilities of forming plastic composites with other materials found in solid wastes.

Experimental

Equipment. The experimental equipment consisted of a mechanical shredder, hydraulic press with compression dyes, vacuum system and vessel capable of achieving 10^{-9} torr, radiation vessel, monomer introduction system, and a Gamma-cell 220 irradiation cell. The radiation and vacuum vessels were fabricated in this laboratory and are identical with the exception that the radiation vessel has no inlet ports. The monomer introduction system including the radiation vessel is shown in Figure 1.

Materials. Methyl methacrylate (MMA) superior grade MCB No. 6683-MX1150 was the monomer used. High-purity nitrogen was employed to provide the inert atmosphere. The solid wastes investigated consisted of used computer printout paper containing green dye and ink printing, newsprint, sawdust from fir lumber, cardboard, wheat straw, bagasse, dried sediment from the primary treatment of mu-

Run 1

nicipal sewage, sludge from secondary sewage treatment, cotton cloth, clothes lint, and human hair.

The major part of the experimental work involved computer printout paper wastes with preliminary studies being done on the other waste materials.

Procedure. In the preliminary studies, all the samples were prepared in the following manner. The waste samples were placed in a petri dish lined with aluminum foil to prevent the polymer from sticking to the glass during irradiation. The petri dish containing the sample together with a beaker of liquid monomer and the radiation vessel were placed in a glove bag which was connected to a high-purity nitrogen source. The samples were purged with nitrogen for 30 min to remove any free oxygen. In addition, nitrogen was forced through a fritted glass tube placed in the monomer to remove the dissolved oxygen. Upon completion of the above, a fixed quantity of the monomer was poured over the solid waste in the petri dish which was then placed in the radiation vessel. The vessel was sealed under a nitrogen atmosphere and placed in the Gammacell where the radiation-induced polymerization occurred. After a given period of time the samples were removed, the weight ratio of polymer to waste material determined, and the samples cut with a saw to compare their sawing properties to those of wood. Each sample was quali-

Table I. Results of Irradiation Experiments

Irradiation time: 6 hr (corresponds to an absorbed dose of 8.01×10^5 rads) All samples were prepared in a nitrogen atmosphere

Sample no.	Waste material	Wt. ratio of monomer added/ waste material (before irradn)	Wt. ratio of polymer/waste material (after irradn)	Bonding properties	Sawing properties	Wt % increase of H ₂ O after 3 days
1	Layered newspaper	1.0	0.0	None		
2	Layered newspaper	0.5	0.465	None		
3	Layered newspaper	0.2	0.019	None		8 - 40.40
4	Layered newspaper	2.0	1.220	Good	Like wood	18.291
5	Layered computer paper	1.0	0.573	Fair	Easier than wood	36.43
6	Layered computer paper	0.5	0.141	None		10101
7	Layered computer paper	0.2	0.033	None		X 408
8	Layered computer paper	2.0	1.518	Good	Harder than wood	11.584

Run 2
Irradiation time: 12 hr (corresponds to an absorbed dose of 16.02×10^5 rads)
All samples, except no. 4-6, were prepared in a nitrogen atmosphere. These three samples were prepared in
the presence of air. Samples 5 and 6 were drilled and turned on the lathe. Both showed good working properties

	the presence of an. Samp	les 5 and 6 were drifted a	ind turned on the la	tille. Dotti silowet	good working propertie	-5.
1	Layered computer paper	2.0	1.913	Good	Harder than wood	7.927
2	Layered computer paper	1.0	0.918	Good	Like wood	19.040
3	Layered newspaper	2.0	1.870	Good	Like wood	7.436
4	Layered newspaper	2.0	1.867	Good	Like wood	6.581
5	Layered computer paper	2.0	1.918	Good	100 B	
6	Layered computer paper	1.0	0.882	Good	23 5	2
7	Pure methyl methacrylate	· 133		Good	Harder than wood	1.137

tatively evaluated with respect to the bonding of the polymersolid waste composites. A representative group of samples were weighed, dipped in water, blotted with filter paper and weighed again to determine the weight of sample which just had water adsorbed on the surface. Using the wet weight as a basis to eliminate the effect of surface pores, the percentage increase in weight of the sample immersed in distilled water for three days was determined.

For the waste paper samples the above procedure was used for the case of layered paper. Additional samples were also prepared under atmospheric conditions to assess the effects of oxygen on the final composite product. The shredded paper samples were prepared using a Waring Blendor inside the glove bag under a nitrogen atmosphere. A known volume of monomer was then added and the blendor turned on again. After thorough mixing, the samples were placed in aluminum foil-lined petri dishes which were then placed in the radiation vessel. The rest of the procedure is identical to the one originally given.

For the detailed study on waste paper, various modifications of a general procedure were employed to ascertain the effects of oxygen and method of preparation on the properties of the final composite. The general procedure will be given first followed by the various modifications.

Run 4

The waste paper was first cut into 2-in. squares and shredded into irregular-shaped pieces less than $1/_2$ in. square. The shredded paper was then compressed into cylindrical shapes (plugs) at various pressures, weighed, and placed in glass beakers.

The modifications are:

METHOD 1. The beaker containing the paper plug was placed in the vacuum vessel and evacuated to a final pressure of 50 mtorr. At the same time nitrogen was bubbled through the monomer for 15 min to remove any dissolved oxygen. The monomer was introduced into the glass beaker via the monomer introduction system, Figure 1. After the plug had soaked in monomer for 6 hr, the vacuum vessel was returned to atmospheric pressure under a nitrogen atmosphere and placed in a glove bag also under a nitrogen atmosphere and containing the irradiation vessel. The vacuum vessel was then disassembled, the excess monomer removed, and the beaker containing the plug placed in the irradiation vessel. The irradiation vessel was then sealed under a nitrogen atmosphere and placed in the Gammacell.

METHOD 1A. This procedure is identical to Method 1 except that the plug was placed in a glass tube of nearly the same diameter and length as the plug which was then put in the beaker. The plug was also left submerged in a pool of mo-

Run 3

Irradiation time: 12 hr (corresponds to an absorbed dose of 16.02×10^5 rads)

Samples 1 and 6 were drilled and turned on a lathe. No. 1 drilled very easily but chipped badly while being turned on the lathe. While drilling no. 6, the bit heated up, but this sample could be easily turned on the lathe.

Table I. Continued

Sample no.	Waste material	Wt. ratio of monomer added/ waste material (before irradn)	Wt. ratio of polymer/waste material (after irradn)	Bonding properties	Sawing properties	Wt % increase of H ₂ O after 3 days
1	Shredded computer paper	Saturated	2.982	Fair		
2	Shredded computer paper	1.00	0.4631	Fair	Easier than wood	59.92
3	Shredded computer paper	Saturated	7.246	Good	Harder than wood	3.654
4	Layered cardboard	Saturated	1.066	Good	Easier than wood	9.057
5	Shredded newspaper	2.00	1.105	Fair	Much easier than wood	40.212
6	Shredded newspaper	Saturated	5.990	Good		
7	Sawdust	Saturated	4.369	Good	Harder than wood	6.571

Irradiation time: 12 hr (corresponds to an absorbed dose of 16	5.02×10^5 rads)	
All samples were prepared in a nitrogen atmosphere.		

1	Bagasse	4.748	4.202	Good	Much harder than wood	6.781
2	Wheat straw	5.711	1.998	Fair	Easier than wood	20.465
3	Cloth	2.293	2.107	Good	Much harder than wood	6.360
4	Clothes lint	12.581	12.238	Good	Harder than wood	13.247
5	Human hair	5.397	5.185	Fair	Harder than wood	7.797
6	Solid waste	1.589	1.385	Good	Much harder than wood	14.217
7	Sludge	1.367	1.008	Poor		
8	Pure monomer			Good		

nomer during soaking and irradiation. After irradiation the homopolymer and glass tubing were removed.

METHOD 2. The paper plug was not evacuated but prepared in air and placed in a weighing bottle. The dissolved air in the monomer was removed as in Method 1 and the monomer was added to the weighing bottle in an amount sufficient to just cover the plug.

The plug was allowed to soak for 6 hr in the pool of monomer and then the excess monomer was removed. The weighing bottle was sealed and placed in the irradiation vessel which was placed in the Gammacell.

METHOD 2A. This method is identical to Method 2 except that the plug was again placed in a glass tube of nearly the same diameter and length as the plug which was then placed in the beaker. The plug was also left submerged in a pool of monomer during soaking and irradiation. After irradiation the homopolymer and glass tubing were removed.

METHOD 3. This is similar to Method 1a except that the monomer was bubbled with air for 15 min instead of nitrogen prior to introduction to the evacuated paper plug.

METHOD 4. This is similar to Method 2a except that the monomer was bubbled with nitrogen for 15 min instead of air prior to introduction to the paper plug prepared in air.

To test the final composite plugs for tensile and compression strength both perpendicular and parallel to the direction of compression, the plugs were first cut as shown in Figure 2. The numbering of the pieces and the shapes of the final test samples are also given in this figure. Cylinder bars numbers 4 and 6 were used for the water adsorption studies. A Riehler Tensile Tester, Model FS-20, and a Southwark-Emery Compression Tester were employed to obtain the tensile and compression strengths. A Rockwell Hardness Tester (method similar to ASTM test D143-52) was used to obtain hardness data.

For the main investigation the procedure for the water absorption studies was similar to that previously given except that the samples were painted on their sides with black gytol paint which is impermeable to water. In this way the absorption of water is in one direction only and the rate of water absorption perpendicular and parallel to the direction of compression of the waste paper can be determined. Samples were immersed in water and removed at various times to determine the percentage weight increase.

Results and Discussion

The results for the preliminary study on various types of solid waste are summarized in Table I. When the polymer-towaste material weight ratio of the composite was ≥ 1 , the polymer appeared to bind well to the waste material. Dissolved oxygen in the monomer did not appear to affect the bonding. The ease with which the composite could be sawed was directly related to the polymer content; the higher the polymer content, the more difficult it became to cut the composite. It was observed that for waste paper composites, the composites with higher polymer content were easier to shape on a lathe and exhibited no chipping. Water absorption studies indicated that the amount of water absorbed was a function of the polymer content. A typical result is shown in Figure 3. Results for other samples are summarized in Table I. These values are quite low compared to the percentage weight increase for various woods observed by Bliss et al. (1963) which ranged from a weight increase of 58% for untreated oak to 150% increase for untreated cottonwood. In all cases studied, the final composites were stronger than the original waste materials. Because of the flammability of MMA, the composites

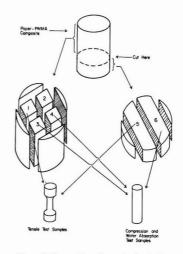


Figure 2. Preparation of samples for testing

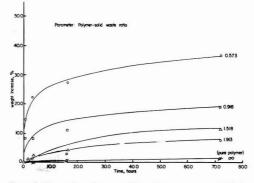


Figure 3. Water adsorption rate of layered computer paper-PMMA composite

ignited and sustained combustion when exposed to an open flame indicating that either a fire retarder or another nonflammable monomer should be used if the composites are to be suitable for wood substitutes.

The absorbed dose needed to obtain a given conversion of monomer to polymer in the final composite was determined. This is presented in Figure 4. The curves labeled "conversions in nitrogen" and "conversions in air" are consistent with the usual observations for free-radical polymerization in which the free radical preferentially reacts with the oxygen until it is consumed; then the polymerization proceeds. These results were obtained when the excess monomer was removed prior to irradiation. However, when the samples are left in a pool of monomer during irradiation (immersion technique), the results lie somewhere between these two outer curves, and no significant effect of the dissolved oxygen on the absorbed dose needed to affect a given monomer conversion is discernible. Table II indicates that while the effect of dissolved oxygen is slight, the removal of oxygen from the liquid monomer is more effective than evacuating the compressed waste paper. From Figure 4 it also can be seen that the pressure used to compress the waste paper has no significant effect on the dose

Table	II.	Effect	of	Oxygen	on	the	Percent	Conversion	of
		Mono	me	r in Paper	-PN	MMA	Compos	sites	

Sam- ple no. ^a	Method of preparation	% polymer	Paper-to- polymer ratio	Conv of monomer
41	Method 3	31.7	0.464	77.4
42	Method 1a	32.8	0.489	81.5
43	Method 2a	31.5	0.461	76.8
44	Method 4	31.5	0.461	76.8

^aAll samples were irradiated for 8.0 hr which sorbed dose of 8.99×10^5 rads.

needed for a given conversion of monomer. The absorbed dose needed to reach a specified conversion of monomer to polymer in the final composite can also be obtained from this figure.

When the samples were machined prior to testing their physical properties, small voids were observed in their interior. This appears to be caused by local heating during polymerization which induces the monomer to vaporize. These bubbles then become trapped in the paper-monomer mixture and form voids in the final composite. The effect is observed in samples which have a high polymer content. It is believed that in this case the paper-monomer-polymer mixture is unable to absorb completely and dissipate the heat of polymerization thus leading to a local heating condition. When the waste paper was compressed to a pressure of 5000 psig or greater, no voids were present. This is due to the fact that a smaller amount of monomer is being polymerized and the large amount of paper present is able to dissipate the heat and keep the temperature below the boiling point of the monomer. Additionally, during preparation of the samples for testing their physical properties, it was observed that the composite samples were somewhat more difficult to machine than the pure polymethyl methacrylate (РММА).

The physical properties of the composites are summarized in Table III. Samples prepared in a nitrogen atmosphere exhibited substantially greater compression and tensile strength than those prepared in air. In all cases the compression strength was greater in the direction of original compression

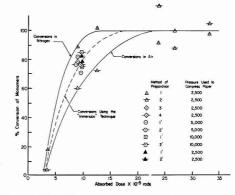


Figure 4. Effects of oxygen and paper compression pressure on conversion of monomer

of the waste paper while the opposite is true of the tensile strength. This is as expected because compressing the paper prior to irradiation should increase the compression strength parallel to the direction of paper compression and also lead to a greater tensile shear strength perpendicular to the direction of paper compression similar to that observed in reinforced concrete or polymers. The hardness of the samples appears to be relatively insensitive to the presence of air during irradiation of the samples and increased with increasing paper compression pressure until the compression pressure reached 5000 psig. Above this value no discernable influence of paper compression pressure was observed. The hardness of Douglas fir wood is given in Table III for comparison. Finally, the amount of water absorbed and the rate of water absorption are inversely correlated with the polymer content of the final composite. The higher the polymer content, the less water absorbed in a given time. Typical results are shown in Figure 5 and Table III. A comparison of the physical properties of the composites with other well-known building materials is given in Table IV. The composites compression strength exceeds that of aluminum while the tensile strength is comparable to that of pure PMMA and greater than that for concrete or concrete-polymer materials.

			Table III. Pr	operties of Pape	r-PMMA	Composites			
Sample no.	Pressure used to compress paper, psi	РММА, %	Compression Parallel	n strength, psi Perpendicular	Tensile Parallel	strength, psi Perpendicular	Water ab Final % wt increase	osorption Time, hr	Rockwell hardness value
\mathbf{I}^{a}	225	71.7	$15,700^{b}$	$6,500^{\circ}$	220 ^b	2,230 ^c	6.9	398 ^d	169
\mathbf{H}^{a}	5,000	34.2	26,300	11,500°	320 ^b	$6,440^{c}$	16.0	402^{d}	176
\mathbf{III}^{a}	5,000	41.8	24,300	$12,000^{\circ}$	190 ^b	5,590°	25.1	265^{d}	175
IV^e	5,000	35.7	19,600	$7,300^{c}$	140^{b}	3,590°	30.0	338^{d}	146
\mathbf{V}^{a}	5,000	38.3	28,700	$12,800^{c}$	280^{b}	$7,060^{c}$	17.7	384^{d}	171
VI^a	500	65.5	9,800	9,800°	80 ^b	$2,890^{c}$	16.5	384^{d}	131
VII ^a	10,000	27.3	$20,400^{b}$	$10,200^{c}$	210 ^b	$4,290^{c}$	34.0	262^{d}	174
10A	Wood (Do fir) para		7,962						24.8
100		endicular ^g	1,029						21.5
a Samples in	radiated in a nitre	agen atmosphe	aro.						

^a Samples irradiated in a nitrogen atmosphere.
 ^b Strength parallel to the direction in which the paper was compressed.

Strength perpendicular to the direction in which the paper was compressed.
 Time in hours to reach 95% of final weight increase.

Sample irradiated in an air atmosphere

/ Force applied parallel to grain of wood.
 / Force applied perpendicular to grain of wood.

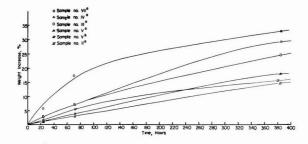


Table IV. Comparison of Properties of Paper-PMMA Composite with Other Standard Materials

Material	Tensile strength, psi	Compression strength, psi	Density, lb/ft ³
Steel ^a	70,000	42,000	490
Aluminum ^a	30,000	25,000	165
Concrete ^a	400	5,000	156
Concrete-polymer			
$(6\% \text{ pmma})^a$	1,600	20,000	165
Wood polymer ^b		12,000	
Wood (Douglas fir) ^c		7,400	30
Wood (Douglas fir) ^d		8,000	
Pure PMMA (TC) ^e	7,000	15,000	62
Pure PMMA (TC) ^f	6,200	15,200	62
Pure PMMA (RI) ^g	10,600	17,700	62
Composite sample (V)			
Paper-РММА composite (38% РММА-рарег compressed at			
5000 psi) ^h	$7,000^{i}$ (280) ^k	$28,600^{j}$ $(12,800)^{l}$	80.5

^a Steinberg et al. (1968).

^b Birch-PMMA combination (Ballantine, 1965). Miner and Seastone (1955).

d Compression strength found experimentally, force applied parallel to grain in wood.

^е Риге Рима, formed thermal catalytically, Steinberg et al. (1968).
 ^f Риге Рима, formed thermal catalytically, properties experimentally

determined. *Pure PMMA*, formed by radiation-induced polymerization, properties

 A to FMM, but for the control of the control of the control of the formation, properties experimentally determined.
 Composite produced using radiation-induced polymerization of MMA monomer, properties experimentally determined.
 Collection of the control of the contro * Pulled in a direction perpendicular to the direction in which the

i Force applied perpendicular to the direction in which the paper was compressed.

* Pulled in a direction parallel to the direction in which the paper was

compressed. ¹ Force applied parallel to the direction in which the paper was compressed.

Conclusions

The basic concept of forming a solid waste-plastic composite is feasible. The process will accommodate a variety of solid waste similar to that found in commercial and industrial wastes, and there are no residual wastes.

The formation of voids in the final composite owing to the vaporization of the monomer by a local heating effect related to the heat of polymerization can be avoided by using higher paper compression pressures and maintaining the polymer content of the composite below some critical value which was found to be about 50 wt % polymer for a MMA monomer using the immersion technique. This corresponded to a volume Figure 5. Water absorption rates for various paper-**PMMA** composites

a. Direction of water absorption was parallel to direction in which paper was compressed b. Direction of water absorption was perpendicular to direc-

tion in which paper was compressed

percent polymer ranging from approximately 40% for samples prepared at a paper compression pressure of 225 psig to 55% for samples prepared at a paper compression pressure of 10,000 psig. The tensile and compression strengths of the composites are significantly greater when the dissolved oxygen is removed from the monomer prior to irradiation. However, the absorbed dose rate needed to achieve a specified conversion of the monomer to polymer is not significantly affected by either the paper compression pressure used or the presence of air in the monomer when the composite is formed using the immersion technique. The Rockwell Hardness of the composites is insensitive to the atmosphere in which the samples are prepared and increased with increasing sample compression pressure only up to 5000 psig. The hardness of the composites was vastly superior to that of wood.

While the results indicate that plastic-solid waste composites can be produced with favorable physical properties for use as wood substitute materials, the use of gamma radiationinduced polymerization to produce such composites is an expensive one. Thus it is desirable to make and test the physical properties of such composites produced by more conventional, and usually less costly, polymerization techniques before an economic assessment of the industrial possibilities of this process can be made. Work in this direction is presently under way in this laboratory.

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Influence of Coriolis Forces on Airflow in Conifuges

Werner Stöber1 and Thomas T. Mercer

Department of Radiation Biology and Biophysics, University of Rochester, Rochester, N.Y. 14642

Due to the radial component of the airflow in the annular conical ducts of centrifugal aerosol size spectrometers known as conifuges, the Coriolis forces of the rotating airflow have a tangential component which accelerates the airflow to an angular velocity relative to the cone. For a given rotor speed, this angular motion limits the range of total airflow rates suitable for the proper operation of the instrument. The influence of the Coriolis forces was estimated earlier by employing a crude mathematical model assuming that the angular velocity of the air relative to the cone remains small compared to the rotor speed. In this paper, a more adequate model is used to determine the maximum permissible Reynolds numbers of the tangential airflow in conifuges. A critical value of 9000 is suggested.

he first operational design of an actual aerosol size spectrometer providing continuous spectra of aerodynamic particle sizes by utilizing centrifugal forces was the so-called conifuge (Sawyer and Walton, 1950). The original instrument and a number of modified versions of more recent years (Keith and Derrick, 1960; Stöber, 1967; Tillery, 1967; Hauck and Schedling, 1968; Hochrainer and Brown, 1969) feature a rotor with a conical, annular duct where the sizeseparating deposition of the aerosol takes place on the outer conical wall. In these centrifuges, particles of equal aerodynamic size are located on concentric rings on the envelope of the cone.

A major modification of the original design is the cylindrical conifuge having a ring slit entrance for the aerosol. In this case, the cone is replaced by a cylinder. Although this model has the advantage of a rigorous theory available for an absolute evaluation (Stöber and Zessack, 1966; Stöber et al., 1969), only a few small devices of this kind have been built so far (Berner and Reichelt, 1969; Hochrainer and Brown, 1969). Apparently, the conical design has prevailed for such practical reasons as extending the range of deposited particles to smaller sizes or facilitating a simpler way of exchanging and positioning the sampling foils.

In an attempt to design a ring-slit conifuge of optimal performance (Stöber and Flachsbart, 1969), a significant drawback of the conical concept was pointed out and taken into consideration: Since the conical duct causes a radial component of the flow velocity, the laminar airflow in the duct will be subjected to tangential Coriolis forces causing an angular acceleration of the air against the direction of rotation of the conifuge. To reduce this influence, a compromise design with a small half-angle of 20° at the apex of the cone was adopted (Figure 1) and an estimate of the general influence of



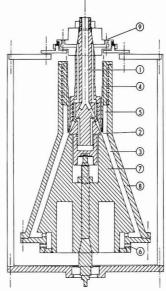


Figure 1. Design of ring-slit conifuge (different shading discriminates rotating from nonrotating parts)

(1) nonrotating aerosol duct, (2) ring slit aerosol inlet, (3) annular conical duct, (4) capillary air inlet, (5) annular cylindrical air duct, (6) outlet jets, (7) rotor cone, (8) rotor lid, (9) coolant duct

¹ To whom correspondence should be addressed.

Coriolis forces on the airflow pattern was made and compared with experimental results.

Although experiments at high flow rates or high rotor speeds indicated that the Coriolis forces were, indeed, a limiting factor for the selection of useful operating conditions for conifuges with conical annular ducts, the theoretical estimate was too crude to give commensurate results. The mathematical model employed in the study was valid only if the angular velocity ω of the air relative to the cone remained small compared to the rotor speed Ω . However, some of the operating conditions exceeded that regime. This is at least one of the reasons why unusually high critical Reynolds numbers for laminar tangential flow were encountered in comparing calculations and experimental results. There were also numerical simplifications influencing the comparative calculations.

A more realistic estimate of the tangential airflow in conifuges must account for the reduction of the Coriolis forces in case the relative angular flow velocity ω reaches a sizable fraction of the rotor speed Ω . The following is a recalculation of the influence of Coriolis forces on the airflow in conifuges without the restraints of the earlier estimate.

Mathematical Model

Figure 2 shows schematically one half of the cross section through a ring-slit conifuge in a plane containing the axis of

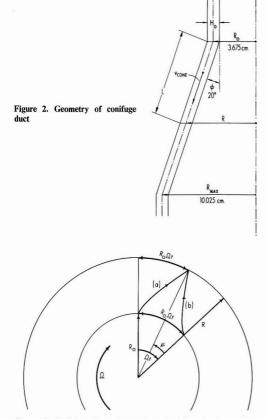


Figure 3. Radial and angular motion of airflow in the conifuge (a) absolute path of volume element of airflow, (b) path of volume element of airflow relative to rotor

rotation. At any location indicated by the radius vector R, the average flow velocity along the cone is given by

$$V_{\rm cone} = \frac{F}{2 \pi R H_o \cos \phi} \tag{1}$$

The radial component of this velocity is

$$\dot{R} = \frac{F \tan \phi}{2 \pi R H_a} \tag{2}$$

Integrating this equation for the initial condition $R = R_o$ at t = 0 gives

$$R = R_o \sqrt{1 + \alpha Ft} \tag{3}$$

where

$$\alpha = \frac{\tan \phi}{\pi R_o^2 H_o} \tag{4}$$

By disregarding the actual velocity profile of the airflow, Equation 3 represents the average radial motion of the air in the duct. Thus, an average residence time, t_{max} , of a volume element of air in the duct can be calculated as

t

$$r_{\max} = \frac{\sigma}{F}$$
 (5)

with

$$\sigma = \frac{R_{\max}^2 - R_o^2}{\alpha R_o^2} \tag{6}$$

In case of ideal, nonviscous flow, the radial motion of the air in the spinning duct would create a tangential component

$$R\psi = -(R - R_o)\Omega t \tag{7}$$

as shown in Figure 3. This implies that the air has no angular velocity relative to the rotor at $R = R_o$. Differentiation with respect to time gives

$$\dot{R}\psi + R\dot{\psi} = (R_o - R)\Omega - \dot{R}\Omega t \tag{8}$$

and

$$\ddot{R}\psi + 2\,\dot{R}\dot{\psi} + R\ddot{\psi} = -2\,\dot{R}\,\Omega - \ddot{R}\,\Omega t \tag{9}$$

From Equations 7 and 8, we find

$$\dot{\psi} = \omega_t = -\Omega \left[1 - \frac{R_o}{R^2} (R - \dot{R}t) \right]$$
(10)

and by utilizing Equation 3

$$\omega_{i} = -\Omega \left[1 - \frac{2 + \alpha F t}{2 (1 + \alpha F t)^{3/2}} \right]$$
(11)

This would be the angular velocity of the air relative to the rotating cone in case of ideal flow.

For the actual case of viscous flow, we make use of Equation 9 which can be rearranged to

$$\ddot{\psi} = \dot{\omega}_i = -\left[\frac{\ddot{R}}{R}(\psi + \Omega t) + 2\frac{\dot{R}}{R}(\dot{\psi} + \Omega)\right]$$
(12)

Equation 12 indicates the apparent angular acceleration inherent to the rotating system. The first term in the brackets accounts for the influence of the deceleration of the radial flow velocity while the second term represents the general form of the Coriolis forces.

The angular acceleration $\dot{\omega}_i$ is counteracted by a drag force,

which may be approximated by the frictional force per volume element calculated for a one-dimensional flow between two planes at distance $H_o \cos \phi$ (Lamb, 1932)

$$-\frac{dp}{dx} = \frac{12 \eta}{H_o^2 \cos^2 \phi} V$$
 (13)

This is equivalent to an angular deceleration

$$\dot{\omega}_f = \frac{1}{R\rho_{\rm air}} \frac{dp}{dx} = -\frac{12\nu}{H_o^2 \cos^2 \phi} \dot{\psi} \tag{14}$$

and the ruling differential equation for viscous flow is then

$$\dot{\psi} = \dot{\omega}_i + \dot{\omega}_f = -\left[\frac{\vec{R}}{R}(\psi + \Omega t) + 2\frac{\dot{R}}{R}(\dot{\psi} + \Omega) + \frac{12\nu}{H_0^2\cos^2\phi}\psi\right]$$
(15)

In terms of the flow in the conifuge as represented by Equation 3, we obtain

$$\ddot{\psi} = \dot{\omega} = \frac{\alpha^2 F^2(\psi + \Omega t)}{4 (1 + \alpha F t)^2} - \frac{\alpha F(\omega + \Omega)}{1 + \alpha F t} - \frac{12 \nu}{H_o^2 \cos^2 \phi} \omega \quad (16)$$

Differentiating this expression with respect to time, and rearranging the equation gives

$$-\ddot{\omega} = \left(\frac{12 \nu}{H_o^2 \cos^2 \phi} + \frac{3 \alpha F}{1 + \alpha F t}\right) \dot{\omega} + \left[\frac{24 \nu \alpha F}{H_o^2 (1 + \alpha F t) \cos^2 \phi} + \frac{3 \alpha^2 F^2}{4 (1 + \alpha F t)^2}\right] \omega + \frac{3 \alpha^2 F^2 \Omega}{4 (1 + \alpha F t)^2} \quad (17)$$

For simplicity we introduce

$$\tilde{\omega} = \frac{\omega}{\Omega} \tag{18a}$$

$$\tau = \frac{t}{t_{\text{max}}} \tag{18b}$$

$$\chi = \alpha \sigma \tag{18c}$$

$$\gamma = \frac{12\nu}{H_{c}^{2}\cos^{2}\phi}$$
(18d)

$$K = \gamma t_{\rm max} \tag{18e}$$

$$f(\tau) = \frac{\chi}{1 + \chi\tau} \tag{18f}$$

so that Equation 17 becomes dimensionless

$$-\ddot{\omega} = [K+3 f(\tau)]\dot{\omega} + [2 Kf(\tau) + \frac{3}{4}f^{2}(\tau)]\tilde{\omega} + \frac{3}{4}f^{2}(\tau)$$
(19)

This equation has no analytical solution except for K = 0, which represents the case of ideal flow already solved by Equation 11. For the general case of viscous flow, it is necessary to make numerical integrations by means of a computer. The solutions have to cover the range $0 \le \tau \le 1$ under initial conditions $\tilde{\omega} = 0$, $\dot{\omega} = -\chi$ at $\tau = 0$.

However, before actually computing specific solutions for various conifuge designs and operating conditons, we can conclude from Equation 19 that besides the pure design parameter

$$\chi = \frac{R_{\max}^2 - R_o^2}{R_o^2}$$
(20)

there is only one more dimensionless parameter

$$K = \frac{12 \nu \pi (R_{\max}^2 - R_o^2)}{H_o F \sin \phi \cos \phi}$$
(21)

ruling Equation 19. This parameter has the quality of an inverse Reynolds number and contains the total flow rate *F* as the only influential operating variable. Thus, by the definition of $\tilde{\omega}$, we conclude that any change of the rotor speed Ω at constant flow rate *F* will only cause a proportional change of the angular velocity ω .

For physical reasons, we can also predict that the solutions of Equation 19 will have a general pattern where the absolute value $|\tilde{\omega}|$ of the angular velocity of the air relative to the cone will first rise from zero to a maximum value until the frictional forces will exceed the tangential Coriolis forces because of the deceleration of the radial flow component. From there on, the angular velocity $|\tilde{\omega}|$ will gradually decrease and finally vanish. Patterns of this kind were obtained in the earlier estimate (Stöber and Flachsbart, 1969).

This implies that there will also be a maximum value for the Reynolds number of the tangential airflow

$$\operatorname{Re} = \frac{R\omega}{\nu} \frac{4 Q}{U} = 2 \frac{R\omega}{\nu} H_o \cos \phi \qquad (22)$$

which will be reached at

$$\operatorname{Re}_{\max} = \frac{2 \, \Omega H_o \cos \phi}{\nu} \, (R\tilde{\omega})_{\max} \tag{23}$$

Depending on the design and the operating conditions of the conifuges under consideration, this maximum value will appear somewhere between R_o and R_{max} of the cone.

Since the proper performance of the conifuges relies on the continuity of the laminar flow of the air in the duct, Equation 23 must not exceed the critical Reynolds number Re_{crit} for laminar flow in the tangential direction. The actual value of Re_{crit} is not precisely known, but we can conclude that

$$\frac{2H_o\cos\phi}{\nu} \Omega(R\tilde{\omega})_{\rm max} < {\rm Re}_{\rm erit}$$
(24)

is complied with for all values of $\Omega(R\tilde{\omega})_{\max}$ pertaining to operating conditions which produce regular concentric deposition patterns for monodisperse aerosols throughout the deposit foil.

For a given design, $(R\tilde{\omega})_{\max}$ increases with increasing flow rate *F* as the only variable. Thus, the choice of operating conditions for *F* and Ω is limited by

$$\Omega(R\tilde{\omega})_{\max} < \frac{\mathrm{Re_{erit}}}{2 H_{\varrho} \cos \phi}$$
(25)

In other words, if Re_{crit} is independent of the operating conditions, then the maximum permissible rotor speed Ω will decrease with increasing flow rate *F*.

Numerical Results and Discussion

By using the appropriate values for the parameters of design and operation, Equation 19 can be solved numerically for various conifuges to give the instantaneous dimensionless angular velocity $\tilde{\omega}$ of the air along the cone. At this point, however, it should be recalled that these solutions have the quality of an approximation, because of the restraints of Equation 13, which is derived for a flow at an equilibrium of forces. Since the centrifuge rotors represent an accelerated system, the solutions of Equation 19 will probably give somewhat enhanced values for the maximum angular velocities $\tilde{\omega}_{max}$ because the inertia of the air is not accounted for. With this reservation, the solutions of Equation 19 may be combined with Equation 22 to estimate the instantaneous Reynolds numbers of the tangential flow at different locations (Figure 2):

$$L = \frac{R - R_o}{\sin \phi} \tag{26}$$

Corresponding calculations were made on an IBM 360 computer for the standard operating conditions of several conifuge designs. Table I gives the parameters employed in the calculations and Figure 4 illustrates some of the results with regard to the instantaneous angular velocities of the angular flow relative to the cone. The graph indicates that, indeed, sizable fractions of Ω are encountered for the angular velocity ω of the air relative to the cone. This is particularly prominent with the instrument designs where the aerosol is entrained at the apex of the cone. In these cases, an increase in flow rate creates a steep increase of ω to a maximum at short distances from the apex and a rapid decline afterwards. For the Hauck and Schedling design, the maximum angular velocity was calculated at 74% of the rotor speed Ω at a distance L of 1.7 cm from the apex. Similar values between 40 and 68% were obtained for the Keith and Derrick conifuge. The maxima for the ring-slit conifuge are less pronounced, thus permitting higher flow rates. However, for a flow rate of 23 l./min, the maximum angular velocity of the air approaches 37% of the

Table I. Conifu	ige Desi	gn Para	neters	
Design	$H_o,$ cm	φ	$R_o,$ cm	R _{max} , cm
Keith and Derrick (1960)	1.00	45°	0.5	9.33
	1.00	45°	1.5	10.33
Hauck and Schedling (1968)	1.00	45°	0.5	9.50
Tillery (1967)	1.00	45°	0.5	9.70
Sawyer and Walton (1950)	0.58	30°	0.3	3.52
Hochrainer and Brown (1969)	0.57	30°	0.84	4.43
Stöber and Flachsbart (1969)	1.00	20°	3.68	10.03

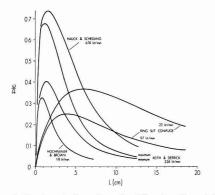


Figure 4. Angular velocity patterns for different conifuge designs

rotor speed and drops less rapidly toward the base of the cone, where it still amounts to about 20%.

Apparently, for high flow rates in the ring-slit conifuge, there is an appreciable reduction of the effective centrifugal force depositing the aerosol particles in the conifuge. At 23 L/min, the tangential airflow reduces this force to at least 65% or less. As a consequence, the indirect experimental determination of the flow rate must have given figures in excess of the true value, because for the computations a simple equation (Stöber and Flachsbart, 1969)

$$\frac{N^2 (D_{\rm kin}{}^2 + 2 \ \beta D_{\rm kin})}{F} = f(L) \tag{27}$$

had been utilized which does not account for the tangential motion of the air relative to the cone. This is qualitatively in keeping with the finding that the flow rates thus determined were considerably higher than in simulating static experiments.

Figure 5 gives the results for the instantaneous Reynolds number of the tangential flow relative to the cone under the actual standard rotor speeds of the different conifuges. The graph shows that the unusually high values for the Reynolds numbers of the Keith and Derrick centrifuge as obtained in the earlier estimate by Stöber and Flachsbart (1969) are now substantially reduced. The new values are of the same magnitude as found for the ring-slit conifuge. The values for the Hauck and Schedling conifuge and the instrument of Hochrainer and Brown are considerably smaller for their respective standard operating conditions.

Considering the performance of the conifuges under the specified operating conditions, it appears that, with regard to the tangential flow, relatively high Reynolds numbers of the order of 10,000 as derived in the above calculations will still permit a laminar tangential airflow. For reasons already mentioned, the actual angular velocities and, thus, the actual Reynolds numbers may be somewhat lower. In any case, the value of 10,000 derived as above must be considered critical. This can be concluded from the data for two different operating conditions of the ring-slit conifuge in Figure 5. At 6000 rpm and a total flow rate of 9.7 l./min, the maximum Reynolds number is 10920 at L = 5.74 cm and there is a drop to 6500 at the base of the cone. In this case, the deposits of monodisperse aerosols are regular over the whole deposit, thus indicating laminar flow conditions. However, at 3000 rpm and 23 l./min, the maximum Reynolds number is only 9530 at L = 10 cm with a drop to about 8000 at the base of the cone, but there is a dis-

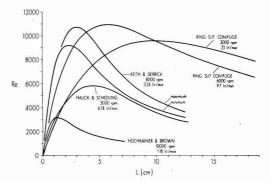


Figure 5. Instantaneous Reynolds numbers for the tangential airflow relative to the cone of different conifuges

Table II. Maximum Angular Air Velocity and Maximum Reynolds Numbers of Tangential Flow Compared to Deposit Patterns at Different Operating Conditions of the Ring-Slit Conifuge

Rotor speed, rpm	Total flow rate, 1./min	$\tilde{\omega}_{\max},$	$L(\tilde{\omega}_{\max}),$ cm	Remax	$L(\text{Re}_{\max}),$ cm	Deposit pattern
1500	8.3	23.1	3.6	2,450	5.0	Regular
1500	5.2	17.8	2.8	1,760	3.8	Regular
3000	23.0	36.5	6.0	9,530	9.9	Diffusive for $L > 14$ cm
3000	10.0	25.4	4.1	5,580	5.7	Regular
3000	4.3	15.9	2.5	3,060	3.3	Regular
3000	2.4	10.8	1.9	1,940	2.2	Regular
6000	46.0	46.1	8.4	28,800	15.3	Diffusive for $L > 8$ cm
6000	24.0	37.1	6.2	19,560	10.1	Diffusive for $L > 10$ cm
6000	9.7	25.0	3.8	10,920	5.7	Regular
6000	7.4	21.8	3.3	9,070	5.0	Regular
6000	3.2	13.1	2.2	4,880	2.8	Regular
9000	14	29.8	4.8	20,890	7.2	Diffusive for $L > 7$ cm
9000	6	19.4	3.0	11,720	4.1	Convective turbulence

turbance of monodisperse aerosol deposits toward the air exit for L > 14 cm. As Table II shows, this is the only inconsistency with regard to a fixed value of the critical Reynolds number being compatible with all experimental operating conditions of the ring-slit conifuge. It may indicate that the critical Reynolds number for tangential laminar flow is slightly increasing with increasing rotor speed. Such effect is known (Schlichting, 1965) as being due to density stratifications of the air in the centrifugal field.

Thus, in comparison with the earlier estimate, the critical Reynolds number indicating unstable flow is reduced by almost a factor of two, and with an approximate value of 9000 it is somewhere in the transitional range between stable and fully developed turbulent flow in ordinary pipes (Dryden et al., 1956). This numerical value may be in excess of the true figure. However, when calculated according to the above derivation, it is probably the limiting value in Equation 25 for the free choice of design and operating conditions of any aerosol centrifuge with an annular conical air duct.

Acknowledgment

The authors gratefully acknowledge the support of David A. Goldstein in programming the computer.

Nomenclature

- $D_{\rm kin}$ = aerodynamic diameter of particle, cm
- = total flow rate in duct, cm³ sec⁻¹ F
- H_{a} = radial width of duct, cm
- L = coordinate of duct length along the side of the cone, cm
- N = rotor speed (see Ω), sec⁻¹
- = pressure, dyn cm⁻² p
- Q cross section of duct, cm²
- R radial coordinate of duct, cm =
- Ŕ radial flow velocity, cm sec-1 =
- R _ radial flow acceleration, cm sec-2
- R_o = initial radial coordinate of duct, cm
- R_{max} = final radial coordinate of duct at base of cone, cm
- Re = Reynolds number = time coordinate, sec t
- = maximum residence time of volume element of air Imax in duct, sec
- U circumference of cross section of duct, cm
- V air velocity, cm sec-1
- $V_{\rm cone}$ = air velocity in the direction of L, cm sec⁻¹

GREEK LETTERS

- ß Cunningham correction (8.2 \times 10⁻⁶ cm)
- η = viscosity coefficient (1.83 \times 10⁻⁴ gram cm⁻¹ sec⁻¹)
- half angle at apex of cone φ =
- density of air (1.29 imes 10⁻³ gram cm⁻³) Pair
- = kinematic viscosity (η/ρ_{air}) , cm² sec⁻¹ v
- ψ = angular coordinate of volume element of air moving relative to cone
- į angular velocity, sec-1 =
- ÿ = angular acceleration, sec-2
- = angular velocity $(=\dot{\psi})$, sec⁻¹ ω
- ώ = angular acceleration, sec-2
- angular velocity of ideal flow, sec-1 ω_i =
- angular acceleration of ideal flow, sec-2 = $\dot{\omega}_i$
- angular acceleration due to viscous forces, sec-2 ŵr
- $\Omega =$ angular velocity of rotor (=2 πN), sec⁻¹

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Growth Rate Determinations of the Macrophyte Ulva in Continuous Culture

Thomas D. Waite,¹ Lloyd A. Spielman, and Ralph Mitchell²

Division of Engineering and Applied Physics, Laboratory of Applied Microbiology, Harvard University, Cambridge, Mass. 02138

■ Continuous culture experiments were run with the benthic macrophyte *Ulva lactuca*. Using oxygen evolution as a monitor of photosynthesis and dry weight determinations for biomass synthesis, growth rates and stoichiometric growth constants were evaluated. The data showed that the ratio of oxygen production to algal mass synthesis is relatively independent of nutrient concentration and growth rate, but is affected by light intensity. The data also showed that the amount of oxygen evolved per unit of algal material was almost a factor of 10 higher than is predicted from carbohydrate synthesis. It appears that *Ulva* is capable of synthesizing compounds with carbon oxidation states of +1 or +2, thus estimates of biomass synthesis may be in error when the average algal material is assumed to be carbohydrate.

Recent evidence has shown that benthic algae may contribute substantially to the productivity of coastal waters (Waite and Mitchell, 1971); however, very little is known about the kinetics of macrophyte growth. The reason for our poor understanding of the growth kinetics of this important group of algae has been the difficulty of accurately measuring productivity responses to changes in nutrient concentration.

Continuous-flow culture assays using chemostats or algastats have recently become popular for growth rate determinations. Biological populations can be kept at steady state while their growth rates are varied over a wide range. It is also possible to determine the maximum cell yield in a given hydraulic situation using a continuous-flow culture system. Microbiologists have long utilized the chemostat for heterotrophic growth kinetics, and the theory and mathematics have been reviewed by Málek and Fencl (1966). The use of continuous culture methods for algal growth kinetics was introduced by Pipes and Koutsoyannis (1962), and a review of algastat theory is given by Retovsky (1966) and Pearson (1969).

The most used type of chemostat or algastat is a flowthrough, mixed reactor. A cellular material balance can be written around the reactor to define the growth of any unicellular microorganism.

(input - output) + (growth - decay) = change

or

$$Qc'_o - Qc' + R'_g V - R'_d V = V \frac{dc'}{dt}$$
(1)

where Q = flow (ml/min), c' = concentration of cellular material (cells/ml), $R'_g =$ volumetric growth rate of microor-

ganism (cells ml/min), R'_d = volumetric decay rate of microorganisms (cells/ml/min), and V = volume of reactor (ml).

Chemostats or algastats are run at steady state as cells are washed out, thus dc'/dt = 0. In most cases the influent flow does not contain microorganisms, therefore $c'_o = 0$, and with the introduction of first-order rate experiments with specific growth rate constant, μ , and specific decay rate constant, σ_d , Equation 1 becomes:

 $O_{2}I = 2IV$

$$-Qc + \mu c v - \sigma_d c v = 0$$

or

$$\mu - \sigma_d = Q/V = 1/\tau \tag{3}$$

(2)

where μ and σ_d are in min⁻¹ and τ = hydraulic residence time, min.

Equation 3 is the basic relationship describing an algastat operating at steady state, and has proved useful for evaluating phytoplankton growth kinetics as a function of nutrient enrichment. The data derived from such experiments have helped elucidate the response of phytoplankton in systems fertilized by domestic sewage. The problem of productivity stimulation owing to nutrient enrichment is not limited to the phytoplankton, however, as recent data have shown that benthic macrophytes are also stimulated by nutrient fertilization (Waite and Gregory, 1969). It has also been shown that benthic plants may have a higher productivity per unit area than the phytoplankton (Waite and Mitchell, 1971).

Methods for measuring primary production of aquatic plants are still inaccurate and poorly developed. Several methods have been proposed for in situ determinations, including oxygen evolution, ¹⁴CO₂ uptake, and biomass changes (Kanwisher, 1966; Wetzel, 1964; Westlake, 1965). Because of the size and growth complexity of the plants, laboratory culturing is difficult; thus, reliable growth rate relationships have not been well developed.

We have investigated the possibility of utilizing a continuous-flow stirred reactor for growth rate determinations of benthic macrophytes. This is a novel case as there is no washout of cells, thus Vdc'/dt in Equation 1 does not equal zero. The purpose of this study was to derive a simple relationship that would reliably model the growth response of benthic plants in a reactor using oxygen production. It has been found that in addition to obtaining the growth rate, the stoichiometric ratio of oxygen produced to biomass produced can also be evaluated provided measurable increases in biomass occur over the duration of an experiment, even though only the final biomass is measured.

Theory

A mass balance of oxygen can be written for the reactor in the same manner as a cellular balance was written in Equation 1.

$$Qc_o - Qc + R_g m - R_d m = V dc/dt \tag{4}$$

where Q = flow-through reactor (ml/hr), c = concentration of dissolved oxygen in reactor (mg O₂/ml), $R_g =$ specific algal growth rate in the light reactor (mg O₂/mg/hr), $R_d =$ specific

¹ Present address, Dept. of Civil Engineering, University of Miami, Coral Gables, Fla. 33124.

² To whom correspondence should be addressed.

algal decay rate (respiration) in the dark reactor (mg $O_2/mg/hr$), m = algal mass (mg), and V = volume of reactor (ml).

The plant material was fastened in the reactor, thus algal material accumulated as the plant grew during the experiment. Figure 1 shows the conceptual difference between a reactor running at steady state—i.e., washout of material, and one with an increasing biomass. It can be seen that the slope for the nonsteady reactor data should be proportional to the rate of biomass increase in the reactor.

Initially it was assumed that the increase in biomass in the light reactor would follow a first-order relationship:

$$dm/dt = (k_L R_g - k_d R_d)m \tag{5}$$

where k_L = stoichiometric growth constant (mg/mg O₂), and k_d = stoichiometric decay constant (mg/mg O₂) thus

$$m = m_0 e^{(k_L R_g - k_d R_d)t} \tag{6}$$

Realizing the oxygen increase in the light reactor represents the net productivity, Equation 4 may be rewritten as:

$$Qc_o - Qc + (R_g - R_d)m_o e^{(k_g R_g - k_d R_d)t} = V dc/dt \qquad (7)$$

Taking $c = c_0$ at t = 0, this equation can be solved to yield:

$$c/c_o - 1 = \frac{(R_g - R_d)m_o\tau/Vc_o}{1 + (k_L R_g - k_d R_d)\tau} \left[e^{(k_L R_g - k_d R_d)t} - e^{-\frac{t}{\tau}} \right]$$
(8)

If we assume there will be only a small amount of growth in the reactor—i.e. $(k_L R_g \tau - k_d R_d \tau)t/\tau \ll 1$, then exp $(k_L R_g - k_d R_d)t/\tau$ can be replaced by $1 + (k_L R_g - k_d R_d)t/\tau$, and Equation 8 simplifies to

$$\frac{c/c_o - 1}{1 - e^{-t/\tau}} = \left[\frac{(R_g - R_d)m_o\tau/Vc_o}{1 + (k_L R_g - k_d R_d)\tau}\right] + (k_L R_g - k_d R_d)\tau \left[\frac{(R_g - R_d)m_o\tau/Vc_o}{1 + (k_L R_g - k_d R_d)\tau}\right] \left[\frac{t/\tau}{1 - e^{-t/\tau}}\right]$$
(9)

Equation 9 is seen to be linear in form so that one can plot

$$\left[\frac{c/c_o-1}{1-e^{-t/\tau}}\right] \text{vs.} \left[\frac{t/\tau}{1-e^{-t/\tau}}\right]$$

and obtain a slope

$$(k_L R_g - k_d R_d) \tau \left[\frac{(R_g - R_d) m_o \tau / V c_o}{1 + (k_L R_g - k_d R_d) \tau} \right]$$

and an intercept

$$\left[\frac{(R_g - R_d)m_o\tau/Vc_o}{1 + (k_L R_g - k_d R_d)\tau}\right]$$

When the slope is divided by the intercept, a value for $(k_L R_q - k_d R_d)\tau$ is obtained, thus m_q may be readily calculated from Equation 6, and values for k_L and R_q can be determined by first evaluating k_d and R_d from the dark reactor and assuming the latter are unchanged between light and dark. The relationship for the dark reactor is similar to the light chamber:

$$-dm/dt = k_d R_d m \tag{10}$$

where k_d = stoichiometric decay constant (mg/mg O₂) and R_d = decay rate of the plant (mg O₂/mg/hr) and

$$Qc_o - Qc - k_d R_d m = V dc/dt \tag{11}$$

Making the same assumptions as those leading to Equation 9, Equation 11 may be solved to yield:

$$\begin{bmatrix} \frac{1-c/c_o}{1-e^{-t/\tau}} \end{bmatrix} = \begin{bmatrix} \frac{R_d m_o \tau/V c_o}{1-k_d R_d \tau} \end{bmatrix} - (k_d R_d \tau) \begin{bmatrix} \frac{R_d m_o \tau/V c_o}{1-k_d R_d \tau} \end{bmatrix} \begin{bmatrix} \frac{t/\tau}{1-e^{-t/\tau}} \end{bmatrix}$$
(12)

This is then treated by plotting in the same manner as Equation 9 to obtain values for k_d and R_d .

Methods and Materials

The benthic macrophyte selected to test our model was *Ulva lactuca* (sea lettuce). This particular plant was chosen because it was easy to handle in the laboratory, and its growth characteristics had been previously studied by Waite and Mitchell (1972). To optimize the reactor design, very shallow chambers were fabricated by cutting 2-in. slices from 12-in. diam polyvinyl chloride tubing, yielding a reactor volume of 4.08 liters. The top of the light chamber was made of $^{1}/_{4}$ -in. thick opaque vinyl plastic. The tops were bolted to the reactor walls to allow removal for filling the chambers with algae. The two reactors were mounted in a $36 \times 20 \times 4$ -in. polyvinyl chloride box, which served as a water bath in which the reactors were submerged. Tap water was circulated through the water bath at a velocity sufficient to maintain a temperature of 16°C inside the reactors.

Samples of algae were fastened to a section of nylon screening which was mounted about mid depth in the reactor. Each reactor was mixed with two magnetic spinbars, and lighting was provided by four 40-W Sylvania Cool White fluorescent lights which gave a visible light intensity of 480 ft-candles incident on the algae. Seawater enriched with ammonia nitrogen and phosphate phosphorus was pumped through the reactors using a Sigmamotor Model T8 directdrive pump.

Oxygen production was used as a monitor of algal response in our experiments, as it could be recorded electronically with dissolved oxygen electrodes. Using our specifications, the Harvard University Electronic Design Center designed and fabricated a four-channel constant-readout dissolved oxygen meter. Incorporated with the meter was a Rustrak Model 288/F109A recorder and four YSI-4004 Clark oxygen probes. This apparatus allowed for dissolved oxygen measurements in both chambers and influent lines simultaneously (Figure 2).

The seawater entering the reactors during the experiments was saturated with oxygen. It was expected that the oxygen concentration would decrease in the dark chamber and increase in the light chamber. To prevent bubble formation in the supersaturated seawater, a deoxygenation chamber was built into the feed lines of the light chamber. The influent

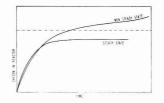
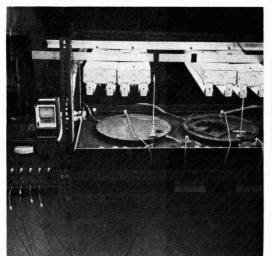


Figure 1. Conceptual difference between an algastat running at steady state and nonsteady state owing to no cell washout



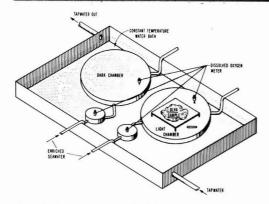


Figure 2. Reactor apparatus utilized in growth rate and stoichiometric analyses

solution of enriched seawater was fed through a 4-in.-diam \times 4-in.-high acrylic plastic cylinder. A gas mixture of 15% oxygen and 85% nitrogen was bubbled through the cylinder, thus lowering the oxygen concentration by 25%. The gas exchange cylinder was mixed with a magnetic spinbar, and a $^{a}/_{a}$ -in.-diam standpipe on top of the cylinder allowed the gases to escape, while maintaining the correct pressure head.

The samples of *Ulva* to be analyzed were acclimated for 24 hr in flasks containing nitrogen and phosphorus concentrations similar to those to be used in the experiment. The plants were then transferred to the reactors, equilibrated in the water bath and analyzed. Flow rates were varied from about 40 ml/min to 80 ml/min and length of experimentation varied between 6 and 12 hr. At the end of the analysis, the samples were removed from the reactors, washed, and dried at 80°C overnight. The samples were then weighed and this value was used as the algal mass (m).

Results and Discussion

Initial experiments were run to determine if the predicted model would fit the observed laboratory data obtained from nutrient enrichment assays. Figure 3 shows typical plant responses to enrichment with optimal and suboptimal nutrients. It can be seen that no increase in oxygen concentration could

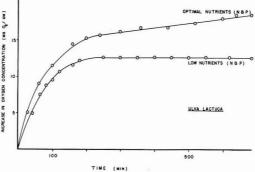


Figure 3. Typical photosynthesis data from plants growing with optimal nutrients and with low nutrients

be noted for the duration of our experiment when there was only a small addition of nutrients. The plants were apparently growing at a rate too slow to obtain the biomass change by a 12-hr analysis with oxygen electrodes. Figure 3 also shows the photosynthetic response of a plant at optimal nutrients and in this case, a discernible increase in oxygen concentration is observed. Our testing showed that the plants had to exhibit a high growth rate before any increase in oxygen was observed in the reactor after two or three detention periods. Growth rates can be determined from our reactor on a steady-state basis, but the stoichiometric constant k_L can be determined only when an apparent slope is realized.

Respiration. The respiration rate measured in the dark chamber represents oxygen utilization by both the plant and its associated microflora. It is impossible to differentiate between the contribution from the bacteria and algae as the plant will not grow normally in an axenic state. Figure 4 shows some typical data from the dark chamber. It can be seen that the decrease in oxygen is small and approaches a steady state. Because the decay of algal material in the reactor is small, the value of $k_d R_d$ is also small and can thus be neglected in Equation 10. While the slope from Equation 12 is immeasurably small, the intercept is still readily discernible to give R_d .

Our model assumes there will be no appreciable variation in nutrients throughout the experiments. Figure 5 shows the concentration of nitrate and phosphate over four detention times for various initial concentrations. While there appears to be a slight decreasing trend in the lower nutrient concentration data, the higher concentrations (greater than 1 mg/l.) show no systematic trend. Stoichiometry would suggest that the nitrogen should be utilized 10–15 times faster than the phosphorus, but this uptake ratio was not observed in our experiments. From these data we conclude that nitrogen and phosphorus were not changing significantly in the reactors during the experiments.

Assays that showed high growth rates were analyzed in our model. Several environmental factors were manipulated to observe possible variations in growth rates and stoichiometric constants. Figure 6 shows typical data obtained from the light reactor. An error analysis was performed on each sample by analyzing the maximum variation in slope and intercept. It can be seen from Equation 12 that the slope of Figure 6 is the main determinate of k_L while the intercept reflects the growth rate R_g . The error analysis showed that even with a

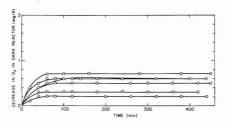


Figure 4. Respiration data in dark chamber (values are due to both plant respiration and heterotrophic activity of the associated microflora)

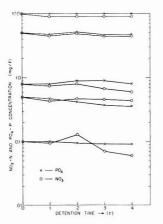


Figure 5. Nitrate and phosphate concentrations in the light reactor with time at different initial values

variation in slope, the intercept remained comparatively constant.

Figure 6 also shows a short period immediately after startup, during which the normalized oxygen concentrations fall significantly below the linear dependence expected from Equation 9 and observed at longer times. While it is not certain, this systematic discrepancy might be a result of inherent error in the initial concentration c_0 or possibly in the startup time. Such effects would be pronounced at short times when quotients of small numbers must be evaluated to obtain the coordinates of Figure 6. At long times these effects should be unimportant.

Some values of growth rate R_g , decay rate R_d , and stoichiometric constant $1/k_L$ obtained in our experiments are presented in Table I. The most surprising information in Table I is the values obtained for $1/k_L$. It can be seen that $1/k_L$

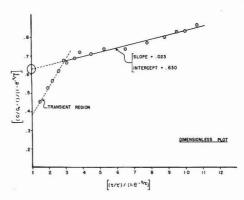


Figure 6. Dimensionless plot of reactor data showing a typical slope and intercept (note transient region for small times)

Table I.	Giowui Kates an	a Stotemomente Gro	the Constants for Orea	laciaca Onuci vario	us Nutrient and L	agint Conditions
				Growth rate (R_g) ,	Decay rate (R_d) ,	(mg O ₂ /mg)
Sample	Plant condition	Nutrients, mg/l.	Lights, ft-candles	(Mg O ₂ /g/hr)	(mg O ₂ /g/hr)	1/k _L
	Fresh	0.05 NH ₄ -N	Fluorescent			
1			480	5.4 ± 1.2	3.1	0.04 ± 0.01
		0.05 PO ₄ -P				
	Fresh	0.2 NH ₄ -N	Fluorescent			
2			480	9.4 ± 3.0	2.6	0.07 ± 0.02
		0.2 PO ₄ -P				
	Old	0.5 NH ₄ -N	Fluorescent			
3			480	5.7 ± 0.3	2.0	0.12 ± 0.03
		0.5 PO ₄ -P				
	Old	0.5 NH ₄ -N	Fluorescent			
4			480	4.2 ± 0.2	3.6	0.13 ± 0.01
		0.5 PO ₄ -P				
	Fresh	0.5 NH4-N	Fluorescent			
5			480	13.2 ± 1.8	2.9	0.08 ± 0.02
		0.5 PO ₄ -P				
	Fresh	0.7 NH ₄ -N	Fluorescent			
6			480	26.5 ± 1.0	4.1	$0.08 \pm$
		0.7 PO ₄ -P				
	Fresh	0.5 NH ₄ -N	Incandescent			
7			1000	11.2 ± 0.5	3.5	0.35 ± 0.7
		0.5 PO ₄ -P				
	Fresh	1.0 NH4-N	Incandescent			
8			1000	4.0 ± 0.1	3.2	0.11 ± 0.03
		1.0 PO ₄ -P				
		1 -				

Table I. Growth Rates and Stoichiometric Growth Constants for Ulva lactuca Under Various Nutrient and Light Conditions

remains substantially unchanged as the growth rate varies over a wide range, but its absolute value is a factor of 5-10 times lower than predicted from carbohydrate synthesis (Odum, 1971). If the plants were synthesizing material that was equivalent to carbohydrate-i.e., the oxidation state of carbon is equal to zero-then

linh 4

$$CO_2 + H_2O \xrightarrow{\text{nglit}} CH_2O + O_2$$
 (13)

and the quotient $1/k_L$ (gram O₂/gram mass) would be 1.07. It is generally assumed that algae reduce the oxidation state of carbon from +4 (CO₂) to a lower level during metabolism. The actual compounds synthesized and stored by algae depend on the environmental conditions to which the plant has been acclimated. Brandt and Rabin (1920) analyzed the composition of various plants and found that Ulva grown under their test conditions had a CHO ratio of 100/175/65. This implies that the oxidation state of carbon was about -0.4, or material more reduced than carbohydrate was being synthesized by the plant. Our data show that algal material with carbon oxidation states of +1 or +2 was being synthesized by Ulva under our test conditions. It is possible that the use of nitrate nitrogen instead of ammonia nitrogen as an enrichment would have changed the value of $1/k_L$. We did not attempt experiments with nitrate enrichment because of the complexity of growth interactions when ammonia is already present in the seawater.

The effect of different concentrations of ammonia and phosphate on growth rate and $1/k_L$ can be observed by comparing samples 1, 2, 5, 6, and 8. It can be seen that as the ammonia and phosphate concentration is increased from 0.05 mg/l. to 0.7 mg/l. the growth rate increases, but $1/k_L$ remains constant at about 0.07. When the concentration of nutrients is increased to 1.0 mg/l. nitrogen and phosphorus (sample 8), the growth rate decreases and $1/k_L$ increases slightly. Sample 8 was run with a higher light intensity, but the growth rate was much less than observed with a lower light intensity and nutrient concentration. This result agrees with findings of Waite and Mitchell (1972) that ammonia nitrogen concentrations above 0.8 mg/l. inhibit photosynthesis in Ulva regardless of other environmental influences.

Aging of test material was investigated in samples 3 and 4. These plants were left in culture flasks for two weeks before testing and their production was compared to fresh samples. Comparing samples 3 and 4 to sample 5, it can be seen that the growth rate of the plants was hindered by the long acclimation, but the value for $1/k_L$ was higher indicating a build-up of more reduced compounds in the algae.

The effect of higher light intensity was also investigated in our experiments. Samples 7 and 8 were run at 1000 ft-candles or approximately twice the intensity used in samples 1-6. By comparing sample 7 with sample 5, it can be seen that the increase in light intensity did not alter the growth rate significantly, but the stoichiometric constant increased by a factor of four. This finding is not easily explained because presumably with a high light intensity the plant would not be manufacturing appreciable amounts of chlorophyll, as the synthesis of chlorophyll would tend to increase $1/k_L$:

where $1/k_L = 2160/892 = 2.24 \text{ mg O}_2/\text{mg}$.

Table I also shows that the decay rate, R_d , does not fluctuate appreciably. The values only vary from 2.0 to 4.1 while the growth rate varies from 4.0 to 26.5. As was mentioned earlier, the decay rate reflects both the plant respiration and heterotrophic activity of the associated microflora. Thus, it appears that even if the rate of plant respiration is varying with growth rate, the variation is dwarfed by the larger contribution from the microflora.

Our laboratory data have shown that it is possible to analyze the growth rate and stoichiometric ratio of mass synthesis of benthic plants in a nonsteady state reactor. Because of the nature of the reactor, the increase in plant biomass can be calculated from the slope of the oxygen vs. time curve. We have also shown that the ratio of oxygen production to algal mass synthesis is relatively independent of nutrient concentration and growth rate, but is affected by light intensity. The most interesting observation is that the value of $1/k_L$ is almost a factor of 10 lower than predicted. It appears that Ulva is capable of producing compounds with carbon oxidation states of +1 or +2, thus estimates of biomass production may be in error when the average material is assumed to be carbohydrate.

Although untested, it is possible that this method may be applied to growth kinetic studies of phytoplankton. By preventing washout of cellular material, our analysis would be applicable, and a stoichiometric coefficient could be calculated as well as growth rate. This method may also prove useful for assessing low growth rates of phytoplankton, because no washout would occur as happens in a conventional algastat when the doubling time is less than the detention time.

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Aerosol Filtration with Slip Flow

Richard D. Chmielewski and Simon L. Goren¹

Department of Chemical Engineering, University of California, Berkeley, Calif. 94720

New measurements of the pressure drop and filter efficiency when slip at the fiber surface becomes significant are presented. With slip the filter efficiency increases and pressure drop decreases. These data are adequately predicted by a model extending the theory of Stechkina and Fuchs (1966) and Friedlander (1958, 1967) to the slip regime. The model, however, fails to predict certain data of other authors.

echanisms of particle capture in aerosol filtration by fibrous media include impaction, interception, and Brownian diffusion. For each of these mechanisms, the efficiency of particle capture by individual fibers, and by the mat as a whole, increases as the fiber radius decreases. This fact has encouraged development of filters fabricated from fine fibers. It is now known that if the fibers are reduced to a size comparable to the mean free path of the gas (6.45 \times 10⁻⁶ cm for air at normal temperature and pressure), the efficiency of particle removal is further and significantly increased while at the same time the pressure drop requirement for a fixed degree of removal is reduced. The explanation of these trends is that an object in the slip flow regime gives less disturbance to the oncoming plug flow so that the streamlines remain straighter and closer to the object than for continuum flow. Since there is less disturbance to the flow field, the frictional drag on the object is reduced and thereby the pressure drop lowered. Since the fluid streamlines are straighter, there is less hydrodynamic force tending to divert oncoming particles from collision with the collector when impaction or interception is dominant and the fiber efficiency is thereby increased; when diffusion or interception is dominant, the larger velocity near the surface of the collector results in a smaller diffusion boundary layer giving an increased deposition rate.

The present paper is an experimental and theoretical investigation of the effects of slip on low-speed filtration efficiency and pressure drop in fibrous filters. A model is proposed which successfully correlates our own data but underestimates the efficiencies reported by Stern et al. (1960). It is hoped that by pointing out the advantages of working in the slip flow regime and by providing a scheme for correlating pressure drop and filter efficiency our work will stimulate the development of submicron fibers and their incorporation into filter media.

Theory

Spielman and Goren (1968) examined slip flow past a cylindrical fiber immersed within a fibrous bed. The basis of their calculation was Brinkman's model in which the influence of neighboring fibers is accounted for as a body force proportional to the local velocity of the fluid, and slip effects enter through the boundary condition on the tangential velocity at the fiber surface. They derived the following

formula for the dimensionless pressure drop as a function of the fraction solids and the modified Knudsen number:

$$\frac{\Delta p a_F^2}{4 \alpha \mu U \Delta L} = \Phi(\alpha, \mathrm{Kn}') \tag{1}$$

The function $\Phi(\alpha, Kn')$ was computed and displayed graphically in their paper. For the present paper, it is useful to factor $\Phi(\alpha, Kn')$ into two terms

$$\frac{\Delta p a_F^2}{4 \alpha \mu U \Delta L} = \Phi(\alpha, \mathbf{Kn'}) \equiv \Phi(\alpha, 0) / C(\alpha, \mathbf{Kn'})$$
(2)

where $\Phi(\alpha, 0)$ gives the pressure drop that would occur in the complete absence of slip and $C(\alpha, Kn')$ accounts for the slip. In the range of fraction solids usually encountered in fibrous filters, $0.03 \leq \alpha \leq 0.1$, $\Phi(\alpha, 0)$ is in good agreement with Davies' empirical equation $\Phi(\alpha,0) = 4 \alpha^{1/2} (1 + 56 \alpha^3)$ although an even better representation of the calculated results at higher α is provided by $\Phi(\alpha, 0) = 4 \alpha^{1/2} (1 + 175 \alpha^3)$. The utility of factoring $\Phi(\alpha, Kn')$ rests partly on the observation that for $Kn' \leq 0.4$, $C(\alpha, Kn')$ is nearly linear in Kn' and is not strongly dependent on α ; this is illustrated in Figure 1 where $C(\alpha, Kn')$ is plotted against the modified Knudsen number for $\alpha = 0.032, 0.061, \text{ and } 0.090$. A second benefit is that pressure drop data for a given filter can be compared on a relative basis, as was done by Pich (1971), so that uncertainties in the fraction solids, filter thickness, and velocity largely cancel.

As part of their calculation, Spielman and Goren (1968) derived an expression for the stream function for flow past a cylinder. Correct to second order in the power series ex-

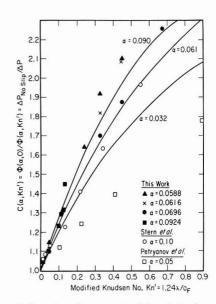


Figure 1. Comparison of measurements of the reduction in pressure drop due to slip effects for flow-through fibrous media with that predicted by the model of Spielman and Goren (1968)

¹ To whom correspondence should be addressed.

pansion in the distance from the fiber surface, their stream function is

$$\psi = 2 A U a_F \sin \theta \left[\left(\frac{y}{a_F} \right)^2 + B \left(\frac{y}{a_F} \right) \right]$$
(3)

where

$$2 A = (1 - 2 \alpha) \Phi(\alpha, \mathrm{Kn}') \tag{4}$$

and

$$B = 2 \operatorname{Kn}'/(1 + \operatorname{Kn}')$$

The stream function for slip flow derived by Pich (1966a) using the cell model of Kuwabara and that derived by Natanson (1962) for flow past an isolated fiber can be cast into a form identical to Equation 3 except that the coefficient A takes on different values.

$$\frac{4 (1 - \alpha)(1 + Kn')}{(\alpha^2 - 1 - 2 \ln \alpha)(1 + 2 Kn') - 2 + 4 \alpha - 2 \alpha^2}$$
(5)

Natanson: 2 $A = \frac{1 + Kn'}{(2.50 - \ln Re)(1 + 2 Kn') - 0.50}$

We shall confine our attention to the capture mechanisms of interception and Brownian diffusion. When the particles are sufficiently small that Brownian diffusion is important, but the Peclet number is sufficiently large that the concentration boundary layer is confined to a thin zone adjacent to the fiber where the two-term expansion of Equation 3 is valid, the particle concentration satisfies the following approximate convective diffusion equation:

$$\sin \theta \left[2 \left(\frac{y}{a_F} \right) + B \right] \frac{\partial c}{\partial \theta} - \cos \theta \left[\left(\frac{y}{a_F} \right)^2 + B \left(\frac{y}{a_F} \right) \right] \frac{\partial c}{\partial y/a_F} \cong \frac{1}{A \operatorname{Pe}} \frac{\partial^2 c}{\partial (y/a_F)^2}$$
(6)

with the boundary conditions at

$$y/a_F = a_p/a_F = R, c = 0$$
 (7)

and at

$$y/a_F = \infty, c = c_{\infty}$$

The capture efficiency is computed from the rate of particle deposition on the fiber surface

$$\eta = \frac{\int_0^{\pi} D\left(\frac{\partial c}{\partial y}\right)_{y=a_p} a_F d\theta}{U a_F C_{\infty}}$$
(8)

With B = 0 we recover the set of equations previously investigated by Friedlander (1967) and Stechkina and Fuchs (1966) for no slip. Inspection of the above set of equations for nonzero B in the manner used by Friedlander (1967) shows that the fiber efficiency is a function of two dimensionless groups:

$$\eta R P e = f^n (2 \ A R^3 P e, \ B/R) \tag{9}$$

For the complete absence of slip, B = 0, Stechkina and Fuchs (1966) numerically solved the convective diffusion boundary layer equation. Their results for the limit of small interception number and large Peclet number in the absence of slip can be accurately represented in the formula

$$\eta R Pe = 2 A R^{3} Pe + 1.25 (2 A R^{3} Pe)^{1/2} + 2.9 (2 A R^{3} Pe)^{1/3} (10)$$

When 2 $AR^{a}Pe \gg 1$ so that the first term on the right-hand side of Equation 10 dominates, the major mechanism of capture is interception; when 2 $AR^{a}Pe \ll 1$ so that the last term on the right-hand side of Equation 10 dominates, the major mechanism of capture is Brownian diffusion of "point" particles. For 2 $AR^{a}Pe$ of order unity, both mechanisms are comparable.

The approximation of no slip for the capture of "point" particles by Brownian diffusion will be valid when the diffusion boundary layer, δ , extends sufficiently far into the fluid so that the quadratic term in the stream function dominates the linear term (but the cubic term is still unimportant). This will occur when $(\delta/a_F) \approx (A \operatorname{Pe})^{-1/3} \gg \operatorname{Kn}'$ or

$$2 AR^{3}Pe \cdot (Kn'/R)^{3} \ll 1$$
 (11)

Since 2 *AR*³Pe must be small for Brownian diffusion to dominate, slip effects can be neglected in the diffusion zone if the modified Knudsen number is not much larger than the interception number. Pich (1966) has extended the diffusion calculation to give a first-order correction when 2 *AR*³Pe· (Kn¹/*R*)³ is small but not negligible.

For sufficiently small particles, however, the modified Knudsen number, even though small, may be much larger than the interception number so that $2 AR^{3}Pe(Kn'/R)^{3} \gg 1$ even though $2 AR^{3}Pe \ll 1$. Under these conditions, Brownian diffusion of "point" particles is still the dominant mechanism, but the diffusion boundary layer does not extend beyond the linear zone of the stream function. With the neglect of the quadratic term in Equation 3, the convective diffusion equation easily solved approximately (Chmielewski, 1972 to yield the following expression for the Brownian capture of "point" particles with very large slip effects:

$$\eta R P e = 2.26 (2 \ A R^3 P e \cdot B/R)^{1/2}$$
(12)

for 2 $AR^{3}Pe \ll 1$, $B/R \gg 1$, and 2 $AR^{3}Pe \cdot (B/R)^{3} \gg 1$. Thus, Equation 12 and

$$nRPe = 2.9 (2 \ AR^{3}Pe)^{1/3}$$
 (13)

for $2 AR^{3}Pe \ll 1$, $B/R \ll 1$, and $2 AR^{3}Pe(B/R)^{3} \ll 1$ are expected to be the limiting forms for the capture of "point" particles with large and negligible slip, respectively. For intermediate values of $2 AR^{3}Pe(B/R)^{3}$ the diffusion equation must be solved numerically. We have estimated the increased efficiency due to slip (including interception due to the finite size of the particles) relative to the efficiency in the complete absence of slip by solving the diffusion equation near the forward stagnation point. The resulting expression is

$$\frac{\frac{\eta}{\eta_{0}}}{\int_{1}^{\infty} \exp\{-A_{0}R^{3}\operatorname{Pe}[^{1}/_{3}(Y^{3}-1)]\}dY}{\int_{1}^{\infty} \exp\{-AR^{3}\operatorname{Pe}[^{1}/_{3}(Y^{3}-1)+\frac{1}{2}B/R(Y^{2}-1)]\}dY}$$
(14)

Some results for small 2 $A_{\alpha}R^{3}$ Pe are shown in Figure 2. In the above, A_{α} is the hydrodynamic parameter of Equation 4 for no slip—i.e., $A_{\alpha} = (1 - 2 \alpha) \Phi(\alpha, 0)$.

At the other extreme, $2 AR^{3}Pe \gg 1$, one may immediately write down the fiber efficiency for interception of nondiffusing particles from the relation of the stream function to the volumetric flow rate:

1102 Environmental Science & Technology

$$\eta = \frac{\psi\left(\theta = \frac{\pi}{2}, y = a_p\right)}{Ua_F} = 2 \ AR^2(1 + B/R)$$
(15)

or

$$\eta R P e = 2 A R^{3} P e \{1 + B/R\}$$
(16)

and

$$\eta/\eta_o = \frac{A}{A_o} \left\{ 1 + B/R \right\} \tag{17}$$

Figure 2 shows the significant increase in fiber efficiency possible when slip at the fiber surface becomes important. Here the relative fiber efficiency, η/η_0 , calculated from Equation 17 for 2 $A_o R^a Pe \gg 1$, from Equation 14 for 2 $A_o R^a Pe < 1$ and interpolated in the intermediate zone is plotted against 2 $A_o R^a Pe$ with B/R as a parameter; for the purpose of constructing these curves Kn' is taken so small that $A \approx A_0$. It is seen that the effects of slip are more pronounced in the interception zone where increases in efficiency of more than an order of magnitude are possible.

For the purpose of correlating experimental filtration efficiency data in the slip flow regime and comparing it with data for continuum flow, we found it most convenient to plot ηRPe against 2 $AR^{3}Pe(1 + B/R)$ with B/R as a parameter. This is because the interception zone [see Equation 16] is accounted for exactly, and in the diffusion zone for any fixed finite value of B/R the curves all approach the same asymptote, Equation 13, as 2 $AR^{3}Pe$ becomes sufficiently small. When plotted this way the various curves largely coalesce to a single curve for the range of variables of usual concern as can be seen in Figure 3. With B = 0 this is the same curve found by Spielman and Goren (1968) to give good agreement with the semiempirical equation of Friedlander (1958) and the continuum regime data of Chen (1955), Ramskill and Anderson (1951), and Thomas (1953).

Experimental Methods

An experimental program was undertaken to measure pressure drop and filter efficiency in fibrous filters under conditions where slip at the fiber surfaces is important. Filters were made from low-twist yarns of uniform diameter glass fibers supplied by Owens Corning, and their fabrication and characterization were determined by the methods described by Spielman and Goren (1972). Fibers of nominal diameter 3.5 μ and 12 μ were used. At atmospheric pressure the mean free path of air is 0.064 μ so that the modified Knudsen numbers, $Kn' = c_m \lambda / a_F$, for the two fibers are 0.046 and 0.013. To increase effects of slip, measurements were made at reduced pressures, the lowest pressure being 5 cm Hg for which the modified Knudsen numbers increase to 0.70 and 0.20, respectively. The thickness of the mats varied from about 0.1 to 0.3 cm. Mats fabricated from the 3.5-µ-diam fibers had fraction solids between 0.05 and 0.07 and those fabricated from the $12-\mu$ -diam fiber had fraction solids of about 0.09.

The aerosol particles used were Dow monodisperse polystyrene latex particles of diameter 0.126, 0.264, 0.357, 0.557, and 0.796 μ . The aerosol was formed from liquid suspensions by atomization and evaporation in a Royco Aerosol Generator, Model 255. To minimize the role of electrical forces, the aerosol was mixed with a stream of bipolar ions to neutralize the electrical charge on the particles. The bipolar ionizer was constructed according to the specifications of Whitby and Peterson (1965). The aerosol was then passed between parallel copper plates 36 cm long, spaced 4.5 cm apart, and maintained

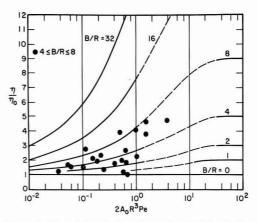


Figure 2. Theoretical estimate and experimental data showing increase in fiber efficiency due to slip for the Brownian diffusion and interception mechanisms; for data shown, B/R = 2 Kn'/R(1 + Kn') varies between 4 and 8

at a potential difference of several kilovolts to remove any remaining highly charged particles (particles of large mobility in an electric field).

The filters were mounted in a holder with taps for pressure measurement and the withdrawal of samples from the upstream and downstream sides. The pressure drop was measured with a Decker Corp. Model 308 differential pressure gage with a transducer capable of detecting pressure differences of 0.0042 cm Hg. The aerosol concentration was measured with a Sinclair-Phoenix Photometer, Model JM-3000-AL. As the photometer measures scattering from a cloud of particles, it is necessary to assume the aerosol is monodisperse and of known diameter. To the extent that small secondary particles formed in the atomization process were present, our interpretation of the photometer signals may be in error. However, the consistency of the results suggests that such secondary particles were not generated in significant numbers to influence the photometer measurements or that they were largely removed in the electrostatic precipitator. The temperature, humidity, and absolute pressure were measured at the inlet to the filter, and the flow rate measured with a rotameter.

Experimental Results

Pressure Drop. A number of runs were made with particle free air to measure the pressure drop at reduced pressures. At each pressure the pressure drop was observed to vary linearly with velocity as reported by Stern et al. (1960). From the best slope of each line $\Phi(\alpha, \mathbf{Kn}') \equiv \Delta p a^2 _{F}/4 \alpha_{\mu} U \Delta L$ was computed and compared with the theoretical value calculated from the model of Spielman and Goren (1968). The agreement was very good. For the 3.5- μ -diam fibers, three mats with a total of 11 runs (11 slopes), $\Phi_{expt1}/\Phi_{theory}$ ranged from 0.85 to 1.20 and averaged 0.97; for the 12- μ -diam fibers, one mat with a total of five runs, $\Phi_{expt1}/\Phi_{theory}$ ranged from 1.05 to 1.15 and averaged 1.12.

An experimental value of $\Phi(\alpha,0)$ was determined by extrapolation of the experimental values of $\Phi(\alpha,Kn')$ to zero Knudsen number. Then $C = \Phi(\alpha,0)/\Phi(\alpha,Kn')$ was plotted against Kn'. The results, displayed in Figure 1, show good agreement with the theory of Spielman and Goren for Kn' \leq 0.2, but the theory underestimates C by about 15% at the

higher Knudsen numbers used. At Kn' \cong 0.4, the pressure drop is about half of what it would be in the complete absence of slip.

Also shown in Figure 1 are the pressure drop data of Stern et al. (1960) for a mat of 17-µ-diam fibers and 0.1 fraction solids. The theoretical values of C are larger by about 15%than those observed by these authors. Moreover, $\Phi_{exptl}/$ Φ_{theory} varied from 1.31 to 1.57 averaging 1.44—i.e., the observed pressure drop is about 44% larger than the expected value. This discrepancy might be explained by a small underestimate of the fraction solids ($\alpha = 0.12$ instead of $\alpha = 0.10$) for the filters used by Stern et al. Also shown in Figure 1 are the data of Petryanov et al. (1965) for a filter of 1.5-µ-diam fibers and 0.05 fraction solids; these data were taken from the paper of Pich (1971). The theory of Spielman and Goren considerably overestimates C for these data. In contrast, Pich found that the hydrodynamic model developed by him gave good agreement with these data. However, this agreement is fortuitous since the Knudsen numbers for the experiments of Petryanov et al. are too large to justify the approximation of Equation 2 by Equation 3 in Pich's paper (1971) and his more exact result, based on Equation 5 of this paper, also considerably overestimates the observed values of C.

It is difficult to compare the pressure drop data of Wheat (1963) and Clarenburg and Van der Wal (1966) with the theory because of the nonuniform diameter fibers used by these authors.

Fiber Efficiency. Filtration performance was measured at reduced pressures for five mats made from 3.5- μ -diam fibers and one mat of 12- μ -diam fibers. The superficial velocity varied from about 1 to 16 cm/sec with a few runs as high as 200 cm/sec. The range of the important dimensionless groups covered in our experiments are as follows:

$$\begin{array}{l} 0.046 \leq Kn' \leq 0.70 \\ 0.01 \leq R \leq 0.23 \\ 600 \leq Pe \leq 1.3 \times 10^{\circ} \\ 0.0024 \leq 2 \ AR^{3}Pe \leq 230 \\ 0.37 \leq B/R \leq 31 \\ 0.42 \leq [2 \ AR^{3}Pe(1 + B/R)]^{1/3} \leq 10.4 \end{array}$$

In the above the Peclet number 2 $a_F U/D$, is based on the Brownian diffusion coefficient at the pressure of the experiment so that slip effects at the particle surface are accounted for even if slip at the fiber surface is negligible. Single fiber efficiencies were computed from the formula

$$\eta = \frac{\pi a_F \Delta \ln N}{2 \alpha \Delta L} \tag{18}$$

where N is a number proportional to the dust concentration given by the photometer reading. Calculated fiber efficiencies varied between 0.010 and 0.16. The dimensionless group ηR Pe suggested by the theory for simultaneous interception and Brownian diffusion varied from:

$$0.27 \leq \eta RPe \leq 1360$$

In Figure 2 we have plotted the ratio of the measured fiber efficiency to that predicted by Equation 10 for the complete absence of slip at the fiber surface vs. the group 2 $A_oR^{3}Pe$. For clarity, only the data for B/R between 4 and 8 are shown. As can be seen from the figure, about half the points fall within the band defined by the theoretical curves for B/R = 4 and 8. The data falling below this band had the smaller values of B/R. The theory, in the diffusion zone at least, while giving the correct trend and order of magnitude, somewhat

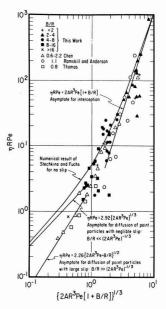


Figure 3. Correlation of the fiber efficiency due to Brownian diffusion and interception with slip at the fiber surface

overestimates the enhancement of the fiber efficiency due to slip.

Figure 3 shows our data plotted as ηRPe vs. [2 $AR^{3}Pe(1 +$ B/R)]^{1/3}. Also shown are the filtration data at atmospheric pressure of Chen (1955), Ramskill and Anderson (1951), and Thomas (1953). The theoretical curve of Stechkina and Fuchs (1966) for simultaneous interception and Brownian diffusion with no slip (B = 0) and the three asymptotes discussed in this paper are also shown. With the factor [1 +B/R]^{1/3} applied to the abscissa, most of our data fall toward the interception zone, $[2 AR^{3}Pe(1 + B/R)]^{1/3}$ greater than unity, where it is well correlated and adequately predicted by the theory. For $[2 AR^{3}Pe(1 + B/R)]^{1/3}$ less than unity, the data fall below the predicted values. Similar theoretical overestimates of filter efficiencies where slip is of minor importance are evident in the extensive data of Chen (1954) and Wong and Johnstone (1953) as shown in Figure 5 of the paper by Spielman and Goren (1968).

Experimental filter efficiencies of Stern et al. (1960) expected to be in the interception- and diffusion-controlled regimes and measured at reduced pressure where slip at the fiber surface is expected to play an important role were plotted as suggested in Figure 3 (but are not shown here). For $0.1 < [2 AR^3Pe(1 + B/R)]^{1/3} < 10$, the experimental values of $\eta RPe \approx$ 2.5 [2 $AR^3Pe(1 + B/R)]^{2/3}$: The efficiencies of these authors are from six- to tenfold greater than those measured by the present authors and by others working at atmospheric pressure. The explanation of this discrepancy is not known, but it is probably related to the pressure drops found by Stern et al. for their filters also being larger (by about 44%) than expected.

Nomenclature

- a_p = particle radius
- a_F = fiber radius
- A = hydrodynamic parameter depending on flow model

- Ao hydrodynamic parameter for zero Knudsen number
- B $\equiv 2 \text{ Kn'}/(1 + \text{Kn'})$
- *c* = particle concentration
- $C \equiv \Phi(\alpha, 0)/\Phi(\alpha, Kn')$, slip coefficient for fibers
- D = Brownian diffusion coefficient
- $Kn' \equiv$ $c_m \lambda / a_F$, modified Knudsen number; $c_m \approx 1.24$
- Pe $\equiv 2 a_F U/D$, Peclet number
- $R \equiv a_p/a_F$, interception number
- Re $\equiv 2 a_F U/\nu$, Reynolds number V
- = coordinate normal to fiber surface U superficial velocity
- fraction solids α
- $\Delta L =$ filter thickness
- $\Delta p =$ pressure drop across filter
- fiber efficiency = η
- θ = angular coordinate
- μ = viscosity
- Φ $\equiv \Delta p a_F^2 / 4 \alpha \mu U \Delta L$, dimensionless pressure drop
- ¥ = stream function

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X-ray Spectrometric Analysis of Air Pollution Dust

Connell L. Luke, Thomas Y. Kometani, John E. Kessler, and Thomas C. Loomis¹ Bell Laboratories, Murray Hill, N.J. 07974

John L. Bove² and Benjamin Nathanson

New York City Department of Air Resources, New York, N.Y. 10003

 Using X-ray fluorescence spectrometry, direct nondestructive analysis of airborne particulate matter for common metals can be performed rapidly on collection filters without need for chemical processing. Commercially available instrumentation has been used for the direct determination of Pb, Zn, Cu, Ni, Fe, Mn, and Co in samples collected on filter paper. Calibration is achieved by precipitating known amounts of these elements and collecting them on a Millipore filter disc which then serves as a reasonably permanent standard. Results obtained by this X-ray technique on typical samples collected by the New York City Department of Air Resources compare favorably with those obtained on the same samples after ashing.

articularly appealing features of the direct X-ray method are its rapidity and the ease with which it could be automated to handle large numbers of samples even where chemical processing facilities are not available.

Introduction

An important part of any program for monitoring air quality is the analysis of airborne particulate matter. Samples obtained by drawing appropriate volumes of air through suitable filters are usually analyzed by atomic absorption spectrometry (aas). The method involves a preliminary ashing to ensure that all of the elements to be determined are dissolved in the specimen presented to the spectrometer. This requirement makes the analysis expensive and-especially for large numbers of samples-somewhat cumbersome.

Direct X-ray fluorescence spectrometry has been proposed for the analysis of air particulate matter by Cares (1968), Luke (1968), and Leroux (1970). So far, however, there is no indication that X-ray methods have actually been employed in routine measurements. One reason may be that no serious evaluation of the range, scope, and reliability of an X-ray fluorescence method for particulate matter characterization has as yet been reported. The opportunity for just such an investigation arose when the New York City Department of Air Resources suggested a joint undertaking by that organization and the analytical group of Bell Laboratories.

The New York City people were in a position to supply representative samples and expertise regarding such matters

¹ To whom correspondence should be addressed.

² Present address, Chemical Engineering Department, The Cooper Union for the Advancement of Science and Art, 51 Astor Place, New York, N.Y. 10003.

as which elements need to be determined, precision and accuracy requirements, and techniques for sampling and sample handling. Bell Laboratories, on the other hand, had the equipment and personnel for developing and evaluating the actual methods of analysis.

Preliminary Considerations

Some previous unpublished work by Luke had established, at least semiquantitatively, that certain elements present in airborne particulate matter collected on filter paper could be determined by direct X-ray spectrometry or by Coprex analysis (Luke, 1968) following wet or dry ashing. He also demonstrated that the Coprex method is suitable for the determination of trace elements collected in aqueous solutions in standard air pollution impingers. The present investigation, then, was aimed principally toward ascertaining which elements could be determined by a direct method to the desired accuracy at the expected level of contamination.

New York City has been collecting daily samples of particulate matter on 8 \times 10-in. sheets of Schleicher and Schuell (S & S) No. 589 Green Ribbon filter paper in high-volume samplers at many locations throughout the city. Approximately 1500 m3 of air pass through each filter in the 24-hr period. These samples are then dry-ashed at 350-500°C and analyzed for 12 metals by aas (Bove et al., 1972). The glass fiber filters used by most laboratories contain such large amounts of some of the elements of interest that X-ray intensities from the blanks are prohibitively high and variable. According to Kometani et al. (1972), these impurities also cause such serious difficulties during sample preparation for aas that the use of glass filters for air sampling should be avoided. The earlier study by Bove et al. (1972) indicated that less than 3% of the solids retained by glass fiber mats escapes the paper filters at the flow rates employed.

The list of metals measured—lead, iron, nickel, cobalt, copper, zinc, manganese, chromium, vanadium, cadmium, barium and beryllium—is long and may eventually have to be expanded. The very large number of determinations required makes desirable the development of simple, rapid, direct Xray analyses that could be fully automated using a computer or electronic calculator to control the spectrometer and sample changer. Accordingly, efforts were centered on producing such a direct X-ray method for the first 10 elements in the above list. As a by-product, however, information also was obtained concerning the simultaneous determination of several metals in solution by Coprex analysis.

Experimental

The X-ray spectrometer used in this work was a General Electric XRD-6S air or helium path instrument equipped with a four-crystal changer, bulk sample holder, dual (W-Cr) target X-ray tube, and dual (scintillation and flow proportional) counter tube detector system with solid state electronics. The direct X-ray analyses were made on 25-mm-diam disks cut from the large filter paper samples using a special die. These disks were held firmly by a clamping ring to an aluminum mask having an ¹¹/₁₆-in.-diam opening. The area of the particulate matter sample thus exposed to the X-ray beam is 0.371 in.² and is comparable to the area of Coprex specimens.

Preliminary tests on typical pollution samples indicated that, except for cobalt, the net X-ray intensities of the most favorable analytical lines were sufficiently high to provide a reasonable precision for all 10 elements using only 10-sec counting times. Separate experiments showed the sensitivity for cobalt to be adequate, but none of the preliminary samples happened to contain a significant quantity. For most of the elements, the scattered background measured for blank S.S. paper was about twice that found for the Millipore disks used in Coprex analysis, and the paper appears to contain traces of iron, copper, and zinc which make the background X-ray intensities somewhat higher than is desirable. Other experiments demonstrated that variations observed in the background intensity over the full area of an 8 \times 10-in. sample were within the 2 σ statistical counting error limits.

Metal deter-	Method			μg of N	Aetal/in. ² of :	sample of pa	per						
mined	used	1	2	3	4	5	6	7	8				
Pb	aas	41.1	21.1	57.9	40.4	31.4	25.4	44.1	22.8				
	Coprex	40.0	23.5	61.5	46.3	33.9	25.5	47.0	26.7				
	Direct X-ray	41.3	21.0	63.8	41.0	32.5	25.2	47.0	23.0				
Zn	aas	100.9	7.4	15.8	2.1	10.9	7.8	14.9	9.9				
	Coprex	95.8	7.1	15.3	1.8	10.4	7.3	14.1	9.5				
	Direct X-ray	101.0	8.7	19.0	2.1	12.0	8.3	16.0	10.2				
Cu	aas	18.9	2.9	10.5	2.2	2.3	3.2	5.3	2.1				
	Coprex	16.3	3.3	10.6	2.4	2.1	3.5	5.4	2.5				
	Direct X-ray	17.0	2.2	10.7	2.4	3.4	2.9	6.0	2.1				
Ni	aas	0.9	0.8	1.4	0.9	0.9	0.5	1.5	0.5				
	Coprex	1.0	0.9	1.4	0.9	0.9	0.6	1.5	0.5				
	Direct X-ray	1.1	0.8	1.6	0.9	0.9	0.7	1.5	0.5				
Fe	aas	17.2	24.9	42.5	12.5	27.1	21.7	46.0	22.1				
	Coprex	12.1	26.2	39.6	12.4	27.2	20.5	38.8	25.8				
	Direct X-ray	15.0	25.6	42.3	11.5	28.0	22.0	45.8	22.5				
Mn	aas	0.9	0.5	1.0	0.2	0.6	0.8	1.1	0.7				
	Coprex	0.7	0.4	0.8	0.2	0.6	0.8	1.1	0.7				
	Direct X-ray	0.6	0.3	0.9	0.2	0.7	0.7	1.2	0.7				
Cr	aas	0.2	0.3	0.9	0.2	0.8	0.3	0.5	1.2				
	Coprex	0.3	0.3	1.3	0.2	1.2	0.4	0.9	1.3				
	Direct X-ray	0.4	0.3	1.7	0.2	1.2	0.4	1.2	1.4				

Table I. Direct X-Ray Determination of Traces of Heavy Metals in Air Pollution Dust Collected on S & S Filter Paper

Also, differing amounts of pollutants on samples from various locations throughout the city did not cause statistically significant changes in the background. These data gave a strong indication that a satisfactory analytical method could be developed provided a suitable means of calibration could be found.

Coprex analysis (Luke, 1968) involves X-ray spectrometric counting of the desired element or elements as a precipitate collected on a Millipore filter. Standard wet chemical separation procedures are employed and a suitable coprecipitant is chosen to ensure quantitative recovery with minimum interference in the X-ray analysis. As a calibration technique, this approach appears to be simpler and probably more accurate than that proposed by Cares (1968). Since the amounts of the metals were very small, interelement effects, except for possible line interference, were predictably negligible. An adequate calibration for eight of the 10 elements was obtained using only a blank Millipore disk plus another carrying the mixed hydroxide and carbamate precipitate from a solution containing a know optimum amount of each element to be determined. Difficulties encountered in the measurements of cadmium and vanadium that could not be resolved are discussed in some detail below.

To establish the validity of the direct X-ray analysis, the amounts of the remaining eight elements on eight representative samples were also determined by aas and Coprex analysis as described below. The results shown in Table I indicate a reasonably good agreement between the three methods. The value given for aas is the average of separate analyses following wet ashing and dry ashing. The Coprex analysis was carried out on wet-ashed samples except for chromium which was measured after dry ashing. Data for cobalt are not included in the table since none was detected in any of the samples by any of the methods. The minimum detectable limit for cobalt by direct X-ray counting is estimated to be $0.02 \ \mu g/in^2$. The results for cadmium by the destructive methods are shown in Table II.

Ashing Procedures. The atomic absorption and Coprex analyses required a preliminary destruction of the filter paper. A wet-ashing technique was used along with the dry ashing (Bove et al., 1972) routinely employed by the New York City Department of Air Resources. Indications are repeatedly found in the literature of losses of metals during dry ashing, but the work of Kometani et al. (1972) and the results reported here clearly vindicate this method.

For wet ashing, place 1-5 in.² (depending on the metal content) of the filter paper samples in 125-ml conical flasks. To another flask add the same amount of clean S & S paper as a blank. Add 5–15 ml of HClO₄–HNO₃ mixture (1+4) and heat the open flask on a medium temperature hot plate to destroy the paper. When all of the HNO₃ has been expelled (ca. 20 min) and white fumes of HClO₄ are being evolved, swirl the flask over a direct flame until the volume has been reduced to about 0.25 ml. (This should be done in a perchloric acid– proof hood.) Cool and add 5 or 10 ml of deionized water, ignoring any slight cloudiness due to silica or silicates.

For dry ashing, heat the blank or sample paper in a 30-ml platinum crucible at 350° C for 30 min in a muffle furnace. Then raise the temperature to 500° C over another 30-min period. After an additional 2 hr at this temperature, cautiously remove the crucible—avoiding loss of ash by air currents—allow it to cool, and add 1 ml of HF–HNO₃ solution (1+2) from a plastic beaker using a plastic pipet or graduate. Heat the uncovered crucible to near dryness on a medium temperature hot plate, avoiding baking the residue. Cool, add 2–4

Table II. Determination of Cadmium by Coprex and aa Analysis

	H	g of Cadmium/in	.2
Sample no.	Dry ash, Coprex	Dry ash, aa	Wet ash, aa
1	1.0	1.2	1.1
2	0.2	0.2	0.2
3	0.6	0.5	0.5
4	0.3	0.2	0.2
5	0.1	0.1	0.1
6	0.3	0.2	0.2
7	0.7	0.6	0.6
8	0.3	0.2	0.1

drops of HCl [or, for aas, 2 ml of HNO₃ (1+10)], warm the crucible to dissolve the residue, and add 10 ml of deionized water.

Determination of Metals After Ashing. With the exception of vanadium, as noted below, and chromium, metals in the solutions from either ashing may be precipitated using essentially the procedure for the preparation of the standard Millipore disk and X-ray intensities compared with those from a multimetal standard disk. During wet ashing, perchloric acid oxidizes chromium to the hexavalent state, enabling some to escape precipitation. While this is undoubtedly correctable, the simpler answer is dry ashing. As an alternative to the Xray method the analyses can be carried out by aas. However, if vanadium is to be determined, larger samples must be employed because of low sensitivity.

Direct X-Ray Analysis

Standard Metal Solutions (1 mg/ml). The 1000-ppm standard solutions may be purchased or prepared. Chloride salts are preferred and enough acid should be present to prevent hydrolysis. Because of the presence of lead, solutions containing sulfate should especially be avoided along with any oxidizing agents that will destroy carbamate.

Multimetal Standard Solution. Transfer 5 ml of hydrochloric acid plus 50 ml of deionized water to a 100-ml volumetric flask and add appropriate amounts of the standard metal solutions. Any solutions containing nitrates should first be combined in a small conical flask, treated with 1 ml of perchloric acid, and evaporated to white fumes to expel all nitrates. After cooling, quantitatively transfer this perchlorate solution to the volumetric flask with the other standard solutions. Fill the flask to the mark with deionized water. In the present work, the expected amounts of pollutants dictated that 1 ml of the multimetal standard should contain 50 μ g of Pb and Fe, 10 μ g of Zn and Cu, and 2 μ g of Ni, Co, Mn, and Cr. These amounts should, of course, be adjusted to the analytical problem at hand.

Preparation of Standard Millipore Disk. Transfer 1.00 ml of the multimetal standard solution to a 50-ml beaker, add 10 ml of deionized water, 2 or 3 drops of hydrochloric acid, and a drop of 0.1% meta cresol purple indicator. Neutralize by the dropwise addition of ammonium hydroxide (1+1) and then add 1 drop in excess. Add 5 ml of a clear 2% solution of sodium diethyldithiocarbamate, swirl, and allow to stand for 5 min. (The carbamate solution should be stored in a plastic bottle and refrigerated when not in use.) Filter the solution slowly through a 25-mm, 0.8- μ porosity Millipore disk and wash with about 2 ml of deionized water. Dry the disk thor-

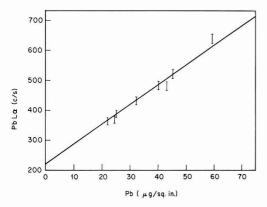


Figure 1. Lead L_{α} intensity vs. average aas and Coprex results

oughly at room temperature. For faster drying, place the wet disk on a glass surface at 50-60 °C using a lightweight plastic ring on top to prevent curling of the paper.

Analysis of Air Pollution Samples. The analytical line employed for X-ray counting is the K_{α} doublet for all elements except lead, for which $L_{\alpha 1}$ line is used. (The presence of As in the samples would necessitate using the slightly less sensitive Pb $L_{\beta 1}$ line.) The tungsten target of the EA 75 X-ray tube is operated at 50 kVP and 75 mA. Other parameters include a LiF (200) analyzing crystal, 10-mil Soller slit, and helium path.

Take a 10-sec count at the goniometer setting for the X-ray line of each element to be determined on a clean Millipore disk, a blank S & S paper disk, the multimetal standard disk, and all the S & S paper samples to be analyzed. For each analytical line, subtract the total intensity measured on the Millipore blank from that obtained on the multielement standard. Similarly, subtract the intensity on the S & S blank from those found on the pollution samples. Then use the formula

$$\frac{1}{0.371} \times \frac{\text{Net counts for sample}}{\text{Net counts for standard}} \times$$

µg of metal on standard disk

to compute the number of micrograms of each metal per square inch of the sample.

Direct X-Ray Determination of Vanadium. Vanadium K_{α} X-rays cannot be entirely resolved from titanium K_{β} radiation using a standard high-intensity 200 lithium fluoride crystal. A 220 cut LiF crystal provides slightly better separation of the lines, but with a serious loss in intensity. The weaker vanadium K_{β} line suffers an even more serious interference from chromium K_{α} . If the titanium interference were the only problem, the vanadium K_{α} intensities could easily be corrected by subtracting an appropriate fraction of the measured titanium K_{α} radiation.

Attempts to apply such a correction to the vanadium intensities on the New York City samples yielded disappointingly low results due to absorption of the relatively long wavelength radiation by the paper. X-ray counts on samples placed in the sample chamber with the clean side of the filter facing the X-ray beam prove that pollution dust buried deep in the coarse paper contributes fewer X-rays than if it were on the surface. This effect is significant only for X-ray lines with wavelengths as long as or longer than vanadium K_{α} . The absorption loss would be compensated for if the calibration were based on the determination of vanadium by destructive methods on actual samples like those to be analyzed. (This assumes that the particle size distribution of the various pollutants remains reasonably constant.) The best solution would be to collect the sample on a filter medium sufficiently less porous to keep the particulate matter on the surface.

Another problem is that contrary to indications in the literature (Sandell, 1959) not all the vanadium is precipitated at pH 9 by ammonia and carbamate. The recovery of vanadium is consistent, however, hence, no difficulty arises in Coprex analysis since the same loss occurs for the standard and unknown. For calibration of the direct X-ray method, however, a separate carbamate precipitation of vanadium should be made at pH 5 using an acetate buffer cleansed of heavy metals by extraction with chloroform after the addition of carbamate (Luke, 1967). At this pH, reasonable recoveries are made of all elements except chromium and manganese in the original list of 10.

Direct X-Ray Determination of Cadmium. The K radiation of elements like cadmium cannot be used because of inherent low sensitivity for thin layers and the presence of a high scattered background. Cadmium $L_{\beta1}$, the only intense line that can be diffracted by a lithium fluoride crystal, is not suitable due to interference from the strong potassium K_{α} line. Cadmium L_{α} radiation may be analyzed using a graphite crystal, the interfering second-order iron K_{α} line being minimized by pulse-height discrimination. Measurements on standards prepared by precipitation indicate a lower detectable limit for cadmium of 0.1 μ g/in.² for a 100-sec counting time.

The absorption effect previously noted for vanadium is so much more serious for cadmium that the latter cannot satisfactorily be determined in samples on No. 589 S & S paper. However, vanadium, cadmium, and other elements of possible interest that suffer this same limitation—titanium, calcium, potassium, silver, tin, antimony, and barium—could be determined in samples collected on Millipore filters.

Summary

A joint investigation by Bell Laboratories and the New York City Department of Air Resources has shown the feasibility of direct X-ray spectrometric analyses of air particulate matter for lead, iron, nickel, cobalt, copper, zinc, manganese, and chromium calibrated against a single Millipore disk containing known amounts of the eight elements as hydroxides or carbamates. Alternatively, samples analyzed by destructive methods could serve as standards. Figure 1, a plot of lead L_{α} intensity vs. the average of aas and Coprex results, illustrates such a potential calibration curve. The error bars represent $\pm 2 \sigma$ statistical variations in the 10-sec counts. Undoubtedly, other elements can be determined in the same way, but difficulties encountered with vanadium and cadmium warn against assuming that the method is universally applicable to all metals of interest.

The major advantage of the direct X-ray spectrometric technique over existing methods is the elimination of the timeconsuming and expensive ashing step. In addition, however, the actual instrument time amounts to just slightly more than 10 sec per element compared with about 30 sec per element for automated atomic absorption procedures.

Direct X-ray spectrometric analyses are readily susceptible to automation, either by computer control or by a hardwired control system employing an electronic desk calculator for computations. Such an automated system should provide a significant saving in time and expense to any organization with an extensive air monitoring program.

It should be obvious that the Coprex method could also be applied to water pollutants and to air pollutants collected in impingers. Ionized contaminants in aqueous solutions that do not contain substances interfering with precipitation could be simultaneously determined by Coprex analysis. In some instances, preliminary processing to eliminate undesirable components may be necessary or preconcentration may be required to attain sufficient sensitivity.

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Removal of Heavy Metals from Water and Brine Using Silicon Alloys

James P. McKaveney,1 William P. Fassinger, and David A. Stivers

Garrett Research and Development Co., Inc., Division of Occidental Petroleum Corp., La Verne, Calif. 91750

A one-step exchange treatment using silicon alloys is proposed for removing heavy metals from waste waters and brine solutions. Silicon alloys of calcium and magnesium produced the best results both from the view of scope in the exchange (zinc thru mercury in the electromotive series) as well as replacement of heavy metals with elements common to natural waters (calcium and magnesium). Silicon alloys of aluminum, barium, iron, and titanium were also found suitable but their use may require some posttreatment (hydrolysis or sulfate precipitation). Ferrosilicon is fairly selective in removing mercury without significant iron contamination of the water. The alloys can be used in either a column or batch contact type of operation. Brine and other aqueous solutions over the pH range of 2-12 have been treated. However, to minimize acid attack and to conserve alloy, the pH should be \geq 4.0. Metals successfully removed were: Ag, As, Au, Cd, Cu, Cr, Fe, Hg, Pb, V, and Zn. Mercury has also been successfully removed from chlor-alkali brines.

his research was undertaken to develop a suitable method for removing heavy metals (principally arsenic, cadmium, copper, mercury, lead, and zinc) at levels ranging from 5 to 100 ppm from aqueous solutions. Because of scope or economic considerations, the traditional procedures-viz: classical chemical (CaO, NaOH, or H₂S precipitation), ion exchange with resins, activated carbon and solvent extraction-were not extensively investigated. The use of metallic exchangers was appealing, however, since they appeared to offer broad scope, economy, and simplicity of operation for the removal of metallic impurities from water.

Metals such as iron and zinc are used in many water purification processes to displace less desirable metals such as copper and mercury from solution. The exchange is limited to the removal of metals below the exchanger metal in the electromotive series. The well-known cementation reaction for the recovery of metals in hydrometallurgical processes is a classical example of the use of metal exchangers. A recent work (Habashi, 1970), covers the commercial recovery of silver and copper with an iron exchanger as well as gold and silver from cyanide solution with a zinc exchanger. An earlier article (Kamecki and Sedzimir, 1952) treated the history, theory, and selection of the best conditions for the precipitation of metals by metals. Items included were the recovery of gold and silver from cyanide solution with aluminum and zinc, copper with iron, and finally copper, cadmium, nickel, and cobalt with zinc treatment. Other fairly recent cementation approaches involved arsenic with iron (Piontelli and Fracchia, 1949); bismuth and antimony on cadmium, iron, and lead (Kozlovskii et al., 1952); cobalt on zinc (Kamecki and Sedzimir, 1955); and copper on iron (Beck et al., 1964).

Analytical chemists were involved very early in the use of solid metals to separate or reduce less active metals present in solution as ions to a lower valence state by the use of more active metals, chiefly aluminum and zinc. Metallic aluminum was used to remove Bi, Cu, Cd, and Pb from solution (Stone and Waring, 1907) prior to the separation of zinc as the sulfide. The well-known Jones reductor (Jones, 1889) was developed for the determination of iron by using a column of zinc to reduce the ferric iron to ferrous prior to titration with potassium permanganate.

The use of metal reductants in water purification has had application for the removal of a number of heavy metals. Heavy metal impurities such as arsenic (Schegolov, 1947), copper (Wierzbicki et al., 1971; Wartman and Roberson, 1944), mercury (Katsuro, 1970; Karpiuk and Hoekstra, 1962; N.J. Zinc Co., 1971), and ruthenium (Lacey, 1952; Johannsen, 1968) are removed by direct deposition on the metal reductant (aluminum, iron, or zinc). However, metals such as chromium, which is usually present in the higher oxidation state (+6), are reduced by the metal reductant to the lower oxidation state and then separated with alkali as an insoluble hydroxide (Brushkin et al., 1970; Drakhlin, 1964). As zinc very often accompanies hexavalent chromium, the postaddition of alkali in the presence of chromium enables the chromium hydroxide to serve as a collector for zinc (Gaughan et al., 1971). Zinc has also been removed from cadmium and copper mixtures following contact on an aluminum or magnesium exchanger with postalkali addition (Borchert, 1967). Since mercury is fairly low in the electro-

¹ To whom correspondence should be addressed.

motive series (Latimer and Hildebrand, 1940) a fairly wide series of metals has been applied to its removal—viz: bismuth, copper, iron, magnesium, manganese, nickel, tin, and zinc (Gilbert and Rallis, 1962). However, at the present time, it is doubtful if any of the metals with the exception of magnesium would be permitted on a commercial basis unless an alkaline posttreatment were used.

To eliminate the alkaline posttreatment required to remove objectionable exchanged ions such as manganese, nickel, or zinc, the idea occurred that more active exchange metals such as calcium or magnesium would be preferred. In such a method the impurity such as nickel or zinc would be deposited on the metal reductant with the liberation of an equivalent amount of calcium or magnesium. The only obstacle to the concept appeared to be the extreme reactivity of metallic calcium with water. Magnesium metal, while much less reactive, could still not be used because of a high corrosion rate over an extended period. The writer's metallurgical experience indicated a possible solution by use of alloys of the metals whereby an element such as silicon makes the exchanger metal more resistant to aqueous attack.

Calcium and magnesium are present in all natural waters and are considered vital to many metabolic processes. The alloy concept appeared to be extendable to elements such as aluminum, barium, iron, and titanium which are not usually considered as hazardous to health. However, if an objection exists, aluminum, iron, and titanium are readily hydrolyzed and easily separated. Any barium entering solution would be separated by the sulfate present in most natural waters. While the U.S. Public Health Service set a maximum allowable limit of 1.0 mg/l. (1 ppm) in 1962 for barium in drinking water, the medical profession continues to use large dosages of barium sulfate in X-ray examination of gastrointestinal disorders. Therefore barium separated as the sulfate should not be considered harmful according to the medical profession.

Experimental

This initial study was performed to determine the scope of the silicon alloy concept and as such was primarily confined to selection of alloy types and their ability to remove the more common heavy metals. It is expected that currently continuing work will be used to determine rate processes, approach to equilibria, exchange ratios, and pH effects. The heavy metal solutions used were obtained by taking aliquots of

Table I.	Silicon Alloys Used for Heavy	Metal Removal
Alloy	Composition, % ^a	Source
CaSi ₂	32.4 Ca, 63.1 Si, 4.0 Fe, 0.4 Ba	Union Carbide
AlBaCaFeSi	20.5 Al, 12.4 Ba, 10.6 Ca, 39.0 Si, 17.0 Fe	Union Carbide
FeTi	69.8 Ti, 29.0 Fe, 0.03 Si	Union Carbide
AlFeSi	8.0 Al, 42.0 Fe, 49.0 Si	Union Carbide
MgFeSi	8.8 Mg, 45.2 Si, 45.0 Fe	Interlake Inc.
FeSi ₂	49.0 Fe, 49.0 Si, 1.0 Al, 0.1 Ca	Interlake Inc.
AlSi	49:0 Fe, 50.0 Si	Kawecki Berylco
Mg ₂ Si	63.0 Mg, 36.8 Si	Cerac/Pure
CaSiBa	16.6 Ca, 16.7 Ba, 57.0 Si, 8.5 Fe	Union Carbide
SiFe	3.0 Si, 96.0 Fe	Armco Steel

^a Alloys also contain minor amounts of other elements (C, P, S, Mn, N, O) which were not listed on the analytical certificates.

atomic absorption stock solutions (1000 ppm) and diluting to the desired level. The solutions were salts in either the chloride or nitrate form. Table I tabulates the alloys used along with their compositions and commercial sources. Certain of the active alloys such as Mg₂Si, AlBaCaFeSi, and CaSiBa tend to produce silane (spontaneous ignition in air) below a pH of 2.0 and their application should be restricted to values above a pH of 4.0. Subsequent studies have indicated that MgFeSi and CaSi₂ also tend to form some silane in more strongly acid solutions, pH <1.0.

Removal Studies for As, Cu, Cd, Hg, and Pb with Silicon Alloys. Two aqueous solutions were prepared containing 25 ppm of each of the elements: As, Cu, Cd, Hg, and Pb. One of the solutions also contained 30 grams/l. of sodium chloride. Fifty-milliliter samples of each of the solutions were placed into 250-ml Erlenmeyer flasks containing 50-ml volumes of various silicon-metal alloy particles (8 mesh). The flasks were then stoppered and shaken for 10 min on a mechanical shaker. After being shaken, each solution was filtered through No. 40 Whatman paper. The filtrates were collected in 125-ml polyethylene bottles prior to chemical analysis. Copper, cadmium, and lead were analyzed by standard atomic absorption analysis while arsenic was analyzed by arsine evolution into silver diethyldithiocarbamate and photometric measurement. Mercury was analyzed by flameless atomic absorption (Coleman MAS-50 mercury analyzer). Table II lists the results of the analyses.

Column Studies for Removal of Cr(VI), Hg, and Zn. Following the preliminary contact studies, column experiments were performed using the two alloys appearing to have the widest scope for heavy metal removal. The columns were glass ion exchange columns of 30 mm i.d. and contained alloy to a depth of 150 mm. The CaSi₂ alloy weighed 142 grams and was +80 mesh while the MgFeSi alloy weighed 150 grams and was +60 mesh. The initial pH of the metal contact solution was 5.6 and it contained: Cr (VI) at 7.6 ppm, Zn at 9.9 ppm, and Hg at 9.9 ppm.

Zinc and chromium (VI) were selected for study since they are fairly common metal impurities in many rinse waters from the metal-plating industries. Zinc also is fairly high in the electromotive series (+0.76 V) and is not presently removed by the conventional metal contact processes. Mercury was included because it is of current national interest as well as because it provided the lowest reference point for a wide scope of electrochemical exchange (-0.86 V). The complete analytical data on the heavy metals are shown in Table III as well as for the concentration of exchanged metal (Ca or Mg) put into solution.

Mercury Removal from Chlor-Alkali Brines. An actual plant sample from a mercury cell waste was chosen for contact study with several silicon alloys. Aliquots containing 100 ml of the collection tank waste were transferred to four 250-ml Erlenmeyer flasks each containing 5.0 grams of silicon alloys respectively: MgFeSi (+60 mesh), Mg₂Si (-20), AlSi (-20, +40), and CaSi₂ (+80). The flasks were stoppered and shaken for 30 min. After being shaken, the solutions were allowed to settle for 5 min, the pH was measured, and then filtered through No. 40 Whatman paper. The original solution as well as the filtrates were analyzed by atomic absorption for Hg, Al, Ca, Mg, Fe, and Si (Table IV).

The above experiment was repeated after reduction of pH of the collector tank sample to 6.5 with 1:1 hydrochloric acid prior to alloy contact (Table IV).

Heavy Metal Removal with Other Exchangers. An experiment was performed to compare the exchange ability of some

		Tab	le II. Me	etals Remai	ining After S	ilicon Alloy	Contact			
		Water base, a	ll at 25 pp	m to start		Saltwat	er base (30) grams NaC	/l.), 25 ppm	to start
Alloy	As, ppm	Cu, ppm	Cd, ppm	Pb, ppm	Hg, ppm	As, ppm	Cu, ppm	Cd, ppm	Pb, ppm	Hg, ppm
CaSi ₂	$<1^{a}$	<1	<1	<1	0.010	<1	<1	<1	<1	0.050
AlBaCaFe	<1	<1	<1	<1	0.010			Not treated	1	
FeTi	<1	<1	25	<1	0.015	14	13.4	25	25	0.062
AlFeSi	16	<1	25	23	0.015	16	3	25	23	0.055
MgFeSi	<1	<1	<1	<1	0.012	<1	<1	<1	<1	0.045
FeSi ₂	13	21	25	25	0.030	20	21	25	22	9.0
^a < Means	that the material o	contained less t	han one ppr	n and as sucl	n was undetecta	ble in the solu	ition.			
		Table III.	Metal I	mpurity Re	moval from	Water with	Silicon Al	lloys		
	Eluate	Flow								
Alloy column	volume cut, ml	rate, ml/min	Cr(VI), ppm	% Removal	Zn(II), ppm	% Removal	Hg(II), ppb	% Removal	Mg(II), ppm	Ca(II), ppm
Parent			7.6		9.9		9900		<0.01	<0.10
soln										
MgFeSi	400-500	10	5.4	29	<0.01	>99	2.6	>99.9	48	
	500-550	1 Hr static	2.3	70	<0.01	>99	2.4	>99.9	51	
	900-1000	2.5	5.3	30	<0.01	>99	0.8	>99.9	49	• • •
	1000-1050	Overnite static (15 hr)	<0.5	>93	<0.01	>99	2.0	>99.9	38	
	pH = 3.0 Separate	14	2.6	66	<0.01	>99	4.8	>99.9	54	6.00 B
CaSi ₂	400-500	10	6.3	17	<0.01	>99	10.8	99.9		104
-	500-550	1 Hr static	6.0	21	<0.01	>99	47.2	99.5		114
	900-1000	2.5	7.2	5	<0.01	>99	3.6	>99.9		69
	1000–1050	Overnite static (15 hr)	3.5	54	<0.01	>99	75.6	99.3	•••	124

of the silicon alloys with other materials such as activated carbon and ion exchange resins. The experiment involved contacting 100 ml of a heavy metals solution containing Cd, Cu, Hg, and Zn, each at 20 ppm, with the exchanger for 30 min on a shaker. The exchangers were about 20 mesh and weighed about 0.50 gram. The solution pH prior to contact was 4.5. After contact, the solutions were filtered and analyzed (Table V) as before.

Results and Discussion

The data of Table II indicate the wide scope of the silicon alloy concept for removing heavy metals from solution. The CaSi₂ (32.4% Ca), AlBaCaSiFe (20.5% Al, 12.4% Ba, 10.6% Ca) and MgFeSi (9.0% Mg) alloys appear to be effective in removing all of the impurities present (As, Cu, Cd, Pb, and Hg). Since such a large amount of alloy was present (\sim 125 grams) in contact with the 50 ml of heavy metals solution, it is assumed the data reflect true equilibrium capability for maximum metal removal. The test would hopefully be equivalent to a column-type operation where a maximum amount of alloy would contact the heavy metals solution.

The pH value of the heavy metals solution was quite acid (2.0) and this prevented further use of the heavy metals solution with the AlBaCaSiFe alloy in the saline medium. Considerable gas evolution occurred with this alloy on initial water contact, and some ignition of the gas occurred. As indicated earlier, it was suspected that silane (SiH₄) and possibly

hydrogen evolution resulted. The CaSi₂ and MgFeSi also caused gas evolution on initial contact but this was assumed to be entirely hydrogen as no ignition occurred. The gas evolution appeared similar to that experienced when an active metal such as zinc or iron contacts an acid solution. This indicates that any column operation would require venting. In nearly all subsequent experiments the pH was not permitted to go below 4.0 and this appeared to eliminate the heavy gas evolution. The presence of excess hydrogen ion would also cause unnecessary alloy consumption and should be avoided.

Column-type operation is illustrated in the data of Table III. It is apparent that both the CaSi₂ and MgFeSi are capable of removing metals as bigh as zinc in the electromotive series. The data for chromium indicate that removal only occurs after the alloy has had sufficient time to reduce the chromium(VI) to the trivalent state. A lower pH value also assists the reduction and removal as shown by the pH = 3.0 data for the MgFeSi. Obviously a prereduction step with SO₂ would be required to remove chromium in continuous column operation. Subsequent experiments have confirmed this postulate. However, if alloy cost is not critical, particularly for small batch-type operation, then either prolonged alloy contact or acid addition to pH 3.0 can be used.

The data on the chlor-alkali brines (Table IV) are apparently not equilibrium or equivalent to column-type operation as only 5.0 grams of alloy were in contact with 100 ml of solution. However, the data indicate that mercury can be com-

Table IV. Mercury Removal from Chlor-Alkali Brines with Silicon Alloys											
Alloy	Soln pH	Chloride, gram/l.	Ca, ppm	Mg, ppm	Al, ppm	Fe, ppm	Si, ppm	Hg, ppm	% Hg, removal		
Parent											
soln	12.3	32.8	23.9	0.10	11.0	1.4	8.2	10.6	0.0		
AlSi	11.8	32.8			600.0		3.9	1.2	88.7		
AlSi	7.1	35.1	26.1	0.5	11.0	1.2	5.3	2.9	72.6		
Mg ₂ Si	12.3	32.8		0.06			31.0	1.4	86.8		
Mg ₂ Si	10.6	35.1		111.0	11.0		1.2	0.3	97.2		
MgFeSi	12.3	32.8		0.03		1.3	14.5	4.1	61.3		
MgFeSi	10.2	35.1		100.0		1.3	27.0	0.8	92.5		
CaSi ₂	12.3	32.8	43.6				13.2	4.2	60.4		
CaSi ₂	10.0	35.1	155.0				158.0	0.7	93.4		

a All data for samples refer to values on treated filtrates from 100-ml volumes initially in contact with 5.0 grams of alloy for 30 min.

		Table V.	Heavy	Metal Extra	action by I	Exchange	Materials			
			Meta	als remainin	g after cont	act (all at	20 ppm to s	tart), ppm		
Material	Cd	Cu	Hg	Zn	Na	Fe	Mg	Ca	Si	Ba
Activated carbon (Darco-G 60)	19.7	7.4	0.4	19.9						
Cation exchange resin (Dowex HCR-W)	0.4	0.4	19.3	0.5	pH = 2.0					
Anion exchange resin (R-H IRA-93)	19.4	14.5	0.8	18.5	77.1					
Mg ₂ Si	0.3	0.3	0.1	0.4			38.0		1.6	
CaSiBa	<0.1	<0.1	0.1	<0.05				10.0	45.0	129.0
MgFeSi	3.1	0.5	1.3	0.6		0.1	27.0		0.3	
SiFe	16.8	2.3	0.4	6.9		36.2			0.5	
^a 0.50 gram of material in	n contact wi	th 100-ml so	lution for	30 min.						

pletely removed and that Mg_sSi reactivity probably indicates that it has the highest electrochemical potential of the alloys used. Recent laboratory experiments have indicated that the other magnesium alloy (MgFeSi) can remove down to 10 ppb of mercury from chlor-alkali brines in simulated column-type operations.

The AlSi alloy is obviously too reactive in the pH 12 solution as evidenced from the gas evolution during alloy contact as well as the high aluminum content (600 ppm) of the solution after treatment. Pretreatment of the brine with hydrochloric acid prior to alloy contact apparently solves this problem (11 ppm Al after contact).

The data for the magnesium alloys in the high pH brine indicate practically no magnesium pickup, despite the fact that mercury has been removed and significant silicon dissolution has occurred. The higher pH probably favors silicon dissolution in the alloy as well as precipitation of magnesium hydroxide from solution. The data for the magnesium alloys in the lower pH solutions (10.2 after contact) indicate faster mercury removal but at the expense of considerable magnesium dissolution (about 100 ppm).

The CaSi₂ alloy is also attacked more strongly at the lower pH than for the higher pH (131.1 vs. 19.7 ppm net calcium in solution). Obviously in terms of economics the CaSi₂ alloy would be the best to use (at the higher pH) as the removal of 6.4 ppm of mercury required 19.7 ppm of calcium or about 3 ppm of Ca for every 1 ppm of Hg. At an alloy cost of 0.25/lb of CaSi (33% Ca), it would cost 0.40/1000 gal. to remove 20 ppm of mercury. The varying reactivity of the alloys in the chlor-alkali brines is not completely understood; however, we suspect that dissolved chlorine may account

1112 Environmental Science & Technology

for part of it, at least for the calcium and magnesium alloys. The amphoteric nature of aluminum would account for its increased reactivity in the more alkaline solution.

The data of Table V were prepared to give some comparison of the silicon alloy metal removal capability with other commonly used exchange materials. The activated carbon and ion exchangers selected have been used at this laboratory in earlier metal removal studies. The HCR-w resin appears quite good for Cd, Cu, and Zn while the IRA-93 is very good for mercury removal. The activated carbon appears to be good for the most easily reducible metals (Hg and Cu). Mercury was dissolved in solution at the 20-ppm level as mercuric chloride and presumably is present as a cation. However, its behavior with the ion exchange resins would suggest that even in this very dilute solution, it is behaving as an anion.

The Mg₂Si and CaSiBa alloys are obviously quite good for removing all four heavy metals. While the CaSiBa alloy may inject barium into the water, as was indicated earlier, the presence of sulfate in all natural waters would remove it as barium sulfate. The MgFeSi alloy while containing a major amount of iron does not exchange any of the iron while magnesium is still present in the alloy. The low-silicon SiFe data indicate that the alloy removes zinc in preference to cadmium (zinc is above iron while cadmium is below iron in the electromotive series). The explanation for this is probably related to the scavenging effect of ferric iron when it hydrolyzes from solution. Although the solution indicated appreciable iron in the filtrate (36.2 ppm), a brown ferric hydroxide floc was clearly visible prior to filtration. The iron passing through the filter was obviously in the divalent state while that oxidized by dissolved oxygen was removed by the filter. In essence, zinc ap-

pears to be scavenged by ferric iron hydrolysis while cadmium is only slightly affected.

Some cementation-type experiments have been performed with MgFeSi and CaSi2 alloys on the recovery of gold, silver, and vanadium from solution. They were, in general, successful for gold and silver when the elements were complexed with a ligand such as NH3 or S2O32- but failed for the cyanide complexes. Apparently a pretreatment with an oxidizer (chlorine or peroxide) would be required to destroy the cyanide complexes. Vanadium was studied as the vanadate (pentavalent) and like dichromate, required prolonged contact for successful removal. Again a prereduction step with SO2 would be required for a column-type removal of vanadium.

Regarding economics, no detailed costs are available since the procedure requires additional study on alloy saturation, exchange ratios, etc. However, from (limited) bench-scale tests, with an alloy such as CaSi2, cost estimates appear to be about \$0.20/1000 gal. treated at the 20-ppm level for the conventional heavy metal rinse water. As indicated earlier for a brine sample, the cost would about double to \$0.40/1000 gal.

Recovery studies have not been investigated in depth, as the alloys are fairly economical ranging from \$0.08/lb (FeSi2) to \$0.40/lb (AlBaCaSiFe). An exception would be FeTi which sells for \$1.54/lb of contained titanium. Although FeTi is fairly expensive, acid recovery work indicates it to be fairly stable permitting the removal of surface-deposited metals. Mercury has been distilled from MgFeSi by heating the alloy in a nitrogen stream. The MgFeSi also appears to have self-regenerative properties for mercury removal as evidenced from successive contact studies on a fixed sample surface. Calculations of the available surface area had indicated the mercury adsorption would decrease with successive contacts. However, repeated contact failed to show any decline in mercury adsorption ability. The experiments were not continued to alloy saturation.

The concept of alloying has been extended beyond silicon in some very recent work to other metals such as chromium, copper, nickel, and molybdenum so as to metallurgically tailor the alloy to effect selective metal separations. By combining these elements tending to reduce the chemical potential by Raoult's law with the more active elements in the electrochemical series, it should be possible to obtain electrochemical potentials intermediate between two or more elements that one desires to separate.

While the mechanism of heavy metal removal appears to be primarily electrochemical, deposition for elements such as copper and mercury, it appears that additional electrochemical mechanisms may be possible with other metals such as arsenic chromium, vanadium, and zinc. With the higher oxidation states of each element, the initial step may be electrochemical reduction: As⁵⁺ \rightarrow As³⁺, Cr⁶⁺ \rightarrow Cr³⁺ or Cr²⁺, and V⁵⁺ \rightarrow V³⁺ or V²⁺. The second step may be hydroxide formation or hydrolysis

The hydroxide is usually supplied from the slight water reactivity of the more active metals in the alloy

With iron alloys, laboratory data indicate a coprecipitation or scavenging effect is also operating:

 $Fe(OH)_2^+ + Zn^{2+} + 3 OH^- \rightarrow Fe(OH)_3 \cdot Zn(OH)_2$ $Fe(OH)_2^+ + As^{3+} + 4 OH^- \rightarrow Fe(OH)_3 \cdot As(OH)_3$

With barium alloys, the barium ion can undergo further reaction as indicated earlier:

$$Ba^{2+} + CrO_4^{2-} \rightarrow BaCrO_4$$

$$Ba^{2+} + SO_4^{2-} \rightarrow BaSO_4$$

Some recent data indicate the alloy CaSiBa is also very good for removing chromate from metal rinse waters.

An interesting aspect of the structure of the silicon alloy after departure of the exchanger metal relates to the silicon skeleton. If a silica gel-type surface develops at the reacted alloy sites, then enhanced metal removal should result. In addition to this aspect and the prior indicated rate needs, future work is necessary to fully investigate the various mechanisms of heavy metal removal. Also required are data on the effect of codeposited ions (mercury followed by copper) or precipitation of metal interaction products on the active alloy surface.

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NOTES

Improving Linear Relations to Obtain Kinetic Parameters

Dominic M. Di Toro

Environmental Engineering & Science Program, Manhattan College, Bronx, N.Y. 10471

An iterative method is presented which corrects the truncation error inherent in the linearized approximations commonly used in obtaining reaction kinetic parameters. This correction extends the applicability of these convenient methods.

Use of graphical techniques to analyze kinetic data is an established and useful part of the methods that are routinely applied to water quality data. Two examples are the Thomas slope method (Thomas, 1950) for obtaining the first-order reaction coefficient, K_1 , and ultimate BOD concentration, L_0 , from data obtained at times t_i , i = 1, ..., N, and assumed to follow the reaction model

$$L(t) = L_0(1 - e^{-K_1 t})$$
(1)

and a method suggested by Pessen (1961) to obtain both the order, n, and the reaction coefficient, K_n , from data which is assumed to follow the reaction model

$$\frac{dc}{dt} = -K_n c^n \tag{2}$$

or, in integrated form:

It

$$c(t) = c(0)[1 + (n-1)tK_nc_0^{n-1}]^{1/1-n}$$
(3)

where
$$c(o)$$
 is assumed known. Both of these techniques rely
on approximately linear relations that exist between expres-
sions involving time and the measured concentrations vs. time

The Thomas slope method depends on the approximate equation

$$[L(t)]^{1/3} = (K_1 L_o)^{-1/3} + (K_1^{2/3}/6 L_o^{1/3})t$$
(4)

and the nth-order method is based on the relationship

$$\frac{t}{f} = \frac{1}{\alpha} + \frac{n}{2}t \tag{5}$$

where

$$f = \frac{c(o) - c(t)}{c(o)}$$
 (6)

and

$$\alpha = K_n c(o)^{n-1} \tag{7}$$

Since a derivation of this relationship is not readily available, one is presented below. The linearization of the *n*thorder reaction solution is based on the expansion of the solution in an appropriate way. For both n = 0 and n = 2 the linear relation given by Equation 5 holds exactly. Thus it is reasonable to investigate whether t/f vs. t is approximately linear for other reaction orders. Toward this end the solution for an *n*th-order reaction is:

$$(1 - f)^{1 - n} - 1 = \alpha t(n - 1)$$
(8)

Expanding the binomial $(1 - f)^{1-n}$ and simplifying yields:

$$f + \frac{n}{2}f^2 + \frac{n(n+1)}{6}f^3 + \ldots = \alpha t$$
 (9)

or

$$\frac{\alpha t}{f} = 1 + \frac{n}{2}f + \frac{n(n+1)}{6}f^2 + \dots$$
(10)

Since the coefficient of f^2 in this expansion is not small, this expansion does not lead to a good approximation directly. What is required is t/f expanded in powers of t. This can be accomplished using Equation 10 and an expansion for f in terms of t. To obtain the latter expansion, the technique of reversion of series (Abramowitz and Stegun, 1964) is used. This is, if

$$y = a_0 x + a_2 x^2 + a_3 x^3 + \dots$$
(11)

then

$$x = \frac{1}{a_1} y - \frac{a_2}{a_1^3} y^2 + \frac{(2 a_2^2 - a_1 a_3)}{a_1^5} + \dots$$
 (12)

Thus Equation 9 is inverted to give:

$$f = \alpha t - \frac{n}{2} (\alpha t)^2 + \frac{n(2 n - 1)}{6} (\alpha t)^3 + \dots$$
 (13)

And substituting this equation into Equation 10 gives the required expansion:

$$\frac{\alpha t}{f} = 1 + \frac{n}{2}(\alpha t) + \frac{n(2-n)}{12}(\alpha t)^2 + \dots$$
(14)

the first two terms of which are an approximation of the exact solution.

These linear relations are the basis for the simple graphical methods since plots of $(t/L)^{1/3}$ and (t/f) vs. time are approximately linear with respect to time and the slopes and intercepts of the fitted straight lines are used to calculate the unknown kinetic parameters. A difficulty with this procedure stems from the approximate nature of the equations used. These equations are the first two terms of exact infinite series expansions and the resulting truncation error makes them approximate. It is the purpose of this note to suggest a method of iteratively reducing the truncation error to make these methods asymptotically exact.

Estimating Truncation Error

For the sake of being explicit, the *n*th-order method will be used in this exposition; a similar analysis applies to the Thomas method. The expansion that relates t/f to t can be written as

$$\frac{t}{f} = \frac{1}{\alpha} + \frac{n}{2}t + R(n, \alpha, t)$$
 (15)

where $R(n, \alpha, t)$ is the truncation error which is a function of *n* and α , the unknowns. However, a first estimate of these unknowns, n_1 and α_1 , can be obtained by using the approximate linear relation and ignoring the truncation error *R*. Thus n_1 and α_1 , can be obtained by fitting a straight line to t_i/f_i calculated from the data f_i , vs. t_i . Thus fitting

$$\frac{t_i}{f_i} = \frac{1}{\alpha_1} + \frac{n_1}{2} t_i \quad i = 1, \dots, N$$
(16)

gives n_1 and α_1 .

1114 Environmental Science & Technology

Using these estimates it is possible to estimate the truncation error by simply subtracting the exact value of t/f evaluated using the known solution

$$f(n, \alpha, t) = 1 - [1 + (n - 1)\alpha t]^{1/1 - n}$$
(17)

and the approximation, Equation 16. Thus

$$R(n_1, \alpha_1, t_i) = \frac{t_i}{f(n_1, \alpha_1, t_i)} - \left(\frac{1}{\alpha_1} + \frac{n_1}{2}t_i\right)$$
(18)

is the truncation error for n_i , α_1 at t_i . This truncation error can be removed and new estimates, n_2 , α_2 are obtained by fitting a straight line to the improved equation:

$$\frac{t_i}{f_i} - R(n_1, \alpha_1, t_i) = \frac{1}{\alpha_2} + \frac{n_2}{2} t_i \quad i = 1, \dots, N$$
 (19)

With the improved estimates n_2 , α_2 the process can be repeated by recalculating the truncation error $R(n_2, \alpha_2, t_i)$ using Equation 18, fitting the data using Equation 19, and so on. As shown in the accompanying example the process appears to converge in a few iterations. The result is exact in the sense that the method converges to the correct order and reaction coefficient if the data used are exact. The method also appears to work well with actual data although the effects of measurement errors can be troublesome if the errors are large and the data scattered. For such cases it is questionable whether kinetic models that result from any fitting technique such as direct least-square error minimization (Ravimohan, 1969) have much utility. In any case the statistical analysis of the resulting kinetic parameters becomes an important adjunct to the fitting technique.

Example

Synthetic data, f_i , for a reaction order of n = 1.5 and for which $\alpha = 1.0$, calculated for $t_i = 0.5, 1.0, 1.5, \ldots, 4.0$ is shown in Table I together with t_i/f_i and the exact truncation error $R(1.5, 1.0, t_i)$. The iteration procedure described above is applied to this data. The straight line fit is accomplished using a least-mean-square error criteria. The resulting order and reaction rate at each iteration is shown in Table II. Convergence is obtained after 19 iterations.

Algorithm

The algorithm used in the above example is as follows: For the data, the concentration fraction remaining at each observation time $f_i, t_i, i = 1, ..., N$

(1) calculate the ordinate values

$$d_i = t_i / f_i$$
 $i = 1, ..., N$ (20)

(2) least-mean-square error estimate of the slope to calculate order

$$n = 2\left(N\sum_{i}d_{i}t_{i} - \sum_{i}d_{i}\sum_{i}t_{i}\right) / \left[N\sum_{i}t_{i}^{2} - \left(\sum_{i}t_{i}\right)^{2}\right]$$
(21)

(3) least-mean-square error estimate of the intercept to calculate reaction coefficient

$$\alpha = N \left/ \left(\sum_{i} d_{i} - \frac{n}{2} \sum_{i} t_{i} \right) \right.$$
(22)

(4) exact value of f at t_i for n and α calculated above

$$f(n, \alpha, t_i) = 1 - [1 + (n-1)\alpha t_i]^{1/1-n}i = 1, \dots, N \quad (23)$$

(5) truncation error at t_i for *n* and α calculated above

				R(1.5, 1.0,
i	ti	f_i	ti/fi	t_i
1	0.5	0.640	1.388	0.013
2	1.0	0.444	1.800	0.050
3	1.5	0.327	2.227	0.102
4	2.0	0.250	2.667	0.167
5	2.5	0.198	3.115	0.240
6	3.0	0.160	3.571	0.321
7	3.5	0.132	4.033	0.408
8	4.0	0.111	4.500	0.500

Iteration	n	α
1	1.78218	1.10141
2	1.65589	1.05719
3	1.58484	1.03158
4	1.54575	1.01718
5	1.52452	1.00926
6	1.51310	1.00496
7	1.50699	1.00265
8	1.50372	1.00141
9	1.50198	1.00076
10	1.50106	1.00040
11	1.50056	1.00022
12	1.50030	1.00012
13	1.50010	1.00006
14	1.50008	1.00003
15	1.50005	1.00002
16	1.50002	1.00001
17	1.50001	1.00001
18	1.50001	1.00000
19	1.50000	1.00000

$$R(n, \alpha, t_i) = \frac{t_i}{f(n, \alpha, t_i)} - \left(\frac{1}{\alpha} + \frac{n}{2}t_i\right)i = 1, \dots, N \quad (24)$$

(6) ordinate values adjusted for truncation errors.

$$d_i = \frac{t_i}{f_i} - R(n, \alpha, t_i) \quad i = 1, ..., N$$
 (25),

(7) Go to (2) until procedure converges.

Conclusion

The technique described above provides an iterative improvement for reducing the truncation error in approximately linear relations that are used to estimate kinetic parameters. The technique is general and can be applied to any such formulation.

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Concentration of Arsenic from Water Samples by Distillation

Edward J. Farkas,1 Robert C. Griesbach, David Schachter, and Michael Hutton

Department of Chemical Engineering, McGill University, Montreal, Que., Canada

Determination of the amount of arsenic in samples of drinking water is facilitated if the arsenic in the sample can first be concentrated into a smaller volume. The utility of the distillation method described in standard reference works was investigated. Optimum conditions for application of this method were determined based on experiments in which percentage recovery of the arsenic present in the original sample was measured as a function of the amounts of the reagents added to the original sample and of the volume of distillate collected. Improvements in experimental technique are also described.

In the course of measuring in our laboratory the concentration of arsenic in samples of solids and in aqueous samples, it appeared useful to concentrate the arsenic from a large sample into a smaller volume. Determination of the amount of arsenic present by the usual methods (Amer. Public Health Assoc., 1967) would thereby be facilitated.

The distillation method was selected from accepted sources (Furman, 1962; Hillebrand and Lundell, 1959; Scott, 1947). The arsenic comes over in the form of AsCl₃. This compound is supposed to go into solution in the distillate flask but in view of the small quantities involved in analyzing drinking water, for example, the use of a carrier gas is not advisable. The vapor pressure of AsCl₃ (Hodgman et al., 1957) is not insignificant in this application. In the covery of arsenic in the distillation method were studied systematically. The object of this communication is to present the optimum values of these variables and to describe various other improvements in the technique.

Results

We can regard the distillation process as one for which

$$R = f(V, m, c, d, H)$$

where R = percentage of arsenic initially present in the distillation flask which is recovered; V = total volume of solution initially present in the distillation flask, ml; m = number of micrograms of arsenic initially present in the distillation flask; c = number of grams of cuprous chloride present in the distillation flask; d = volume of distillate collected, ml; H= wt % hydrochloric acid in the solution initially charged to the distillation flask.

The range of values of H studied was based on indications (Baumhardt, 1953; Manufactures de produits chimiques du Nord, 1952; Scherrer, 1936; Snell and Snell, 1967) that the acid concentration should be above that corresponding to the azeotropic or constant boiling mixture. The latter value is 20.2 wt % at atmospheric pressure (Bonner and Wallace, 1930). Results are shown in Table I.

To determine exactly the recovery, it was necessary to

consider the amount of arsenic contributed by the reagents. The major contributor was cuprous chloride. Recommendations on amount to be used varied widely in the literature. Different amounts were tried as shown in Table I. From these results it appears that 0.2 gram cuprous chloride would be adequate.

We wished to check the statement to the effect that all the arsenic is contained in the first 10 or 15 ml of distillate (Scott, 1947). The condenser was designed to minimize holdup and after the amounts of distillate indicated in Table I had been collected, the apparatus was dismantled, and arsenic-free wash water was used to rinse any material remaining in the condenser into the distillation flask. There was no quantitative indication that the amount of distillate collected could not have been lower, but intuitively possibly not as low as 10 ml.

Experimental Technique

For reasons indicated, a carrier gas was not used and the vertical condenser was of the Allihn type to minimize holdup of distillate. The apparatus was closed to the atmosphere except for a side arm on the distillate collection flask. From this side arm, flexible tubing led to the top of a dip tube inserted through a stopper in the top of another flask with side arm. The latter flask had a capacity of about 1 liter and was filled to a depth of about 2.5 cm with a concentrated NaOH solution. This solution was slowly swirled with a magnetic stirrer. The dip tube came down to within 0.5 cm of the surface of the basic solution. This arrangement collected any HCl vapors evolved without the risk of suckback. The side arm of the NaOH flask was connected by flexible tubing to the exterior of the building.

The distillate flask (250 ml) was kept cold ($\sim 0^{\circ}$ C) and the condensate was led into it through a dip tube which was not long enough to become immersed in the liquid in the flask. Initially the distillate flask contained 25 ml distilled water and 25 ml concentrated HCl. After the distillation was com-

Table I. Results of Distillation Experiments^a

Test no.	Н	с	m^b	d	μg As recovered
1	34	0.0	15	50	9.0
2	34	2.0	15	50	16.0
3	34	2.0	15	50	17.0
4	34	2.0	0	50	3.4
5	34	2.0	0	50	5.6
6	34	0.5	15	50	14.3
7	34	0.5	15	50	14.3
8	34	0.2	15	50	14.3
9, 10, 11, 12	34	0.5	15	25	16.4 (av)
13	27	0.5	15	25	6.0
14	27	0.5	15	25	8.0
15	30.5	0.5	15	25	15.5
16	30.5	0.5	15	25	12.0

^a V = 165 ml for all runs.

^b Amount added intentionally (not including arsenic introduced with reagents).

¹ To whom correspondence should be addressed.

pleted and before determining the amount of arsenic collected (Amer. Public Health Assoc., 1967), another 100 ml of cold distilled water was added. The HCl was thereby diluted and overly violent reaction with the arsenic determination reagents was avoided.

Discussion

We conclude that the literature was incomplete in its specification of the distillation method. The acid concentration in particular influences the results guite markedly, based on our results.

In applying the results reported here to analysis of drinking water supplies and surface water samples, it must be remembered that all tests involved the same values of V and m. Thus the functionality indicated above has not been completely determined. In particular we did not have the opportunity to vary V with m held constant—e.g., at $15 \mu g$.

The maximum desirable amount of arsenic in drinking water is 0.01 mg/l. and the maximum safe amount is 0.05 mg/l. (Kopp and Kroner, 1967). Contribution of arsenic by reagents and other normal considerations in both the distillation procedure and in the determination of the amount of arsenic collected (Amer. Public Health Assoc., 1967) dictate that as a practical matter on the order of 10 μ g of arsenic be present. Thus in the case of drinking water at or below the desirable arsenic level a sample size of 1 liter or more would be necessary.

For 1-liter samples it would not be practical to add concentrated HCl, which is itself over 60% water. A possibility is to bubble HCl gas into the sample until the acid concentration is on the order of 30 wt % or more. It might be best to do this at a very low rate of bubbling, before adding the cuprous chloride, and with the sample kept cold ($\sim 0^{\circ}$ C). Once the desired acid concentration was reached, the cuprous chloride could be added and distillation could begin.

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Particle Counting by Glow Discharge Perturbation

Nicholas A. Fedrick, Alexis T. Bell,1 and Donald N. Hanson

Department of Chemical Engineering, University of California, Berkeley, Calif. 94720

A constricted glow discharge has been assessed for its use as a means for counting and sizing airborne particulate matter. The device consists of a tube in which two electrodes, separated by a disc containing a small orifice, are mounted. Passage of a particle through the orifice causes a perturbation in the discharge current. The magnitude of these perturbations has been found to be proportional to particle diameter.

easurement of the size and concentration of particulate matter in various media has become an increasingly important problem. Considerable effort has been expended on the development of various methods for these measurements. Of the devices proposed and used, one of the simplest is the Coulter counter (Coulter, 1953), which is extensively used to count and size particles in liquids. The use of the Coulter counter, however, is restricted to particulate

matter suspended in a liquid. If such a counter is to be used for analyzing gas streams, the particulate matter must first be collected and then introduced into the liquid. This procedure necessarily involves uncertainties in collection efficiency and agglomeration. It would be desirable therefore to count the particles directly in the gas phase. This could be accomplished if the gas were made conductive through its partial ionization. The present note describes a technique by which it is possible to develop a gas-phase analog to the Coulter counter through the use of an electric discharge.

The device described here makes use of a dc glow discharge maintained between two electrodes separated by a disc containing an orifice of approximately the same size as the particles to be counted. If a particle of solid material is allowed to pass through the orifice, a momentary interruption occurs in both the discharge current and voltage. These perturbations which can be viewed on an oscilloscope can be used to determine the particle size and number.

Experimental Apparatus and Discussion

The experimental apparatus is shown diagrammatically in Figure 1. The particle counter consists of four glass sections,

¹ To whom correspondence should be addressed.

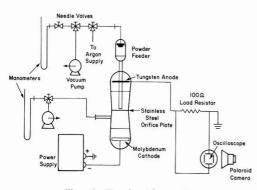


Figure 1. Experimental apparatus

mated to each other by standard taper ground-glass joints. The lowest glass section contains a molybdenum electrode which is connected to the negative output terminal of the power supply. The second glass section functions as a support for a stainless steel disc, 100μ thick, 1 in. in diam containing a $153-\mu$ hole drilled through the center. The orifice is affixed to the tube with Silastic cement. The third glass section consists of a support for the powder feeder and a length of 3-mm i.d. glass tubing used to direct the particles toward the orifice once they have sifted through the feeder. The lower portion of this section houses a tungsten ring-electrode, which is grounded and serves as the anode. The uppermost glass section connects to the gas supply manifold.

The particulate matter used in this work consisted of sized glass beads. Beads having nominal sizes of 50 and 100 μ were sieved before use in order to assure a minimum deviation from the mean diameter. Observations of 25 beads through a microscope showed the following spread in bead diameters:

Nominal 50 μ : high, 56 μ ; low, 44 μ ; av, 50 μ Nominal 100 μ : high, 112 μ ; low, 88 μ ; av, 105 μ

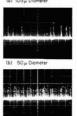
For the experiments, these beads were loaded into a powder feeder consisting of three fine-mesh screens individually sandwiched between flat plastic washers. By gently tapping the feeder a small number of beads could be caused to fall through the screens and into the glass tube leading to the orifice. Because of the pressure differential existing across the orifice the beads were swept through the aperture and out of the system.

The passage of each bead through the orifice produced a perturbation in the discharge current. These perturbations were observed as voltage pulses (with a Tektronix Model 502A oscilloscope) across a 100-ohm load resistor connected in series with the discharge. To isolate the base dc current, the connection to the oscilloscope was made through a capacitor and the oscilloscope was used in its ac mode. Permanent records of the pulses were made by photographing the oscilloscope trace with a Polaroid camera.

Results and Conclusions

Preliminary experiments were performed without the particles to determine the optimum pressures and current to be used. It was concluded that a stable discharge could be maintained using the following conditions: pressure upstream





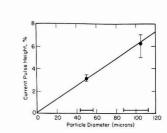


Figure 2. (left) Current pulse

Horizontal sensitivity, 2 msec/cm

Vertical sensitivity, 5 mv/cm

measurements

Figure 3. Correlation between current pulse height and particle size

from the orifice = 20 torr, pressure downstream of the orifice = 2×10^{-3} torr, and current = 2.1 mA.

When glass beads were placed in the feeder and then allowed to fall through the orifice a perturbation could be observed in the oscilloscope trace. The decrease in current was noted as a percentage of the base current. Figures 2a-c are typical photographs of the pulses recorded in such a manner that an upward pulse indicates a decrease in current. As may be seen from Figure 2c the pulse heights for a given particle size are quite uniform. A small amount of noise can also be observed in the photographs as very rapid symmetric pulses. Although it was not possible to eliminate these noise pulses with the experimental arrangement used, they were readily distinguishable from the pulses due to the passage of a particle through the orifice. Each of the pulses is characterized by a sudden rise to a maximum value and a more gradual recovery to the base signal. Average pulse times for both the 50 and $105-\mu$ beads were on the order of 1.5 msec.

Figure 3 shows a plot of the average pulse height vs. the average bead diameter. For each bead size the spread in the current pulses observed is indicated by the vertical error bars, and the spread in measured bead diameters is indicated by the horizontal error bars. The magnitude of the variations from the mean value of the current ranged from +11 to -8% for the 50- μ beads and from +10 to -20% for the $105-\mu$ beads. The spread of particle diameters was $\pm11\%$ for the 50- μ beads and +7 to -16% for the $105-\mu$ beads. The extent of variation in current pulse height and bead diameter are in very close agreement with each other.

From Figure 3 it can be observed that there is a linear relationship between pulse height and particle diameter. Whereas the device with its present orifice of 153μ is limited to detecting particle diameters on the order of 10μ and larger, the principle of the device could be extended to smaller particles through the use of a smaller orifice. Preliminary results have already indicated that it is possible to sustain a discharge through a $30-\mu$ orifice.

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Prediction of Heat of Combustion of Solid Wastes from Ultimate Analysis

Donald L. Wilson

U.S. Environmental Protection Agency, National Environmental Research Center, Cincinnati, Ohio 45268

This paper relates how thermochemical principles can be applied in predicting the heat of combustion of prepared solid waste samples, knowing their ultimate analysis. The resulting formula produces data whose variation from average experimental value is similar in magnitude to the variation between duplicate experiments.

The heat contents and ultimate analyses of the solid waste materials to be incinerated are necessary to determine the efficiency of incinerators and to successfully design future incinerators. Efficiency of an incinerator is calculated by establishing energy and material balances between the flow of materials in and out of the incinerator. Heat of combustion values of these various incineration substances are used in energy balances, and ultimate analyses are used in material balances.

Heat of combustion values of solid waste samples are normally determined experimentally (Parr Instrument Co., 1966). However, some materials, especially incinerator ashes, are difficult to analyze. Furthermore, because the experimental data is not compared to other thermal analyses, the accuracy of the heat of combustion values is questionable.

Correlating the data from ultimate analyses into a formula for the heat of combustion determination causes all the data to have more creditability because the formula would produce an unrealistic or uncommon value if some of the ultimate analyses data is in error.

Approach and Results

By definition (Schmidt and List, 1962a), complete combustion of a fuel involves the conversion of all carbon to CO_2 , hydrogen to H₂O, and sulfur to SO₂. The heat of formation values 14096, 60214, 3982 (Handbook of Chemistry, 1967a) represent the heat of formation of 1 mole of CO_2 from graphite carbon, the heat of formation of $1/_2$ mole of water from hydrogen and oxygen gases at constant volume (Schmidt and List, 1962b), and the heat of formation of 1 mole of SO₂ from sulfur, respectively.

This definition is expressed by the following formula:

Btu/lb = 14,096 C + 60,214
$$\left(H - \frac{O}{8}\right)$$
 + 3982 S (1)

Where C, H, O, and S are the decimal percents of total carbon, total hydrogen, total oxygen, and total sulfur found on ultimate analysis. The available hydrogen is estimated by the term [H - (O/8)] (Schmidt and List, 1962a). Available hydrogen is hydrogen that does contribute to the heating value since it is not present in oxygenated compounds.

Since solid waste materials, especially incinerator residues and incinerator fly ashes, often contain carbonate carbon, the above formula is modified to include the endothermic decomposition of calcium carbonate, assuming all carbonates in solid waste samples are calcium carbonates. The total carbon content, determined experimentally (Wilson, 1971), minus the carbonate carbon content, determined experimentally (AOAC, 1970a) equals the organic carbon content. The formula now becomes:

$$\begin{array}{l} Btu/lb \,=\, 14,096 \; C_{\rm org} \,+\, 60,214 \; \times \\ \\ \left(H \,-\, \frac{O}{8} \right) + \; 3982 \; S \,-\, 6382 \; C_{\rm inorg} \ \ (2) \end{array}$$

H in this formula is determined experimentally (Wilson, 1971); O represents the decimal percent of oxygen not combined in carbonates and is determined indirectly from an ashing procedure similar to oxygen analysis for coal and coke samples (ASTM, 1969a).

One procedure of ashing for solid waste samples is the standard method (APWA, 1970) which involves heating the sample to 600°C, but requires testing for the decomposition of carbonates caused by this heating procedure. A preferred ashing technique is to weigh the ash from the carbon-hydrogen method. This preferred method requires very little extra time or effort and eliminates the need for determining amount of carbonate decomposition during heating. Oxygen values for this paper were determined using both ashing techniques.

The heat of reaction value, 60,214, is for 1/2 mole of liquid water being formed from hydrogen and oxygen gases. Since the hydrogen and oxygen contents of the sample are not gases but are bonded to other elements, Formula 2 must have additional terms. Assuming all hydrogen is bonded to carbon, the energy released in the formation of hydrogen gas is 8929 Btu/lb (Glasstone, 1960). Since hydrogen in the formula is an H and not H₂ the available hydrogen concentration is divided by 2 before multiplying by 8929.

The energy released in the formation of oxygen gas is determined similar to the above for hydrogen gas and found to be 4274 Btu/lb.

Adding two new terms, the formula becomes:

$$\begin{split} Btu/lb &= 14,096 \ C_{org} + \ 60,214 \ \times \\ & \left(H - \frac{O}{8}\right) + \ 3982 \ S - \ 6382 \ C_{inorg} + \ 8929 \ \times \end{split}$$

$$\frac{\left(H-\frac{O}{8}\right)}{2}+4274\left(\frac{O}{2}\right) \quad (3)$$

The concentration of sulfur in solid waste samples is usually about 0.20% and unless actually determined (ASTM, 1969b) is considered to be 0.20% in this paper.

The term "heat of combustion" generally includes not only carbon, hydrogen, oxygen, and sulfur but also other elements such as nitrogen and chlorine which may be present in the sample (Parr Instrument Co., 1966). Since nitrogen is usually present in solid waste samples, another term is added to the formula to compensate for the oxidation of nitrogen to nitrogen dioxide. The heat of oxidation of the nitrogen is 1040 Btu/lb (Handbook of Chemistry, 1967b). In this paper, nitrogen concentration, unless actually determined (AOAC,

			Ultimat	e analysis			Exptl,	Exptl	Calcd,
Type of sample	% Corg	% Cinorg	% H	% Oorg	% N	% S	Btu/lb	variation	Btu/lb
Raw refuse	45.68	0.25	6.28	35.34	0.54	0.24	8425	50	8397
Raw refuse	46.76	0.42	6.23	39.82	0.44	0.22	8336	109	8238
Raw refuse	46.51	0.65	6.38	37.92	0.60	0.14	8442	46	8397
Raw refuse	46.25	0.50	6.35	38.05	0.67	0.10	8377	50	8340
Raw refuse	45.85	0.83	6.61	35.94	1.03	0.10	8707	21	8705
Raw refuse	43.55	0.72	6.25	33.87	0.75	0.20 ^c	8192	197	8139
Raw refuse	45.74	0.59	6.02	35.54	0.75	0.20^{c}	8199	57	8207
Res. $(fines)^d$	6.87	0.15	0.17	0.00	0.12	0.21	1192	142	1079
Res. $(fines)^d$	3.65	0.07	0.08	0.00	0.04	0.12	609	42	567
Res. $(fines)^d$	1.40	0.65	0.18	0.00	0.15 ^b	0.20 ^c	226	74	282
Res. (fines)d	0.72	0.42	0.10	0.00	0.15	0.20^{c}	167	95	149
Res. $(fines)^d$	7.22	0.70	0.42	2.94	0.15	0.20 ^c	959	168	1078
Res. (org) ^e	15.81	0.64	2.24	11.04	0.75 ^b	0.20 ^c	3394	580	2996
Res. (org)e	11.23	1.24	1.48	8.61	0.75	0.20 ^c	2126	193	1962
Res. (org) ^e	33.67	0.50	3.93	20.55	0.75	0.20 ^c	5940	106	6049
Res. (org)e	33.86	0.14	4.74	27.01	0.75 ^b	0.20 ^c	6020	130	6236
Res. (org) ^e	50.45	0.14	4.68	27.88	0.75 ^b	0.20 ^c	8539	20	8490
Fly ash	3.48	0.94	0.13	0.00	0.15 ^b	0.20 ^c	565	75	524
Fly ash	2.15	1.31	0.17	0.00	0.15 ^b	0.20^{c}	278	57	338
Fly ash	2.04	0.96	0.09	0.79	0.15 ^b	0.20^{c}	335	85	253
Fly ash	3.36	0.15	0.22	0.25	0.15	0.20 ^c	483	144	602
Fly ash	1.04	0.35	0.16	0.23	0.15^{b}	0.20^{c}	188	84	223

Table I. Composition and Heating Value of Prepared Solid Waste Incinerator Samples^a

^a All values are on a dry basis. Each value is the average of at least two individual analyses. A total of 22 samples are from five incinerators located throughout the U.S. ^b Nitrogen value is estimated.

Sulfur value is estimated.

 a Residue "Fines" or "Noncombustibles" are materials remaining after most of the readily combustible substances have been removed by manual sorting. The sorting was performed at the incinerator site and a 1/2-in. sieve was employed to assist in the separation.
 Residue "Organics" or "Combustibles" are mostly the readily combustible materials. Unlike "Fines" these materials are readily retained on a 1/2-in. sieve

1970b), is assumed to be 0.75% for raw refuse samples and 0.15% for incinerator residue samples and fly ash samples.

For the formula, N represents the decimal percent of nitrogen content in the sample.

Chlorine, although present in solid waste samples, is not considered a factor in the heat of combustion calculations. Considering the general concentrations of chlorine and sulfur, the overall reaction of the chlorine products is a net thermochemical effect which is almost nil (Barker et al., 1955).

The final formula for solid wastes samples is as follows:

$$Btu/lb = 14,096 C_{org} - 60,214 \times$$

$$\left(H - \frac{O}{8}\right) - 3982 \text{ S} - 6382 \text{ C}_{inorg} - 8929 \times \frac{\left(H - \frac{O}{8}\right)}{2} - 4274 \left(\frac{O}{2}\right) - 1040 \text{ N} \quad (4)$$

This final formula has been applied to some prepared solid waste samples (Cohen and Allen, 1970) from various incinerators (Table I). The Btu/lb contents of these same samples have also been determined experimentally (Parr Instrument Co., 1966). The Btu/lb variation between experimental values and calculated values has about the same magnitude as variation between replicate Btu/lb experimental values. The Btu/lb of incinerator residue samples and fly ash samples are difficult to analyze. The reproducibility of data on these types of samples is poor and the formula is very helpful by presenting another technique to verify the average Btu/lb value.

Conclusions

A formula for calculating Btu/lb of prepared solid waste samples from ultimate analysis now exists. Employing this formula allows either the Btu/lb value not to be determined experimentally or more confidence to be put in the accuracy of the experimental Btu/lb value. The degree of accuracy of mathematical Btu/lb values is sufficient for employing these values in energy-balance formulas about an incinerator.

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Particles Containing Lead, Chlorine, and Bromine **Detected on Trees with an Electron Microprobe**

Gary H. Heichel¹ and Lester Hankin

Departments of Ecology and Climatology and Biochemistry, The Connecticut Agricultural Experiment Station, Box 1106, New Haven, Conn. 06504

Analysis of the bark of trees with an electron probe microanalyzer revealed that lead, chlorine, and bromine are associated with particles, about 7 µm in diam, on or embedded in the bark. The similarity in elemental content is noted between the lead-bearing particles on trees and some of the compounds emitted in automobile exhaust.

he lead concentration on or in plants near highways suggests that the lead burden of plants is increased principally by deposition onto plant surfaces of lead-bearing residues from vehicle exhausts (Cannon and Bowles, 1962; Motto et al., 1970; Smith, 1971). The principal constituents of lead compounds in vehicle exhaust are the lead, chlorine, and bromine from the additives in gasoline (Hirschler and Gilbert, 1964). However, the form of the residues on plants has not been unequivocally demonstrated. We now report that at least a part of the lead on or embedded in the bark of trees growing near roads is combined with chlorine and bromine in particles similar in size and composition to particles in automobile exhaust.

Experimental Procedure

Branches 5 mm in diam of the current year's growth were sampled from limbs 1.75 meters above the ground on a white pine (Pinus strobus) growing in a dense roadside screen 2 to 3 meters west of the curb of a densely and speedily traveled highway in Hamden, Conn. Bark samples that averaged 4 mm in thickness were collected 0.75 meter above the ground from an elm (Ulmus americana) growing 1 meter from the curb of a densely but slowly traveled city street adjacent to the green in downtown New Haven, Conn. The pines had been established for 25 years, but the elm was transplanted to the downtown location 3 years before sampling.

The total lead content was determined on oven-dry subsamples by atomic absorption spectrophotometry. The yearold pine twig contained 120 µg lead/g dry wt and the 3-yearold elm bark contained 1080 µg lead/g. The total lead in the year-old pine twig is less than in the 3-year-old elm bark probably because of the briefer exposure of the pine twig to contamination. Analysis of total lead, however, does not reveal the form and composition of the lead compounds deposited on the surface.

For analysis with an electron probe microanalyzer (Sawhney and Zelitch, 1969; Ter Haar and Bayard, 1971) sections of surface bark 3 mm square and 1 mm thick were cut with a clean razor blade, compressed between two sheets of filter paper, and dried for 48 hr over silica gel. The sections were then mounted on a carbon block with Apiezon N stopcock grease, covered with a thin carbon coating in a vacuum evaporator, and stored in a desiccator. Individual spectrometers were tuned at the wavelengths corresponding to the K_{α} Xrays of bromine, the K_{α} X-rays of chlorine, and the L_{α} X-rays of lead. The microprobe was operated at a beam voltage of 25 kV and a sample current 100 nA on brass.

Particles containing lead were located and identified by scanning the surface of the bark with a 1-µm-diam spot beam at an instrument magnification of 250× until a rapid count rate was detected on the lead spectrometer. Areas of bark containing lead were scanned at an instrument magnification of $1000 \times$ with a 1-µm-wide electron line, and the charac-

¹ To whom correspondence should be addressed.

Location	Sample	Diam, µm
New Haven	1	6
	2	5
	3	10
	4	5
	5	5
	6	3
Mt. Carmel	7	9
	8	13
	9	5
	10	6
Mean \pm s.e.		6.7 ± 1

teristic lead X-radiations were displayed on an oscilloscope. The electron image displayed on the cathode-ray tube of the oscilloscope was photographed to facilitate measurement of the dimensions of the particles.

Results and Discussion

Particles containing lead held on the rough surface or embedded in the surface cells of the bark were found on both pine and elm. Within ten regions selected from bark of the two trees, the diameter of the particles that we examined containing lead ranged from 3 to 13 μ m, with a mean of 6.7 \pm 1 µm (Table I).

Particles on the 3-year-old elm bark were apparently poorer in lead than those on the year-old pine twigs. The count rates of particles on pine and elm having comparable dimensions were 4800 to 10,500 counts per 30 sec on the pine and 1200 to 2250 counts per 30 sec on the elm. These observations suggest that a few of the large particles on the elm bark that had been exposed for 3 years might be more strongly weathered than those on the pine.

Lead, chlorine, and bromine in particles were analyzed by focusing the spot electron beam and simultaneously measuring on three spectrometers the emitted X-radiations as counts per 30 sec. Background measurements were made on areas of bark devoid of lead particles. The levels of chlorine and bromine associated with lead in the particles were significantly above background (Table II). The count rates for lead are lower than those reported above because an alternate spectrometer was used. Although the lead content of the particles that we examined varied over fourfold, the ratio of lead/chlorine/bromine among the particles was similar except

Table II. Analysis of Particles on Bark of Pine

	Eleme	ent, counts/30	sec
Sample	Pb	Cl	Br
1	2040	443	999
2	2305	350	927
3	2511	449	908
4	2647	370	1063
5	1078	314	657
6	599	316	587
7	2113	379	862
Mean \pm s.e.	1899 ± 160	374 ± 21	858 ± 66
Background:	66 ± 7	258 ± 18	306 ± 11
Mean \pm s.e. of			
10 samples			

for sample six. Compounds of the elements that we observed in particles on the bark of trees are also present in automobile exhaust (Hirschler and Gilbert, 1964; Ter Haar and Bayard, 1971). Other lead-bearing compounds like oxides and carbonates have been detected in automobile exhaust (Ter Haar and Bayard, 1971), but the presence of those compounds in particles on vegetation has yet to be confirmed. Knowledge of the form and composition of lead residues on or in plants is clearly important in determining how lead affects plants. In humans, for example, some forms of lead are more toxic than others (Bryce-Smith, 1972). We have now shown that lead retained on or embedded in plant surfaces is associated with at least two other elements, chlorine and bromine.

Acknowledgment

We thank Edward Weber, Geology and Geophysics Department, Yale University, for his assistance with the electron microprobe and Richard Botsford, Analytical Chemistry Department, The Connecticut Agricultural Experiment Station for the atomic absorption spectrophotometry.

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Distribution of Mercury in the Sediments of New Haven (Conn.) Harbor

M. D. Applequist, A. Katz, and K. K. Turekian¹

Department of Geology and Geophysics, Yale University, New Haven, Conn. 06520

Distribution of mercury in sediments of New Haven Harbor indicates that the primary supply is from municipal sewer outfalls in the harbor.

H arbors such as New Haven's are subject to maintenance and developmental dredging. The quality of the dredge spoils is a concern of all who must decide the method of disposition of this material. The metals as a group have been subject to scrutiny because of the known toxic effects of large doses not only on marine life but humans as well. Of these none has received as much attention as mercury. In this report we attempt to assess the distribution and levels of this element in the sediments of a typical industrial harbor and identify the probable sources of the high concentrations where encountered.

Methods

Most of the samples were collected aboard a small boat using a 15-cm-long gravity corer with a polyvinyl chloride (PvC) core liner 4.8 cm in diam. For water depths shallower than about 1 meter, cores were collected using manual depression of an aluminum tube. The cores so obtained were immediately transferred to PvC tubes. A few cores obtained by piston coring from a larger craft and made available to us through the courtesy of the United Illuminating Co. were also analyzed. A detailed environmental impact study of the United Illuminating power plant construction site describes these cores (Turekian et al., 1972).

Segments of the cores to be analyzed were disaggregated, homogenized, and heated at 90–100°C for two days. (Experiments showed no discernible difference between air-dried splits and the heated splits.) Smaller aliquots of the samples were then pulverized and analyzed for mercury generally following the cold atomic absorption spectrophotometric technique of Hatch and Ott (1968) but with the following major modifications: After the acid digestion and permanganate oxidation steps, the insoluble silicate residue was separated from the solution to prevent loss of the mercury by adsorption on the particles during the reduction stage. The absorption cell was wrapped with a heating tape to prevent water condensation on the quartz windows instead of using an in-line desiccant.

The formal reagent blank, determined each day, gave a value equivalent to 0.03 ppm (excluding one bad determination out of nine of 0.14 ppm) and a std dev of 0.06 ppm. Each sample was analyzed once. Where the concentration of mercury was excessively high so that off-scale readings were obtained, a smaller aliquot of the sample was rerun.

Results and Discussion

Generally, the top 8-cm portion of each core was analyzed. Two samples in the Long Wharf area on the west side of the harbor were homogenized over the 15-cm length of the core prior to analysis. Samples analyzed with depth in the cores gave variable results (Table I).

The results for the tops of the cores are plotted in Figure 1 together with the indication of the boundary between the clayey-silty sediments and the sandy. A correlation of mercury with the more organic-rich clayey-silty fraction rather than the sandy one is indicated in New Haven Harbor as it is in several other studies (McColloch et al., 1971; Kennedy et al., 1971). The more important feature is the direct correlation of the highest mercury concentrations with the location of the major municipal sever outfalls. This was shown to be the

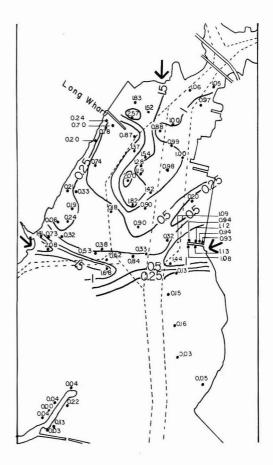


Figure 1. Distribution of mercury (in ppm) in the tops of sediment cores from New Haven Harbor

Long Wharf is on the west side of the harbor. Outfalls of the municipal sewage treatment plants are indicated by arrows and the association with high mercury concentration with them is seen. Sandy areas are found close to shore on the west side of the harbor (from Long Wharf southward) and all sediments south of the sewer outfall on the eastern side of the harbor. Dashed lines outline channel and anchorage

¹ To whom correspondence should be addressed.

Table I. Difference in Mercury Concentration (in ppm)
Between Tops and Bottoms of 15-cm Gravity Cores from
the Long Wharf Area

0–8 cm		8–15 cm	Difference between top and bottom
0.24		0.24	0
0.78		1.07	-0.29
0.74		0.25	+0.49
0.19		0.08	+0.11
0.06		0.08	-0.02
0.73		0.27	+0.46
1.35		2.40	-1.05
0.32		0.14	+0.18
0-5 cm	5-10 cm	10-15 cm	
2.55	1.12	0.52	+2.03

case by Klein and Goldberg (1970) for the Hyperion Sewage plant off Los Angeles. Mercury correlates with the organic carbon content of the sediment as do the other metals (Turekian et al., 1972; Kennedy et al., 1971) and the organic carbon content in New Haven Harbor is related directly to the sewer outfall sites. The different patterns of mercury distribution in the sediments around the outfalls are due to the local current systems. There does not appear to be any evidence of an additional source of supply of mercury to the New Haven Harbor sediments other than municipal sewage treatment plants. The New Haven plants have only primary treatment and until recently accommodated both the sanitary and storm sewer burden of the city. We suspect that the high organic carbon and metals, including mercury, in the sediments around the outfalls are due to escape of sewage sludge which settles into the sediment rather than the consequence of high productivity at these points, resulting in a biological transport of the metals to the sediment.

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CORRECTION

Recent Sedimentary History of Lake Mendota, Wis.

In this article by G. C. Bortleson and G. F. Lee [ENVIRON. SCI. TECHNOL., 6 (9), 799–808 (1972)], in Figure 4, the date at the 40-cm level should read 1895 rather than 1875.

Analygas Systems Ltd. (Scarborough, Ont.) will be the Canadian representative for the eCOlyzer CO monitor.

Combustion Engineering has been awarded contracts in excess of \$30 million to provide two 550-MW controlled circulation steam generators to Oklahoma Gas and Electric Co. The boilers will be installed as Units 4 and 5 of the Muskogee Generating Station, 150 miles east of Oklahoma City.

Cordon International Corp. (Los Angeles, Calif.) has been selected by Cincinnati Gas and Electric Co. to design and manufacture a radioactive gas control system for the William H. Zimmer Nuclear Power Station presently under construction in Moscow, Ohio, just east of Cincinnati.

TRC—**The Research Corp. of New England** has been awarded a \$94,000 contract from EPA to study odor measurement technology. SWECO, Inc. (Los Angeles, Calif.) has received a \$138,724 contract from the city of Ft. Wayne, Ind., for the manufacture of eight Model 60-in. centrifugal waste water concentrators and automatic control panels. Contract is part of demonstration project to determine feasibility of screening systems in treating storm/sanitary flow.

Certain-teed Products Corp. has acquired Ellis & Ford Mfg. Co., a distributor of water and sewer products and manufacturer of pipe tools, for an undisclosed sum of cash. Ellis & Ford had sales of nearly \$5 million last year.

NLT Corp. and Alladin Industries, both of Nashville, Tenn., have jointly acquired a majority interest in Environmental Quality Engineering, Inc. (Oakland, Calif.) EQE's claim to fame is the Accelerated Biological Chemical (ABC) System, made available to the company under an exclusive license from the University of California.

industry trends

Peabody Petersen Co., a subsidiary of Peabody Galion Corp., has received a contract for \$4.1 million to produce an advanced sewage treatment plant which will remove better than 98% of the pollutants from waste water for the City of Fayetteville, N.C. Plant should be completed by the fall of 1974.

Wheelabrator-Frye has completed acquisition of Garden City Fan & Blower Co. for approximately 115,000 shares of Wheelabrator common. Garden City fan has annual sales of about \$4 million.

Dow Chemical U.S.A. has acquired Hydroscience, Inc., a leading water pollution control consulting firm of Westwood, N.J. Terms of the acquisition were not disclosed.

Arthur G. McKee & Co. has been awarded a multimillion dollar contract for construction of Armco Steel Corp.'s new solid waste recycling facility at Middletown, Ohio.

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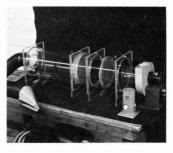
Series 2010 mass flowmeters utilize heat transfer from tubular nickel-sheathed element to measure flow rates of air and other gases. Unit functions from 0-150°C. Response time is 0.1 sec. Thermo-Systems, Inc. 64

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20,000 gph rapidly destroy waterborne bacteria without affecting taste or chemical composition. Operating costs are approximately 1¢/1000 gal. Atlantic Ultraviolet Corp. 72

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Consultants. Firm specializing in consultant services for reduction of natural resources use offers bulletin entitled "The Conservation Approach to Pollution Control." Bataille Assoc. 93

Pumps. Brochure lists company's line of high head, high-capacity pumps for various applications. Allis-Chalmers Corp. 94

Waste treatment. Bulletin describes firm's capabilities in the planning, engineering, construction, and startup of industrial water treatment systems. Illustrated with several photographs of typical installations. Peabody Peterson 95

Carbon regeneration. Case history describes how Vibra Screw Bin Activator promotes positive flow of sluggish, spent powdered carbon for regeneration process. Process is said to be first ever to permit recycling of carbon filtration and purification media. Vibra Screw, Inc. **96**

Compressors. Bulletin describes features of Type MV multistage centrifugal compressors offering pressures up to 10 psig. Bulletin includes special construction features, performance charts for multistage operation, approximate dimensions, and specs guide. Dresser Industries, Inc. **97**

Dust Control. New brochure describesfull line of in-plant air pollution controlequipment including fabric filter dustcollectors and cyclone collectors, mistcollectors, lint collectors, and fume exhausters. The Torit Corp.98

Gas analysis. New bulletin describes use of gas partitioner for fast, economical, and convenient gas analysis. Model 29 uses a dual-column gas detector system which makes possible single-step analysis of complex gas mixtures. Fisher Scientific Co. 99

Environmental services. Brochure lists services of company including consulting services on emission inventories, lab services, review of state and local regulations, process engineering, plant startup assistance, mobile monitoring, and performance evaluation. Betz 100

Impact statements. New brochure, MS72-2, details rapid, economical methods for environmental impact statements. Applies to highways and many other types of construction involving public planning. TRC—The Research Corp. of New England 101

Applications note. Publication describes application of company's total organic carbon analyzer to determining biodegradability of a wide range of organic carbon compounds. Beckman Instruments, Inc. 102

Noise analysis. Bulletin shows value of real-time analysis when applied to measurement of noise. Technique can help engineer to control or reduce noise. General Radio 103

Emissions control. Bulletin describes chemical approaches that have been successfully put into practice for reducing visible emissions from stacks. Apollo Chemical Corp. 104

Data Sheet. Process design sheet gives schematic layout and operating characteristics for an air pollution control system for paper and film coating and saturating lines. The system features both air-to-air and air-to-liquid heat recovery for high-efficiency operation. Midland-Ross Corp. 105

Computer planning. Brochure entitled "Urban and Environmental Technologies" is comprised of three sections: City Planning, Environment, and Energy Systems. Computer Sciences Corp. **106** Tertiary treatment. Booklet discusses centrifugal waste water concentrators which remove most suspended solids from many effluents including those from paper, textile, tobacco, meat packing, potato, and poultry industries. sweco, Inc. 107

Pelletizer. Literature describes an attendant-free, completely automatic system that changes dust to easily disposable pellets. Can be used with existing or planned dust collection system. Hartley Controls Corp. 108

Instrumentation selection. Technical bulletin discusses variables involved in selecting appropriate instrumentation for waste water systems. Photos of typical installations and process flow diagrams illustrate text. Graver Water Conditioning Co. 109

Catalog. New Mikro-Products catalog includes a wide range of equipment for air pollution control and dust recovery, as well as for particle size reduction and separation. MikroPul Div., Slick Corp. 110

Power plants. Reprint of technical paper reviews the status of technology for controlling pollutants from fossil fuelfired power plants. Paper reviews state of the art of particulate collection and control of SO₂ and NO_x emissions. Research-Cottrell 111

Panel design.12-page booklet high-
light's Hach panel design and fabrication
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systems. Hach Chemical Co.112

Published papers. Booklet contains list of papers published by staff members during 1971. Battelle Columbus Laboratories, 505 King Ave., Columbus, Ohio 43201 (Write direct.)

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Environmental Mercury Contamination. Rolf Hartung, Bertram D. Dinman, Eds. ix + 349 pages. Ann Arbor Science Publishers Inc., P.O. Box 1425, Ann Arbor, Mich. 48106. 1972. \$22.50, hard cover.

A comprehensive presentation and analysis of factors in the causes and effects of environmental mercury contamination. Information in the book reassures and dispels the fear that there is total scientific ignorance of the problem, but at the same time, does not allow complacency.

Eater's Digest: The Consumer's Fact Book of Additives. Michael F. Jacobson. xviii + 260 pages. Doubleday & Co., Inc., 277 Park Ave., New York, N.Y. 10017. 1972. \$5.95, hard cover.

Describes over 100 commonly used additives, such as "artificial flavoring and coloring," propylene glycol, EDTA, sodium benzoate, and mono- and diglycerides. Tells how well each chemical has been tested and its effect on human health.

International Environmental Action: A Global Survey. Thomas W. Wilson, Jr. 240 pages. Dunellen Publishing Co., Inc., 145 E. 52nd St., New York, N.Y. 10024. 1972. \$12.50, hard cover.

Assessment of worldwide response to environmental problems from political, legal, social, and economic standpoints. An overview of the actions, reactions, and plans of private and public bodies, both national and international. Includes case studies of what is being done at state and local levels.

Proceedings of the Fifth Mid-Atlantic Industrial Waste Conference. 574 pages. Dean of Continuing and Cooperative Education, Drexel University, Philadelphia, Pa. 19104. 1972. \$15.

Thirty papers presented at the conference held in November 1971. Conference dealt with industrial problems associated with air and water pollution and solid waste disposal.

Water: A Comprehensive Treatise. Vol.
1: The Physics and Physical Chemistry of Water. Felix Franks, Ed. xx + 596 pages. Plenum Publishing Corp., 227
W. 17th St., New York, N.Y. 10011.
1972. \$37.50, hard cover.

Contains contributions written by internationally known authorities in the water field. Stresses the uniqueness of water by comparing it with more normal liquids. The four-volume set, when complete, is intended to serve as a standard reference in the field.

Vector Ecology. World Health Organization Technical Report Series No. 501. 38 pages. World Health Organization, 1211 Geneva 27, Switzerland. 1972. \$1.00, paper.

Concentrates on intensive ecological studies which reveal the quantitative relationships in the vector-pathogen-host system, and surveillance.

Biodeterioration of Materials. Vol. 2. A. Harry Walters, E. H. Hueck-Van Der Plas, Eds. xii + 514 pages. Halsted Press Division, John Wiley & Sons, Inc., 605 Third Ave., New York, N.Y. 10016. 1972. \$49.50, hard cover.

Papers presented at the 2nd International Biodeterioration Symposium held in Holland in 1971. Covers ecology and pollution. Geared to meet the needs of the scientist in research or industry.

Chromatography of Environmental Hazards. Vol. 1: Carcinogens, Mutagens and Teratogens. Lawrence Fishbein. vii + 499 pages. American Elsevier Publishing Co., Inc., 52 Vanderbilt Ave., New York, N.Y. 10017. 1972. \$44.50, hard cover.

Main objective of this four-volume work is to provide the analytical chemist

a practical text as well as a literature source. The present volume centers on toxicants of carcinogenic, mutagenic, and teratogenic environmental significance focusing on pesticides, food and feed additives, drugs, and alkylating agents of industrial utility.

Introduction to the principles of engineering hydraulics and hydrology, as applied in the design of structures and systems for hydraulic developments. This second edition includes two new chapters—one on engineering hydrology and one on groundwater hydraulics. Intended for engineers, geologists, regional planners, and others concerned with water planning, control, and utilization.

The Environment Film Review. 155 pages. Environment Information Center, Inc., Film Reference Dept., 124 E. 39th St., New York, N.Y. 10016. 1972. \$20, paper.

Reference tool for anyone using environmental films. Includes critical reviews of more than 600 films, subject index, industry index, index to sponsors, alphabetical title listing, star rating system, cross-referencing of reviews, coverage of 21 major environmental areas, and names and addresses of distributors.

The Fertilizer Handbook. William C. White, Donald N. Collins, Eds. 208 pages. The Fertilizer Institute, 1015 18th St., N.W., Washington, D.C. 20036.1972.\$4.00, paper.

Underscores the importance of fertilizer by discussing its manufacture, use, and benefits. Includes a chapter on fertilizers and the environment. Eleven authors contribute to the publication.

(Continued on p 1130)

Urbanization and Environment. Thomas R. Detwyler, Melvin G. Marcus. viii + 287 pages. Wadsworth Publishing Co., Inc., Belmont, Calif. 94002. 1972. \$4.95 paper.

Authors aim to demonstrate how man has changed the natural environment by urbanization, to suggest how physical features and processes influence the growth and function of cities, and to reveal some of the feedback between man's actions and environmental processes.

Industrial Processing with Membranes. Robert E. Lacey, Sidney Loeb, Eds. x + 348 pages. John Wiley & Sons, Inc., 605 Third Ave., New York, N.Y. 10016, 1972. \$17.50, hard cover.

Book presents information that will aid the reader in determining whether membrane processes should be considered for a given industrial separation and, if so, which type of membrane process is most promising. Also, basic principles governing membrane processing are covered to aid in the design and operation of membrane processing plants. Fine Dust and Particulate Removal. H. R. Jones. x + 307 pages, Noyes Data Corp., Park Ridge, N.J. 07656. 1972. \$36, paper.

Covers how to reduce fine dust and particulate matter emissions and how to remove particulate matter, preferably at the source. Based on U.S. Government Publication Board Reports and U.S. patents issued since 1960.

The Earth, Our Home in Space. 16 pages. Soil Conservation Society of America, 7515 N.E. Ankeny Rd., Ankeny, Iowa 50021. 1972. \$0.25, paper.

Designed for young people. Relates how the resources of earth provide man with food, fiber, heat, and energy and how man must conserve and manage these resources.

Mercury: A History of Quicksilver. Leonard J. Goldwater. xi + 318 pages. York Press Publishers, 101 E. 32nd St., Baltimore, Md. 21218. 1972. \$15, hard cover.

Primarily a history but is tempered on every page with long experience in

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Political Economy of Environment. 256 pages. Co-Libri, P.O. Box 482, The Hague 2076, Netherlands. 1972. \$6.50, paper.

Social science approach revealing the complex interplay of all interests at stake, digging into the relationship between environmental disruption and the working of social systems, and pursuing the goal of optimization.

Electrodeposition. 3 Vols. Skeist Laboratories, Inc., Livingston, N.J. 07039. 1972. \$950 individual volumes or \$2500 collectively.

Intensive multiclient, technoeconomicmarketing studies directed to new developments and profit opportunities in the coatings industry. Describes different approaches to solve air and water pollution problems.

Water Quality Management: An Analysis of Institutional Patterns. David C. Ranney. xii + 158 pages. The University of Wisconsin Press, P.O. Box 1379, Madison, Wis. 53701. 1972. \$7.50, hard cover.

A case study of the Wisconsin River Basin contrasting the existing regulatory approach to water quality management with the alternative of a systems approach. Purpose of the study was to develop an approach to institutional design from an administrative-political prospective for better utilization of existing water management technology.

Film

The Living Filter. Hornbein-Wood Films, Distributor, Box 174, Lemont, Pa. 16851. 1972. \$200

Reports on a successful experiment at Penn State University proving that treated sewage can be made fit to drink by use of soil as a natural filter. Shows how the system works, enriched crop and tree growth, effects on animals, and laboratory techniques used to analyze water samples.

December 18-22

Florida Technological University

American Association of Professors in Sanitary Engineering Workshop Orlando, Fla.

Topic is mathematical modeling in environmental engineering. Write: Marty Wanielista, Director, Environmental Systems Engineering Institute, Florida Technological University, P.O. Box 25000, Orlando, Fla. 32816

December 26-31 American Association for the Advancement of Science

139th Annual Meeting Washington, D.C.

For more information: D. W. Thornhill, AAAS, 1515 Massachusetts Ave., N.W., Washington, D.C. 20005

January 14–19 Institute of Scrap Iron & Steel

45th Annual Convention and Scrap Industry Exposition

Las Vegas, Nev.

For further information: ISIS, 1729 H St., N.W., Washington, D.C. 20006

January 18–19 Northwestern University and American Chemical Society

39th Annual Chemical Engineering Symposium

Rosemont, III.

Theme "Environmental Impacts of Chemical Engineering." Contact: Jean Kovalcik, The Technicological Institute, Northwestern University, Evanston, III. 60201

January 22-26 Highway Research Board

52nd Annual Meeting

Washington, D.C.

Contact: Hugh Gillespie, Public Information Officer, Highway Research Board, 2101 Constitution Ave., Washington, D.C. 20418

January 27–February 1

National Asphalt Pavement Assoc. 18th Annual Convention

Miami Beach, Fla.

Covers environmental control and other topics. National Asphalt Pavement Assoc., 6811 Kenilworth Ave., Riverdale, Md. 20840

January 29-30 Southern Calif. Chapters of the American Statistical Assoc. and others

Statistics and the Environment Symposium

Los Angeles, Calif.

Sessions dealing with air, population, radiation, and water interconnected with statistics. Write: E. M. Riedinger, Capital Research and Management Co., 611 W. Sixth St., Los Angeles, Calif. 90017

February 7-8

American Water Works Assoc. and others

Fifteenth Water Quality Conference Champaign, III.

Theme is "Organic matter in water supplies: occurrence, significance, and control." Contact: V. L. Snoeyink, Assistant Professor of Sanitary Engineering, 3230 Civil Engineering Bldg., Univ. of III., Urbana, III. 61801

February 12-15 International Snowmobile Congress,

Inc.

Fifth International Snowmobile Congress Sault Sainte Marie, Ont. and Mich.

Topics include land use, noise, and environmental impact. Write: Henry Babbit, Chairman, 5th International Snowmobile Congress, P.O. Box 305, Sault Sainte Marie, Mich. 49783

February 26-27 County of San Diego, Ford Foundation, EPA, and CEQ

Regional Environmental Management Conference

San Diego, Calif.

Will cover technical and management issues relevant to regional environmental management. Write: Patricia Bonner, Public Information Officer, IREM, City of San Diego, 1600 Pacific Hwy., San Diego, Calif. 92101

March 4-9

Engineering Foundation Environmental Engineering in the Food Industry Conference

Pacific Grove, Calif.

Fee: \$175. Contact: Engineering Foundation, 345 E. 47th St., New York, N.Y. 10017

March 5-7

Technical Association of the Pulp & Paper Industry

58th Annual Meeting

Chicago, Ill.

Technical sessions include environmental developments, recycling, energy, etc. Contact: W. B. Manning, TAPPI, One Dunwoody Park, Atlanta, Ga. 30341

March 11-15

American Institute of Chemical Engineers

1973 Meeting New Orleans, La.

Includes session on desalting for water reuse. Contact: AIChE, 345 E. 47th St., New York, N.Y. 10017

meeting guide

March 5-6

University of Louisville and others

Third Annual Environmental Engineering and Science Conference Louisville, Ky.

Will analyze the status of our present environmental problems and present pragmatic solutions. Write: Joseph L. Pavoni, Dept. of Civil Engineering, Speed Scientific School, University of Louisville, Louisville, Ky. 40208

March 13-15 American Petroleum Institute, EPA, and the U.S. Coast Guard

1973 Conference on Prevention and Control of Oil Spills

Washington, D.C.

Covers subjects dealing with administrative, technical, and legal problems concerning oil spills. Contact: J. R. Gould, Secretary, 1973 Conf. on Prevention and Control of Oil Spills, Suite 700, 1629 K St., N.W., Washington, D.C. 20006

March 14-16 Water and Wastewater Equipment Manufacturers Association

Industrial Water and Pollution Conference and Exposition

Chicago, III.

Will focus on the industrial market of water supply, air, water, and noise pollution, and solid waste disposal. Contact: WWEMA, 744 Broad St., Newark, N.J. 07102

March 15-21

National Association of Secondary Material Industries, Inc.

60th Annual Convention

New York, N.Y.

Write: NASMI, 330 Madison Ave., New York, N.Y. 10017

March 18-22

Society of Toxicology

Annual Scientific Meeting

New York, N.Y.

For more information: Robert A. Scala, Secretary, Society of Toxicology, Esso Research and Engineering Co., P.O. Box 45, Linden, N.J. 07036

March 19-23 Ohio State University

Mid-West Workshop in Environmental Science

Columbus, Ohio

Contact: John Lindamood, Dept. of Food Science and Nutrition, Ohio State University, 2121 Fyffe Rd., Columbus, Ohio 43210

(Continued on p 1132)

March 21-24

Utah State University and others

Symposium on Environmental Biogeochemistry Logan, Utah

Logan, Utah

Will emphasize the cooperation of soil scientists, organic geochemists, and biologists in contributing to solving contemporary environmental problems. Contact: J. Skujins, Ecology Center, Utah State University, Logan, Utah 84322

March 26–28 EPA, Cornell University, and National Canners Association

Food Processing Waste Management Conference

Syracuse, N.Y.

Focus will be on results of research and practical applications dealing with waste water management in the food processing industry. Contact: Agricultural Waste Management Program, 1973 Conference Committee, 207 Riley Robb, Cornell University, Ithaca, N.Y. 14850

March 29-30

University of Tennessee and APCA

Third Annual Industrial Air Pollution Control Conference

Knoxville, Tenn.

Contact: Joseph R. Duncan, Program Chairman, University of Tennessee, Dept. of Civil Engineering, Knoxville, Tenn. 37916

April 1-5

Institute of Environmental Sciences

19th Annual Technical/Tutorial Meeting and Equipment Exposition

Anaheim, Calif.

Will provide a forum for exchange between educational, industrial, and governmental activities and to impact U.S. environmental programs with realism. Write: Institute of Environmental Sciences, 940 E. Northwest Hwy., Mt. Prospect, III. 60056

April 1-6

Engineering Foundation

Environmental Needs as Part of the Quality System in Construction Pacific Grove, Calif.

Fee: \$175. Contact: Engineering Foundation, 345 E. 47th St., New York, N.Y. 10017

April 5-9

ASME and others

CIMAC 73—10th International Congress on Combustion Engineering Washington, D.C.

Covers field of internal combustion engines, environmental problems, alternative fuels, etc. Contact: ASME, 345 E. 47th St., New York, N.Y. 10017

April 8-13

American Chemical Society 165th ACS National Meeting Dallas, Tex. Write: A. T. Winstead, ACS, 1155 Sixteenth St., N.W., Washington, D.C. 20036

April 10-12

American Society of Mechanical Engineers

1973 Textile Engineering Conference Charlotte, N.C.

Includes papers on environmental control in the textile industry. Write: ASME, 345 E. 47th St., New York, N.Y. 10017

April 16–18 International Association for Great Lakes Research

Sixteenth Conference on Great Lakes Research

Hiron, Ohio

Covers physical, chemical, biological, engineering, and socio-economic problems of the Great Lakes. Contact: Charles E. Herdendorf, Center for Lake Erie Area Research, 484 W. 12th Ave., Columbus, Ohio 43210

April 22–26 AIChE and EPA

National Meeting on Complete Water Reuse

Washington, D.C.

Sessions will cover legislation and executive action, legal problems, case histories, industry-agriculture collaboration, and other areas. Contact: AIChE, 345 E. 47th St., New York, N.Y. 10017

April 25-27 American Nuclear Society and Marine Technology Society

National Topical Meeting

Palm Beach Shores, Fla.

Theme "The Ocean, Nuclear Energy & Man." Contact: M. Jack Ohanian, Chairman, Dept. of Nuclear Engineering, University of Florida, Gainesville, Fla. 32601

Courses

January 8-10

University of Texas at Austin

Engineering the Control of Air Pollution: Fundamentals Short Course Austin, Tex.

Contact: Engineering Institutes, P.O. Box K, University of Texas at Austin, Austin, Tex. 78712

January 11-12

University of Texas at Austin Engineering Applications of Air Pollution Control Short Course Austin, Tex. Contact: Engineering Institutes, P.O. Box K, University of Texas at Austin, Austin, Tex. 78712

January 15-19

IIT Research Institute Stack Sampling

Chicago, III.

Also will be taught in February and March. Limited attendance. Contact: Ronald G. Draftz, IITRI, 10 W. 35th St., Chicago, III. 60616

January 15-19 University of Tulsa

Recent Advances in Refinery Pollution Abatement

Tulsa, Okla.

Fee: \$225. Contact: Chemical Engineering Dept., University of Tulsa, 600 S. College, Tulsa, Okla. 74104

January 23-25

University of California Extension Center

Groundwater Pollution Short Course San Francisco, Calif.

Write: Continuing Education in Engineering, University of California Extension, 2223 Fulton St., Berkeley, Calif. 94720

February 6-8

Cornell University

Supervision and Inspection of Water, Sewer, and Drainage Installations Short Course

Ithaca, N.Y.

Fee: \$175. Write: Director of Continuing Education, College of Engineering, 252 Carpenter Hall, Cornell Univ., Ithaca, N.Y. 14850

February 8-9

University of Wisconsin—Extension Noise Control in I.C. Engines

Madison, Wis.

Fee: \$95. Contact: University of Wisconsin-Extension, Dept. of Engineering, 432 N. Lake St., Madison, Wis. 53706

February 12-13

Institute on Noise Control Administration

Noise Control

Clearwater, Fla.

Concurrent courses—one for administrators and one for engineers. Contact: James H. Botsford, Director, Institute on Noise Control Administration, P.O. Box 3164, Bethlehem, Pa. 18017

February 15-16

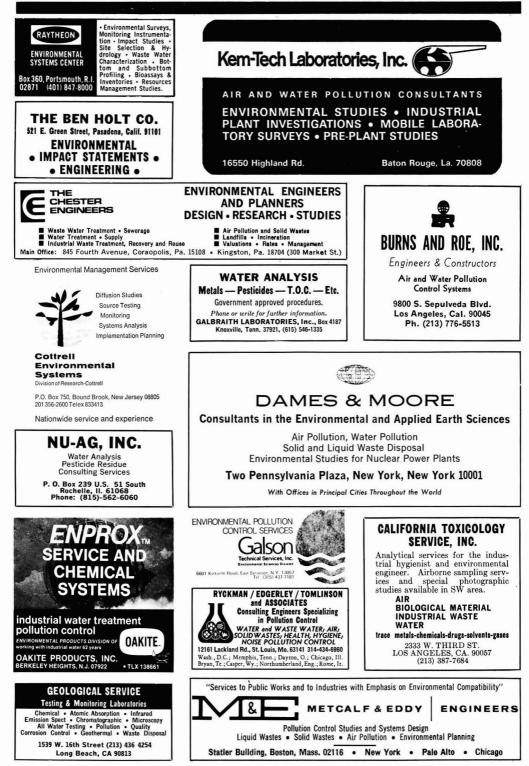
University of Wisconsin—Extension

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Fee: \$95. Write: University of Wisconsin-Extension, Dept. of Engineering, 600 W. Kilbourn Ave., Milwaukee, Wis. 53203

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Volume 6, Number 13

December 1972

January	1–98	July	577-666
February	99-188	August	667-764
March	189–294	September	765–854
April	295-384	October	855-952
Мау	385-474	November	953-1050
June	475–576	December	1051–1148

Pollution Control Directory 1D–162D

Refer to the above listing to determine the issue in which an entry appears.

NAMES INDEX. This index contains the names of ES&T paper and article authors and of people mentioned in staff-written and edited parts of ES&T. If a name is that of an author or coauthor, the page number on which the paper or article starts is in **bold face** in this index. Please note that the titles of papers are not given (they have been in previous years).

SUBJECT INDEX. The basic subject categories are much the same as for Volume 5. The index is recognized to be a relatively shallow one.

All parts of ES&T have been indexed, with the exception of Industry trends, New products, New literature, Bookshelf, and Meeting guide.

NAMES INDEX

A

Abel, Fred, 882 Abrahams, John, 988 Abzug, Bella, 208 Adams, W. A., 928 Adams, W. R., Jr., 114 Adelman, A. H., 933 Adelman, Morris, 388 Adler, Cyrus, 769 Aitken, Bob, 215 Albanese, R. H., 22 Albone, E. S., 914 Alexander, Martin, 732 Alvine, R. G., 22 Anderegg, J. A., 114 Anderson, F. H., 396 Anderson, J. R., 313 Applequist, M.D., 1123 Archer, T. E., 826 Asaro, Frank, 558

В

Badcock, C. C., 72 Bagge, Carl, 28 Baker, Howard, Jr., 208, 303 Bails, J. D., 901 Baldwin, Malcolm, 29, 192 Ballinger, D. G., 130 Banks, W. E. 595 Barber, William, 1065 Barden, C. R., 115 Barker, Bruce, 391 Barnes, G. J., 543 Barrett, Larry, 883 Barth, Ed. 315 Bassham, J. A., 638 Beach, W. A., 313 Beard, C. G., II, 115 Beaver, J. A., 965 Becker, Brooks, 115 Becker, William, 221 Bell, A. T., 1117 Benatsson, Ingemund, 682 Benson, Donald, 307 Berg, K. H., 391 Bernard, Hal, 26 Berry, R. S., 365, 837

Bingham, E. C., 692 Birch, T. J., 224 Bishop, D. R., 622 Black, R. J., 318 Blackwell, Robert, 773 Blanchard, Bruce, 211 Blanche, R. V., 115 Blaser, William, 201 Boden, John, 969 Bordelon, B. R., 1033 Borgwardt, R. H., 350 Borrelli, Peter, 29 Bortleson, G. C., 799 Bosqui, F. L., 518 Boucher, F. R., 538 Boush, G. Mallory, 629 Boushka, Bill, 592 Bove, John L., 617, 1105 Bowen, D. H. M., 7, 103, 197, 393, 583, 673, 771, 861, 961, 1057 Bowen, M. D., 592 Bowman, H. R., 558 Bowyer, James, 199 Boynton, E. B., 115 Braids, O. C., 448 Brennan, Bob, 317 Brezonik, P. L., 719 Briley, Beverly, 780 Brubaker, K. L., 816 Bruce, Robert, 302 Brunner, Carl, 315 Brust, A. T., Jr., 1075 Bryson, J. C., 114 Buckley, D. E., 274 Bufalini, J. J., 816, 837 Bumford, F. H., 115 Burford, M. G., 392 Burgard, Richard, 115 Burns, John, 310 Burton, Philip, 317 Busbee, W. L., 895 Busterud, John, 965 Cahill, W. T., 507

Cahn, Robert, 965

Biglane, Kenneth, 9

Calvert J. G. 72 Campbell, C. L., 114 Carey, William, 219 Carley, G. L., Jr. 1075 Carlin, Alan, 882 Carpenter, Stanley, 1067 Carroll, Thomas, 695 Carver, G. W., 592 Casey, W. J., 303 Cavanaugh, W. T., 23 Chaney, Ed, 210 Chang, S. B., 158 Charam, Philip, 404 Chen, K. Y., 529 Chmielewski, R. D., 1101 Chuah, H. H., 555, 1005 Ciaccio, L. L., 633 Clark, Gene, 317 Clarke, A. J., 671 Clements, J. B., 152 Cliath, M. M., 910 Cohen, Jesse, 315 Cole, Allen, 401 Cole, Criss, 402 Coleman, D. M., 452 Coleman, Emile, 715 Collier, Ledelle, 930 Collom, R. H., Jr., 114 Colonna, Robert, 695 Conn. F. J., Jr., 965 Conway, J. G., 558 Cooper, Charles, 507 Corbalis, J. J., Jr., 1075 Corrado, Frank, 199 Couchman, C. E., 114 Coulter, J. R., 614 Cranmer, Morris, 591 Cranston, R. E., 274 Crawford, Kendall, 769 Cropper, W. V., 23 Crotty, Kenneth, 199 Crow, Larry, 402 Crowe, Robert, 105, 314 Crozier, T. E., 644 Cukor, Peter, 633 Curry, J. H., 114 Cushing, R. L., 391 Cywin, Allen, 786

D

Daley, A. C., 115 Dallas, J. L., 114 Damon, E. K., 72 Dams, Richard, 441 Dante, L. F., 1075 Darley, P. E., 342 Darnay, A. J., 318 David, Edward, 589, 601 Davis, E. N., 161 Davis, Jack, 402 Davis, John, 601 Dea, Stanley, 510 Dean, J. G., 518 De La Rue, R. E., 110 Delves, H. T., 775 Densmore, E. J., 404 Dial. C. J., 318 Dimitriades, Basil, 253 Dillion, Gene, 865 Dinan, F. J., 726 Dingell, John, 208 Di Toro, D. M., 1114 D'Itri, F. M., 901 Dole, Bob. 208 Dominick, D. D., 314 Donaldson, C. duP., 812 Donovan, J. A. K., 1075 Driscoll, J. N., 727 Drivas, P. J., 609 Driver, William, 398 Drobny, N. L., 580 Drobot, Walter, 510 Drucker, Peter, 583 Dubos, Rene, 683 Duce, R. A., 68 Dudley, J. E., 152 Dutriau, Robert, 26 Dwyer, J. B., 21 Dyer, John, 315 Dyer, N. H., 1075 E

Eastland, Mike, 402 Eastman, Paul, 1074 Eggers, A. J., Jr., 508 Eglinton, Geoffrey, 914 Eiguren, A. J., 114 Elia, V. J., 830 Ellington, C. P., Jr., 1017 Ellender, Allen, 209 Elston, John, 785 Engelkemeir, A. G., 338 English, John, 315 Enrione, R. E., 1025 Erickson, Ron, 859 Ernst, Lloyd, 115 Eschwege, Henry, 404 Ettinger, H. J., 614 Evans, Melvin, 1065 Evans, N. C., 914 Evans, R. J., 901

Fairless, B. J., 1036 Falls, C. P., 135 Fan, L. T., 1085

Farkas, E. J., 1116 Farr, W. D., 965 Fassinger, W. P., 1109 Feairheller, Steven, 679 Feder, W. A., 427 Fedrick, N. A., 1117 Feige, W. A., 260 Feldstein, Milton, 250 Ferguson, R. W., 965 Fiarman, Sidney, 79 Fine, D. H., 348 Finelli, V. N., 740 Fisher, L. M., 1075 Fochtman, E. G., 331 Foster, D. H., 872 Francois, Pedrito, 115 Franey, G. J., 43 Franken, P. A., 125 Frankiewicz, T. C., 837 Frankiewicz, Theodore, 365 Freeman, G. O., 115 Freeman, Orville, 681 Freeman S. D. 601 Freese, P. V., 1075 Fri, Robert, 789 Friedlander, Sheldon, 863 Friedman, Mendel, 457, 745 Friend, J. P., 173 Fulgham, Floyd, 203

G

Gaber, Al. 4 Gandrud, Bruce W., 455 Garber, M. J., 71 Garnham, Harry, 309 Gasiewicz, Thomas A., 726 Gay, Bruce W., Jr., 816 Gerhardt, Paul, 883 Gerstacker, Carl, 393 Gesell Gerhard 514 Gianacakes, Peter, 17 Gilmore, Arthur, 316 Gleason, T. W., 965 Gloer, Elmer, 697 Goddard, M. K., 1075 Goetz, Alexander, 143 Goldwater, Leonard, 201 Goodman, Brian L., 794 Goodsel, Arthur J., 268 Goolsby, D. A., 122 Goranson, Stephen, 1019, 1025 Gordon, Eugene, 303 Goren, S. L., 1101 Gottschalk, Robert, 307 Gottwald, F. D., Jr., 307 Graber, Ralph, 698 Graff, R. A., 1009 Greenfield, Stanley, 121, 314, 318, 392, 589, 601, 882 Gregg, Charles, Jr., 789 Gregoire, J. D., 596 Gregor, C. D., 278 Greve, L. C., 71 Griesbach, R. C., 1116 Griffing, Lu, 697 Griffiths, Martha, 681 Grundy, Richard, 300

Guckert, William, 28 Guenther, George, 483, 684 Gutenmann, W. H., 451 Gutow, Bernard S., 790

н

Hale, Samuel, Jr., 318 Hall, Freeman, 585 Hall, S. K., 31 Hamilton, S. D., Jr., 580 Hammond, M. W., 732 Hankin, Lester, 1121 Hanna, Thomas, 112 Hannah, Sid, 315 Hanson, D. N., 1117 Haring, R. C., 602 Harper, Richard, 992 Hart, F. D., 205 Harvey, R. D., 350 Hauser, T. R., 549, 890 Havlick, Robert, 315 Hawes, F. B., 671 Hawkins, D. G., 514 Hay, Kenneth, 315 Hayes, Charles, 402 Hayes, Robert, 115 Healy, M. L., 243 Hechler, Ken, 28 Hecht, T. A., 47 Heichel, G. H., 1121 Hekimian, Kenneth, 205 Hensel, Earl, 1014 Henwood, R. W., 367 Herbert, Bill, 315 Hershaft, Alex, 194, 412 Hickman, H. L., Jr., 318 Hill, J. A., 318 Hill, R. B., 122 Hillman, R. E., 224 Hilst, G. R., 812 Hindenlang, A. W., 219 Hinesly, T. D., 448, 871 Hirsch, L. V., 874 Hirschhorn, M., 404 Hodges, H. E., 115 Hoffman, G. L., 68 Holt, B. D., 338 Holty, J. G., 835 Hope, Malcolm, 1075 Howe Bobert 697 698 Howe, Sydney, 192 Howson, R., 22 Hsieh, D. P. H., 826 Hsu, D. Y., 645 Hughes, Robert, 979 Hume, E. T., 507 Hunter, C. B., 922 Hunter, J. M., 914 Hutton, Michael, 1116 1 Idler, D. R., 982 Ikard. Frank, 15 Innes, W. B., 710 Isberg, Charles, 878 J Jackson, G. E., 72

Jackson, Henry, 29, 199, 208 Jacobson, Robert, 199 Jamieson, Bill, 317 Jefferson, Louis, 199 Jenkins, David, 280 Jenkins, R. E., 884 Jensen, Eugene, 314, 786, 1075 Jessee, Randall, 199 John, M. K., 555, 1005 Johnson, C. J., 1074 Johnson, L. B., 405 Jones, L. A., 821 Jones, P. W., 933 Jones, William, 17 Jordan, B. P., 965 Jorden, R. M., 392 Jores, S. P., 959 Joselow, Morris, 969

Kamphake, L. J., 260 Kanazawa, K., 638 Kanazawa, T., 638 Kardos, L. T., 872 Katz, A., 1123 Kaye, Jim, 1072 Kearney, P. C., 1017 Kemp, C. C., 21 Kennedy, S. R., 397 Kessick, M. A., 642 Kessler, J. E. 1105 Key, Marcus, 684 Kindinger, J. I., 71 Kittredge, George, 222 Klatt, L. N., 452 Klein D H 560 Kleinot, O. J., 143 Kleopfer, R. D., 1036 Klimisch, R. L., 543 Knight, W. E., 115 Kolk, Franklin, 221 Kollar, K. L., 979 Kolpack, R. L., 833 Kometani, T. Y., 617, 1105 Kopczynski, S. L., 342 Kothny, E. L., 250 Krenkel, Peter, 692 Kretchmer, Jerome, 485 Krieger, Barbara, 742 Kugelman, Irwin, 315 Kummler, Ralph, 742

ĸ

La Graff, M. B., 786 Lancy, L. E., 687 Langdon, W. M., 905 Langford, C. H., 367 Lanning, E. W., 633 Lanouette, K. H., 518 Larsoen, Lars, 398 Larson, D. M., 4 Laseter, J. L., 1033 Laufer, D. A., 427 Lazrus, A. L., 455 Lear, Bill, 311 Lear, W. P., 203, 517 Lee, Edward, 199 Lee, G. F., 538, 799, 1031 Lee, R. E., Jr., 929, 1019, 1025 Lee, T. P., 919 Lesher, R. L., 1078 Levaggi, D. A., 250 Levin, A. A., 224 Levin, Alan, 965 Levine, G. V., 280 Levy, Arthur, 423 Lewandowski, G. A., 169 Lewis, H. V., 504 Lindgren, J. L., 922 Lindsay, John, 967 Linford, H. B., 169 Linsky, Benjamin, 859 Lipeles, Martin, 360 Lipke, Herbert, 427 Lisk, D. J., 397, 451 Liskowitz J W 43 Lochboehler, C. M., 821 Lombardo, L. V., 513 Lonneman, W. A., 342 Loomis, T. C., 1105 Low, M. J. D., 268 Luke, C. L., 1105 Lussier, Denis, 315 Lyon, Lee, 594 Lyons, D. W., 37, 391

MacDonald, Gordon, 483, 773, 965 Maga, J. A., 114 Magee, W. S., Jr., 821 Magyar, Martha, 617 Malina, J. F., 438 Malki, Mazen, 742 Malmuth, Norman, 360 Mandancy, Robert, 315 Mandel. Marvin, 1075

Marcus, Martin, 775 Margolis, Geoffrey, 727 Marienthal, George, 211 Mark, H. B., Jr., 833 Masri, M. S., 745 Masse, A. N., 260 Masse, Art, 315 Masselli, J. W., 391 Masselli, N. W., 391 Masterton, W. L., 919 Mathewson, J. H., 644 Matos, C. A., 115 Matson, R. S., 158 Matsumura, Fumio, 629 Matteson, M. J., 895 Mattson, J. S., 833 McCabe, Robert, 697 McCormack, Mike, 601 McCutchen, G. D., 884 McDonald F H 423 McDonald, Louis, 300 McGowan, F. E., 826 McInnis, Noel, 697, 698 McKaveney, J. P., 1109 McKelvey, Vincent, 120, 965 McKewen, T. D., 324 McMahon, Thomas, 22 McWhirter, J. W., 878 Meier, S. W., 1075 Menden, E. E., 740, 830 Mendenhall, I. F., 965 Mercer, T. T., 1091 Messinger, William, 866 Meyer, W. R., 480 Michael, L. W., 830 Middlebrooks, E. J., 979 Miller, G. R., III, 389 Miller, R. H., 872 Milner, Maclin, 397 Minear, R. A., 431 Minnick, L. J., 874 Minor, Paul, 315 Minto, W. L., 517 Mitchell, Balph, 809, 1096 Molina, J-A, E., 448 Moore, F. L., 525 Morel, Francois, 58 Morgan, G. B., 1025 Morgan, J. J., 58, 642 Morris, J. C., 529 Morton, Rogers, 863 Moss, O. R., 614 Moyers, J. L., 68 Mueller, R. F., 4 Mumford, R. A., 427 Munnecke, D. M., 826 Munroe, W. A., 115 Muskie, E. S., 585 Mustoe, G. E., 158 Mvers, E. A., 872 Myers, W. G., 369

N

Naik, P. P., 905 Natarajan, O. Y., 114 Nathanson, Benjamin, 617, 1105 Nekomoto, R. S., 114 Nelson, Gaylord, 28, 123 Ness, Howard, 701, 993 Newill, Vaun, 589, 882 Nixon, R. M., 103, 199, 208, 585, 591, 683, 684, 779, 863, 1059, 1065, 1068, 1071 Nobe, Ken, 239 Nolan, W. F., 37, 391 Noonan, L. W., 778

0

O'Boyle, Thomas, 868, 976 Odum, E. P., 507 Ogilvie, Richard, 11 Okey, R. W., 602 Okun, D. A., 480, 671, 958 Olsen, Marvin, 697 Onnen, J. H., 994 Ordin, L., 71 O'Sullivan, D. A., 682

P Pack, Donald, 675 Page, D. G., 125 Parizek, R. R., 679, 872 Parsons, W. A., 976 Partee, F. P., 114 Pascal, C. C., 21 Patil, K. C., 629 Patterson, H. M., 115 Pattison, J. N., 549 Patton, E. L., 398 Patton, V. D., 114 Payne, Howard, 697 Payne, J. S., 922 Peckham, Alan, 510 Petering, H. G., 740, 830 Peterson, J. R., 871 Petrie, Finlay, 696 Phelps, E. R., 28 Philhower, William, 881 Pieper, A. W., 977 Pipes, W. O., 645 Pircon, Ladd, 865 Plass, G. N., 736 Poirrier, M. A., 1033 Poland, J. F., 122 Pope, Bruce, 588 Porter, J. J., 37, 391 Potworowski, H. S., 835 Pou, Charles, 199 Powers, W. A., 1075 Pradzynski, A. H., 922 Pratt, George, 695 Pratt, J. H., 673 Pressley, T. A., 622 Pritchard, C. B., 390 Pritchard, Edward, 517 Proulx, L. J., Jr., 114 Purcell, T. C., 715 Purdue, L. J., 152

0

Quarles, J. R., 514, 786, 1059

R

Rabinowitz, M. B., 705 Radway, J. E., 959 Rahm, K. A., 441 Raihle, J. A., 621 Raines, G. E., 224 Rall, David, 589 Reinhard, Donald, 879 Reinhardt, G. W., 72 Renfro, W. C., 1001 Retzloff, D. G., 1085 Rhead, M. M., 914 Rhodes, J. R., 922 Ricci, Roy, 510 Rihm, Alexander, Jr., 115 Roan, S.,G., 622

Robbins, J. A., 164 Roberts, J. J., 114 Robertson, J. H., 22 Robinson, Aubrey, 105, 786 Rockwell, W. F., Jr., 205 Roebuck, Gordon, 590 Rogers, C. J., 715 Rogers, Lester, 211 Rome, Samuel, Mrs., 965 Rosenberg, Fred, 406 Ross, W. D., 155 Rother, J. P., 404 Rubino, R. L., 633 Ruckelshaus, W. D., 207, 303, 395, 483, 512, 599, 965 Ruger, P. H., 194 Ruth, L. A., 1009 Rvan, Jim, 502 **Bylands, Bobert, 866**

S

Sadow, Ronald, 122 Sagik, B. P., 438 Saiger, Howard, 114 Samuelson, A. T., 404 Santaniello, R. M., 21 Scarpino, P. V., 715 Schachter, David, 1116 Schell, N. E., 114 Schindewolf, U., 679 Schleyer, W. L., 302 Schneier, Gary, 79 Schomaker, Norbert, 410 Schork, John, 16 Schueneman, J. J., 114 Schultz, Walter, 406 Schweiker, R. S., 28 Sebastian, Frank, 976 Seinfeld, J. H., 47 Serdoz, Richard, 115 Shaffer, C. B., 789 Shair, F. H., 609 Shannon, E. E., 719 Shapiro, Joseph, 280 Shell, H. D., 115 Shy, C. M., 890 Sidebottom, H. W., 72 Siebenberg, Stanley, 617 Sieafried, R. E., 21 Sievers, R. E., 155 Silverman, Mark, 199 Simison, Herbert, 199 Simmonds, P. G., 609 Siu, Wayman, 250 Skully, R. P., 220 Slote, Lawrence, 672, 958 Smith, C. F., 929 Smith, H. D., 115 Smith, J. M., 260, 315 Snitz, F. L., 164 Sober, C. A., 438 Soet, J. C., 114 Sopper, W. E., 872

Sorensen, L. L. C., 239 Southall, Jarrell, 114 Spencer, W. F., 910 Spielman, L. A., 1096 Spink, J. D., 22 Spino, D. F., 715 Squires, A. M., 1009 Sridharan, Nagalaxmi, 1031 Staats, E. B., 303, 405, 585 Stara, Jerry, 590 Stallman, Robert, 121 Stans, Maurice, 103 Stephan, David, 314 Stephens, N. T., 173 Stephenson, H. A., 21 Sternberg, Arnold, 1075 Stivers, D. A., 1109 Stober, Werner, 1091 Stringer, H. L., 895 Strong, Maurice, 682 Sundin, R. E., 115 Sussman, V. H., 115 Sutterfield, F. D., 342 т

Takezawa, Nobutsune, 268 Talty, J. T., 318, 409, 992 Taylor, E. W., 400 Taylor, G. J., 112 Taylor, O. C., 71 Terry, S. L., 513 Teuscher, Lynn, 1071 Thompson, C. R., 1014 Thompson, R. J., 152 Tihansky, Dennis, 882 Tomlinson, R. D., 1001 Touhill, C. J., 784 Train, Russell, 9, 39 682, 789, 882, 1059 395, 589, 675, Trott, Trevor, 367 Trygg, J. E., 114 Tucker, Fred, 119 Turekian, K. K., 1123 Turk, Amos, 602

11

Udall, Stewart, 309, 580, 673 Uhlenhaut, Rudolph, 516 Umscheid, Cliff, 1075

Vacker, Donald, 402 Valentinetti, R. A., 115 Van Atta, R. E., 452 Vanderpool, W. O., 1085 VanLaerhoven, C. J., 555, 1005 Varga, L. P., 135 Varro, Stephen, Jr., 971 Venters, Adolph, 338 Volpe, John, 588 Von Frank, A. J., 216 Vuceta, Jasenka, 642

Wachsmuth, William, 313 Waddell, Thomas, 883 Wagner, A. J., 489 Waiss, A. C., Jr., 457 Waite, T. D., 1096 Wake, B. F., 115 Walker, Peter, 1076 Wallace, George, 778 Wallen, L. L., 161 Walsh, Fraser, 809 Ward, Barbara, 683 Wasan, D. T., 905 Watkins, Clyde, 115 Watts, Joe, 402 Webb, H. J., 115 Weiss, H. V., 644 Wetherill, G. W., 705 Whitmore, S. A., 71 Whitten Jamie 209 Wiik, E. M., 114 Wilkenfeld, Jerome, 217 Willard, Beatrice, 965 Williams, Harry, 114 Williams, Sam, 517 Williams, T. F., 318 Willis, M. T., 114 Wilson, D. L., 1119 Wilson, W. E., Jr., 423 Winchester, J. W., 441 Wingerter, Eugene, 409 Winn, G. S., 115 Wnek, W. J., 331 Wolman, Abel, 4, 776 Wood, G. P., 114 Woods, R. J., 404 Woolson, E. A., 1017 Worsley, P. K., 769 Wright, Ray, 26 Wright, T. E., 113

Y

Yamamoto, Sachio, 644 Yee, W. C., 232 Young, Harold, 876 Youngs, W. D., 451 Yu, Ta-Ching, 833

7

Zadrozny, M. G., 965 Zengel, Alan, 679 Zirino, Alberto, 243 Zoller, W. H., 68 Zurn, E. F., 495 Zurn, Frank, 495, 979 Zurn, J. A., 495 Zurn, M. A., 495

w

SUBJECT INDEX

Acid mine drainage detection by conductivity mea-surement, 587 interception of groundwater, 679 Kentucky control regulations, 305 role of bacteria in formation, 809 Advanced waste treatment status of physical-chemical pro-cesses, 984

A

Aerocolloids

- effect of uv, humidity, nucleating matter, and ammonia on, 152 formation and degradation by uv radiation, 143
- instrument for measurement of, 143

Aerosols

- density measurement with a spectrometer, 614 neutron analysis activation spec-
- troscopic technique, 441 removal with fibrous filter mats, 821

Agricultural wastes

- feedlot controls in Montana, 774 Minnesota to control feedlots, 586 pollution from leather tanning, 594 power from the peanut, 592 Aircraft
- noise control, 124 noise and emission control. 220
- Air monitoring network cascade impactor network first year results, 1019
- Air pollutants
- fate of materials in the liquid phase, 933 Air pollution
- see more specific listings Air pollution control basis of new source performance standards, 885
- clean magnesium-from-aluminum removal process, 865 from coke ovens, 118
- at Ford Motor Co. foundry, 110
- NO_x reduction from oil combus-tion, 109
- odor reduction processes, 602 reducing aircraft emissions, 220 Research-Cottrell Co. business
- 16 review of SO2 removal technolo-
- gy, 688 small precipitator for apartment houses, 587
- survey of state program, 111 Air quality modeling
- NEXUS program, 1071
- Air quality monitoring around the states, 113 ASTM's Project Threshold, 23 automated system, 110 CO concentrations. 397 NOAA program, 675 NO measurement, 969 NO_x measurement, 890 statewide system in Pennsylvania, 586

around the states, 113 EPA approves state implementation plans, 599 EPA has state implementation plans, 395 nondegradation issue, 673 Alabama superport proposed in Mobile Bay, 778 Alaska comments on oil pipeline, 398 Interior views on pipeline, 863 proposed refinery for North Slope oil, 681 sues on tin can dumping, 774 Algae growth rate factors influencing, 1096 Aliphatic materials in air analytical technique for, 549 Allied Chemical Corp. environmental coordinator, 216 phosphate removal tests, 201 Aluminum Co. of America (Alcoa) magnesium-from-aluminum moval process, 865 Aluminum in water analysis by atomic absorption spectrophotometry, 645 Alyeska Pipeline Service Co. Alaskan oil pipeline, 398 American Iron and Steel Institute coke oven emission control, 118 American Petroleum Institute spending for control programs, 15 surveys of waste lube oil, 25 American Society for Testing Mate rials (ASTM) Project Threshold, 23 Analytical methods neutron activation spectroscopy for 35 trace elements in air, 441 Aquaculture, 232 Arizona controls on copper smelters, 485 regulates SO² from smelters, 396 Arsenic in water distillation technique for analysis of 1116 Arthur G. McKee & Co. role in water and waste water treatment business, 974 Asbestos classified by EPA as hazardous air pollutant, 9 Illinois establishes regulations 201 Atlantic City Electric Co. converts from coal to oil, 396 Atomic absorption spectrophotome try for aluminum water analysis, 645 Atomic Energy Commission (AEC) can issue temporary licenses, 585 court rules agency regulations preempt those of states, 396 radioactive discharge data, 105

Air quality standards

Atomics International Co. SO2 removal process, 306, 689 Automotive emissions advanced power systems, 769 API research program, 15 Bromine Bell and Engelhard oxidation catalysts, 776 California requires control devices on old cars, 864 Bureau of Mines, U.S. California to test on assembly line, 774 cars in cities, 769 catalysts tested in California, 1061 effect of lead on control systems, 199 EPA lead-free gasoline regulations, 303 Cadmium exhaust treatment system, 1062 GAO report on EPA capability. 585 highway air quality prediction, 1071 ICI exhaust catalyst, 15 imported autos and standards, 205 lead concentrations in particulate matter, 164 lead from gasoline, 31 lead level and volatility of gaso-lines, 485 natural gas service stations, 588 California NO_x reduction catalyst, 487 Philadelphia fines owners of smoky autos, 11 progress and future, 512 rental car experience, 109 state testing program in N.J., 785 steam turbine vehicle, 203 Volvo requests standards waiver, 398 Avco, Inc. freeze purification process, 487 в Babcock & Wilcox Co. SO₂ removal process, 689 Bacteria role in formation of acid mine waters 809 Bell Labs Catalyst auto exhaust catalyst, 776 Beryllium classified by EPA as hazardous air pollutant, 9 Beryllium in the air in 14 urban samples, 155 gc analytical method for, 155 Bethlehem Steel Corp. clean coking process, 1063 pollution control revenue bonds, 866 B. F. Goodrich Chemical Co. to build new treatment plant with Ohio city, 866 **Biospherics Inc.** PhoStrip process tests, 15 Blueside Co. tanning process, 594 Chemico Breakpoint chlorination physical-chemical treatmen method for nitrogen removal, 622 Chicago

Brewing industry

- water pollution control, 504 in the atmosphere, 68
- from lead halide automobile exhaust particulates, 164
- - ceramics from waste glass, 990 citrate SO₂ removal process, 969 materials recovery from phos-phate sludge, 1062

C distribution in cigarette smoke, 830 effects on oats, 555 Cadmium in the soil factors affecting uptake by plants, 1005 Cadmium in water field polarographic technique for, 452 Calgon Corp. role in water and waste water treatment business, 974 assembly-line testing of auto emissions, 774 bans ocean waste dumping, 677 emission tests for older, resold autos, 864 secondary effluent recycling study, 677 tests catalytic auto exhaust system, 1061 **Carbon adsorption** heavy metals removal, 522 **Carbon dioxide** equilibrium equations for iteration between the sea and atmosphere, 736 Carbon monoxide model for urban air concentrations, 397 nature's contribution to atmosphere 679 for automotive exhaust emission control, 543 Catalytic converter effect of lead on activity of copper oxide-alumina catalyst, 239 for hydrocarbon removal in automobile exhaust, 239 Cellulose waste conversion to protein, 715 Cement plants basis of new source performance standards, 885 Chemfix Corp. sludge-fixing process, 203 Chemical Separations Corp. ion exchange treatment process, 692 SO₂ removal process, 689 Chem-Pure Systems, Inc. sewage treatment process, 397 bans detergent phosphate, 485

data for air and water pollutants, 338 fines noise ordinance violators 774 Chlorine in the atmosphere from lead halide automobile exhaust particulates, 164 Chlorodioxins impurities in 2,4,5-T manufacture 1017 Chlorodioxins in the soil persistence and metabolism of, 1017 Chrysler Corp. requests emissions standards waiver, 398 **Cigarette smoke** presence of toxic metals in, 830 Cleveland saves on solid waste collection 864 upgrades old sewage plant, 782 Clow Corp. role in water and waste treatment business, 974 wate Coal disadvantages of gasification, 388 fluidized limestone process, 865 **Coal industry** conservationist views on strip mining, 192 roundup of strip-mining contro legislation, 27 strip-mining control in Missouri 194 strip-mining regulations in Penn-sylvania, 107 TVA strip-mining regulations, 15 Colorado challenged on air law, 677 emergency evacuation procedure 677 public utility concept for solid wastes, 677 Coloring colloids in water ability to concentrate DDT, 1033 Combustion methods for reactive organics in air, 710 particulate matter size distribution from lignite, 929 Combustion Engineering, Inc. SO2 removal process, 689 **Combustion Equipment Assoc.** SO₂ removal process, 689 Commerce Dept., U.S. atmospheric research program. 675 effect of pollution laws on busi-ness, 1059 inversion experiment, 585 supports deep port construction 773 Commonwealth Associates Inc. SO2 removal process, 689 Compaction of solid wastes, 412 Composting N.Y. City to pay Ecology, Inc. 971 of solid wastes, 417 Computer program computing equilibria in aqueous chemical systems, 58 Conifuge for particle size determination of Delmarva Power and Light Co. particulate matter, 1091 Connecticut contract with GE for statewide solid waste system, 864 Conservall, Inc. spent acid regeneration process 969 **Conservation Foundation** position on strip mining, 192 **Consolidated Coal Co.** clean coking process, 1063 Consolidated Edison Co. closed-cycle cooling at Indian Point 2, 967 Coors, Adolph Co. brewery waste control, 505

Copper industry Arizona SO₂ regulations, 485 Japanese smelting process, 13 Core analysis for determination of accelerated enrichment of lakes, 799 Corn pollen germination prevented by ozone, 427 Corps of Engineers, U.S. Army discharge permit program, 105 **Council on Economic Priorities** praises pulp and paper industry on cleanup, 776 Council on Environmental Quality (CEQ) estimates cost of cleanup, 773 impact of freight rates on recycling, 1059 study of harbor facilities, 483 summation of U.N. Conference, 675 two new members appointed, 965 urges debate on concept of growth, 395 views on proposed water amend-ments, 9 views on weather modification, 773 Crane Co. role in water and waste water treatment business, 974 Crown Zellerbach Co. converts paper mill to recycling, 1063 D Dames & Moore Co. to investigate hazardous spills, DDT INVE concentration by coloring colloids in surface waters, 1033 in estuarine sediments, 914 DDT in lake trout residues as a function of fish age, 451 **Decision Sciences Corp.** national energy model, 205 **Decontamination techniques** for pesticide containers, 826 Deep well disposal survey of practice, 120 **Defense Department, U.S.** demise of management system. 199 military installations in N.J. to clean up, 11 De La Rue, Robert, and Associates report on consulting profession, 110 Delaware deep port off coast proposed, 773 EPA grant for resource recovery system, 1061 goose mortality due to lead shot, 201 to be reimbursed for hurricane damage, 201 shredding plant opens at landfill, 866 wins air pollution case against utility, 967 Delaware River Basin Commission drops plans for regional treat-ment plant, 1061 cited for air violation by EPA, 305 Denitrification, columnar conditions for plant design, 260 for nitrogen removal from waste waters, 260 Depolarization method for suspended solids in sewage, 43 Desalination falling film distillation unit, 109 piezodialysis process, 397 unit powered by trash burning, 587 **Desulfurization of fuels** with half-calcined dolomite, 1009 Detergents

BCPA's as builders, 203

Chicago bans phosphate, 485 Environmental impact statements manufacturers list ingredients on packets, 681 phosphate replacements, 300 Dieldrin adsorption by lake bottoms, 538 Differential voltammetry analytical method for trace elements in water, 243 **Dominion Foundries and Steel Co.** deep-bed filtration unit, 398 Dorr-Oliver, Inc. role in water and waste water treatment business, 974 Dow Chemical Co. water reuse at latex plant, 1072 Dravo Corp. role in water and waste water treatment business, 974 **Drinking water** presence of organic compounds in, 1036 removal of disagreeable odors and tastes in, 529 Е Eastman-Kodak Co. smoke suppression system, 489 Ecodyne Corp. interview with company president, 868 role in water and waste water treatment business, 974 Ecology, Inc. to be paid by N.Y. City to com-post trash, 971 small incinerators, 219 Editorials antitechnology league, 583 calling a spade a spade, 393 degradation of clean air, 673 environmental information, 961 environmental numbers game, 7 the EPA credibility gap, 861 guys in the white hats, 197 how clean is clean enough?, 103 the need for more R&D, 1057 runoff poses control challenge, 771 Effluent standards Illinois adopts water standards, 201 Eldib Engineering & Research Co. detergent builder study, 203 Electron microprobe for measuring lead particles in trees, 1121 Electrostatic precipitator efficiency role of sulfuric acid mist on, 895 Emissions monitoring stack monitor, 397 **Emission standards** stationary pollution for air sources, 105 Energy congressional hearings on nation-al policy, 601 insulation could save fuel, 588 NAE report, 105 national energy model, 205 oil imports through superports, 778 switch to efficient uses could slow demand, 776 Energy Co. of Alaska propsed refinery for North Slope oil. 681 Enforcement of clean air laws, 113 **Engelhard Industries** auto exhaust catalyst, 776 Enviro Control, Inc. water pollution monitor, 510 Envirogenics Corp. health risk computer technique, 205 Environment/One Corp. role in water and waste water treatment business, 974 Environmental health

EPA responsibilities and pro-grams, 589

Alaskan oil pipeline, 398 on North Carolina dam, 303 Environmental Protection Agency, U.S. (EPA) action on auto emissions control. 512 action on pesticides, 395 acts against Baltimore harbor polluters, 485 air pollution order challenged in Delaware, 489 approves state air implementation plans, 599 auto certification ability rapped by GAO, 585 awards to employees, 9 bans DDT uses, 675 charges for training courses, 675 classifies As, Be, Hg as hazardous air pollutants, 9 community environmental plan-ning, 483 construction grants awarded, 863, 1059 degradation of air quality, 1059 dislocation warning system, 9 effluent guidelines for water pol-luters, 786 estimates of radiation sources, 965 fiscal 1973 budget analysis, 207 funds resource recovery plant in Delaware, 1061 gasoline additive regulations, 303 guidelines for sediment control, as state air implementation plans, 395 has state health effects of pollutants, 589 issues 180-day notices in N.Y. City area, 774 manpower requirement studies. 694 marine sanitation standards, 675 new source performance stan-dards, 884 NO_x measurement rationale, 890 readies lead-in-gasoline regulation, 675 receives state air implementation plans, 199 refers Ford case to Justice, 965 rental car emissions program, 109 resource recovery grants, 971 sanitary landfill studies, 410 serves 30-day notice on Delaware utility, 305 solid waste program's new directions, 318 state implementation plans, 773 taps firms for hazardous spill studies, 1063 technology transfer program, 105, 314 toxic substances controls, 788 turn-key proposal, 303 upcoming performance standards, 105 view of municipal incineration, 992 view on deep well disposal, 120 waste treatment grant provisions attached, 303 work of e branch, 880 economic analysis **Environmental Science & Technolo**gy editorial policy and guide for au-thors, 523 Pollution Control Directory, 1D Envirotech Corp. role in water and waste water treatment business, 974 Equilibria computation method for aqueous chemical systems, 58 ERCO, Inc. pollution control at phosphorus plant, 980 Erosion EPA guide to sediment control, 965

Esso Research and Engineering Co. fluidized limestone coal-burning process, 865 smog formation studies, 487 SO2 removal process, 689 Ethers in the air bischloromethyl ether, 930 Ethvi Corp opposed to EPA lead regulations, 307 European Economic Community pollution control effect on U.S. exports, 681 Eutrophication data on 55 lakes in Florida, 719 mathematical simulation models for, 135 phosphorus control into Minnesota lake, 967 role of soluble phosphorus compounds in, 431 F Farmers Chemical Association, Inc. ammonium nitrate recovery, 692 Farmland Industries, Inc. fluoride emission control, 400 Federal budget, 207 Ferric hydroxide filtration effects of five variables, 169 Fertilizer industry fluoride emission control, 400 Fibrous bed coalescence unit for separating oil dispersions, 905 Filter paper advantages over glass in atomic absorption spectrographic work, 617 evaluation for neutron activation spectroscopic technique, 441 removal of soluble impurities in for atmospheric aerosol work, 455 Fish pesticide residues in trout, 451 Fish protein concentrate manufacture mercury extraction technique for, 726 Florida large bond issue in Dade Co., 967 nutrient supply and loadings in lakes, 719 phosphate detergent ban in Dade Co., 11 prohibits some new sewer connections, 1061 Flue gases nitrogen oxides analysis for, 727 Fluoride emissions control at phosphate rock plant, 400 water samples, 79 proton activation Fluorometry technique for selenium analysis in water samples, 621 Food industry fungal treatment of canning wastes, 587 Ford Motor Co. EPA suit on test irregularities re-ferred to Justice, 965 opens clean casting center, 110 requests emissions standards waiver, 398 sued over air emissions, 774 Forestry alternative wood fiber sources, 876 Foster-Wheeler Co. SO2 removal process, 689 Fry Roofing Co. challenges Colorado on air law, 677 G Garrett Research and Development

San Diego pyrolysis project, 971

General Accounting Office, U.S. Congress (GAO) attacks EPA grant requirements, 303 environmental program audits, 404 report on EPA auto certification, 585 General Electric Co. will develop solid waste plan for Conn., 864 General Motors Corp. catalytic exhaust system tested, 1061 requests emissions standards waiver, 398 Georgia court ruling on county's responsibility, 11 Getty Oil Co. sues EPA over air pollution order, 489 Glass reclamation plant goes on-stream, 1063 recycling programs status, 988 **Glass Containers Manufacturers In**stitute glass recycling programs, 988 Graphical techniques for analysis of kinetic data, 1114 Graver Water Conditioning Co. physical/chemical plant, 306 treatment tube settler design, 312 Groundwater on Long Island, 192 nitrate ion analysis for, 835 pollution and conservation, 213 USGS storage project, 199 Grumman Aerospace Corp. engineer retraining program, 316 **Gulf Coast Waste Disposal Authority** organization and programs, 402 **Gulf of Mexico** superport location studies, 779 Gulf Research and Development Co. reduction catalyst for NO_x con-trol, 487 н Hawaii given EPA planning grant, 863 sugar companies sued over water pollution, 1061 **Heavy metals** removal from water with silicon alloys, 1109 in sewage sludge, 397 zinc recovery from waste stream, 880 Heavy metals in water analytical differential voltammetry method for, 243 field technique for lead, cadmium, zinc, and iron, 452 Hercules, Inc. receives EPA grant for Delaware plant, 1061 Hooker Chemical Co. environmental coordinator, 216 Houston Research, Inc. microbial oil slick degradation. 306 Hudson River Fishermen's Assoc. presses AEC on closed-cycle cooling, 967 Hydrocarbons photooxidation at atmospheric concentrations, 837 Hydrocarbons in the air photosulfoxidation of materials in liquid phase, 933 technique for reactive smog-form-ing materials, 710 Hydrogen peroxide role in polluted atmospheres, 816 Hydrogen peroxide in the air formation from formaldehyde photooxidation, 816 Hydrogen sulfide removal from liquid streams, 487

Hytek International, Inc. ion exchange treatment process, 865 Т Idaho new state environmental department, 677 Illinois adopts water effluent standards, 201 approves steel company abate-ment program, 107 establishes asbestos regulations, 201 fines owner of improperly operated landfill, 967 radiation incident team, 305 rejects proposal to incinerate de-foliant, 201 releases funds for sewage plants. 11 tight controls of nuclear power plants, 11 Imperial Chemical Industries Ltd. auto exhaust catalyst, 15 Incineration basis of new source performance standards, 885 model incinerator at MIT, 13 of scrap tires, 775 solid waste burned as energy source, 780 of solid wastes, 415 status on the municipal front, 992 waste heat recovery, 219 Industrial pollution control impact on foundry industry, 790 at phosphorus plant, 980 Industrial waste control at brewery, 505 at carpet latex plant, 1072 chrome tanning process, 594 deep-bed filtration of steel ef-fluent, 398 at fertilizer plant, 692 heavy metals removal, 518 hydrolysis of cyanide wastes, 679 Lancy Labs adsorption process, 687 lead and fluoride waste treatment, 321 package treatment plant, 487 processes for spent acid wastes, 969 protein from tannery wastes, 679 spray irrigation system, 587 tube settlers, 312 Industrial waste streams precipitation of ferric hydroxide from 169 precipitation of iron compounds from, 169 Infrared technique for oil analysis in presence of biological matter, 833 Interstate Commerce Commission freight rate increase and NEPA, 1059 Interstate Commission on the Potomac River Basin organization and goals, 1074 Interuniversity Consortium for Envi-ronmental Studies, 201 Interviews Gov. Melvin Evans, 1065 Vaun Newill, 589 John Schork, 16 Stewart Udall, 309 Frank W. Zurn, 495 Ion exchange heavy metals removal, 521 plating waste treatment, 865 used for waste treatment, 692 lonics. Inc. membrane process, 397 Ionics/Stone & Webster SO₂ removal process, 689 lowa closes open dumps, 305 Iron foundry industry impact of pollution controls, 790 use of wet scrubbers, 994

Iron in water

field polarographic technique for, 452 Isotopic analysis

for lead sources, 705

ITT Rayonier, Inc.

Raycycle pulping process, 596 IU/Conversion Systems, Inc.

sludge encapsulation process, 874 J

Jacobs-Hochheiser method reevaluation for atmospheric nitrogen dioxide analysis, 152

Justice Dept., U.S. files water pollution suits in N.Y., N.J., 774

κ

Kentucky air pollution control deadlines, 396 control of mine water drainage. 305 federal air pollution sources and permits; 1061 Keystone Reservoir mathematical models for simulation of eutrophication, 135 Kinetics desulfurization with dolomite. 1009 graphical techniques for data analysis, 1114 oxidation of sulfides in water by oxygen, 529 L Labor Department, U.S. dislocation warning system, 9 Lake Mendota, Wis. sedimentary history of, 799 Lake Michigan models for predicting pollution loads, 331 Lake Ontario U.S.-Canada study kicked off, 305 Lake Superior Justice Dept. sues Reserve over taconite tailings, 305 Lancy Laboratories, Inc. adsorption treatment process, 687 Land disposal sludge and sewage effluent, 871 Land use planning New Jersey's Meadowlands, 506 study in Maine, 677 Lead effect on auto exhaust control systems, 199 EPA gasoline regulation, 675 in lake sediments, solubilization by NTA, 278 levels in gasoline sold in N.Y. City, 485 measurement of blood concentration, 969 in plants, correlation with vehicle exhaust compositions, 1121 in polluted atmospheres, 68 pollution and poisoning, 31 quick blood level test, 775 Lead in the air concentrations in Berkeley, Calif. (1963-70), 558 identifying different sources, 705 isotope technique for, 705 monitoring by nondispersive X-ray fluorescence, 558 Lead in water field polarographic technique for, 452 Lederle Laboratories, Inc. Unox treatment process, 878 Legislation: federal conference agrees on water bill, 863 conference on water pollution act, 483 congressional backlog, 208

NEPA amendments, 675 noise control bill, 303 noise law, 1059 pesticides law, 1059 progress under 1970 air amend-ments, 111 Senate committee approves interstate compact, 303 tax incentive bill for solid waste recycling, 681 Toxic Substances Control Act, 788 water bill in conference, 675 Water Pollution Contro amendments, 786, 1068 Control Act water pollution act is delayed, 395 Legislation: state Metals New Jersey bill requires industrial waste pretreatment, 396 Pa. empowered to set air pollution rules, 1061 in Pa. strip-mining control law, 107 Letters to the editor, 4, 192, 300, 388, 478, 580, 670, 769, 859. 958 Lindane adsorption by lake bottoms, 538 dissolved salts effect on solubility in water, 919 Los Angeles irradiation of polluted air samples from, 342 Lucas American Recyclers, Inc. scrap tire incineration, 775 M Maine land use control study, 677 Management role of environmental coordinator, 216 Manganese in water spectroscopic technique for, 642 Manpower and training aerospace eng 316, 671, 958 engineer retraining EPA training courses, 675 iob opportunities, 302 pollution control caused layoffs, 790 report on environmental jobs, 694 retraining program deficiencies, 478 Manufacturing Chemists Assoc. pollution control spending, 398 Maryland activities of Maryland Environmental Service, 324 Massachusetts forces small city to upgrade sew-age treatment, 586 Mass spectrometric analysis for bischloromethyl ether in the air, 930 use for air and water pollutants near cities, 338 Mathematical modeling for aerosol filtration, 821 for aerosol filtration on fibrous media, 1101 for atmospheric photochemical reactions, 812 for eutrophic simulation in reservoirs, 135 for industrial and combined wastes in lakes, 331 simplified method for photochemi-cal smog formations, 360 for slip flow in aerosol filtration, 1101 for water pollutants in lakes, 331 Mercuric chloride in water inhibition on lipid biosynthesis in freshwater algae, 158 Mercury analysis of, in water, 1062 classified by EPA as hazardous air pollutant, 9 compounds in water, removal by agricultural products, 457 in fish, levels compared with mu-seum samples, 901

in fish protein concentration manufacture, 726

in industrial waste waters, remov-al by solvent extraction technique, 525 pathways in LaHave River system (Nova Scotia), 274 removal by synthetic and natural polymers, 745 removal from water with silicon alloys, 1109 in the sea, concentrations at two stations off coast of Mexico, 644 in water sediments, data for New Haven (Conn.) Harbor 1123 Metal finishing industry treatment of spent acids, 969 copper recycling, 700 removal from industrial effluents, 518 tobacco smoke condensate, 740 in urban soils for metropolitan Grand Rapids, Mich., 560 Metals in the air monitoring network for trace met-als, first-year results, 1025 nondestructive analysis technique. 1105 presence in particulate matter, 922 X-ray fluorescence technique for particulate matter, 1105 Metal plating wastes adsorption treatment process, 687 Avco freeze purification process, 487 treatment by hydrolysis, 679 Methanol aids in anaerobic waste treat-ment, 775 as natural gas substitute, 775 Methyl mercuric chloride inhibition on lipid biosynthesis in freshwater algae, 158 Met-Pro Water Treatment Corp. role in water and waste water treatment business, 974 Michigan county sues Ford over air emissions, 774 emergency spill cleanup funds, 864 National Guard to collect water samples, 967 Michigan State University receives EPA support for lagoon spray system, 864 Microbial degradation of aliphatic acids in soils, 732 Minnesota advanced waste treatment at Ely. 967 animal feedlot control, 586 may not seek tougher radiation regulations than AEC, 396 Missouri contracts with school for water analysis, 586 new air pollution source permit system, 1061 Mitsubishi Metal Mining Co. copper smelting process, 13 Monitoring networks national network for particle size measurements, 1019 Monsanto Enviro-Chem Systems pyrolysis system for Baltimore, 971 SO₂ removal process, 689 Montana feedlot controls and permit program, 774 implementation plan omits smelter SO₂ controls, 396 Municiplex, Inc. financing municipal treatment plants, 406 N Nashville Thermal Transfer Corp. proton activation analysis for energy from waste project, 780

Academy of Engineering

(NAE)

Nitrogen dioxide in the air report on energy problems, 105 analytical methods for, 152 study of small particles needed, 863 Nitrogen oxides (NO_x) National Aeronautics and Space Adin the air, quantitative separation ministration (NASA) of NO and NO2, 250 ERTS remote-sensing satellites, analysis of flue gases containing 775 SO2, 727 National Center for Resource Rein automobile exhausts, chemistry of, 543 covery, Inc. organization and goals, 1078 control at nitric acid plants, 110 resource recovery site in Florida, control in oil combustion, 109 586 measurement techniques and im-plementation, 890 National Constructors Association supports turn-key proposal, 20 reduction catalyst for auto ex-National Environmental Policy Act haust, 487 (NEPA) use of Saltzman technique for. 348 analysis of impact, 209 National Industrial Pollution Control Nitrogen removal from waste waters breakpoint chlorination technique Council for, 622 report of steel subcouncil, 971 by columnar denitrification, 260 National Science Foundation (NSF) RANN program, 508 Chicago fines ordinance violators, National Steel Corp. clean coking process, 1063 774 National Water Commission control of aircraft, 220 control begins in New Jersey, 305 review of U.S. water problems, 9 federal legislation passes, 1059 Neptune Meter Co. new regulations in N.Y. City, 967 role in water and waste water report to Congress, 9 treatment business, 974 review of sources and controls, Neptune Microfloc, Inc. 124 tube settler design, 312 Nerve gas Nondestructive test method decomposition in the ocean, 928 for trace elements in air, 441 for metals in air, 1105 New Jersey abatement order to Delaware North American Rockwell Corp. River polluters, 1061 air pollution fine on insulation views of executive on pollution control, 205 manufacturer, 967 Nuclear power approves ocean outfall pipe, 11 AEC radioactive discharge data, auto exhaust checking program, 105 785 community recycling projects, 485 controls noise emissions, 305 cracks down on illegal dredging, 396 land use planning for Meadow lands, 506 offshore power plant proposed. 971 orders military installations to NVF Co. clean up, 11 several cities sued over water pollution stricter particulate emission stan-dards, 485 visible smoke regulation, 107 wetland dumping restrictions, 485 **New York City** noise regulations, 967 Oceans pretreatment of metal-plating wastes, 864 sewage treatment plant, 586 sued over water pollution, 774 tests on-site hypochlorite generation. 201 underground sewage plant, 107 Odors uses glasphalt pavement at air-port, 677 Ohio New York State curbs commercial waste collectors 774 given EPA planning grant, 863 groundwater on Long Island, 192 Nickel distribution in cigarette smoke, 830 Nitrate ion analysis brucine methods for, 835 Nitric acid plants basis of new source performance standards, 885 Nitric oxide (NO) NBS monitor, 969 Nitrilotriacetic acid complex with iron and photochemical deg-radation of, 367 **Oil spills** solubilization of lead in lake and reservoir sediments by, 278 Nitrogen analysis

water samples, 79

Illinois imposes tight controls, 11 proposed N.J. offshore plant, 971 temporary A censes, 585 AEC operating liwarm water discharges, 224 Nutrient removal at new Cleveland treatment plant, 782 phosphate removal with alum, 201 plant in Minnesota, 306 zinc recovery from waste stream, 880 0 Ocean dumping banned in California, 677 of toxic materials, 928 decomposition of nerve gas in, 928 Occupational health and safety OSHA's second year, 483 purpose and programs of OSHA, 684 technology for control, 602 permits for air emission sources, 586 possession of nonreturnables illegal in Oberlin, 305 radioactive release from Piketon gaseous diffusion plant, 368 studies new state environmental agency, 396 Oil analysis in presence of biological matter, 833 **Oil dispersions** separation by fibrous bed coalescence, 905 **Oil shale** prospective development in Colo-rado, 863 API research program, 15 chances reduced in offshore ports, 778 cleanup with microbes, 306 contingency plan in Wisconsin, 107

Michigan appropriates cleanup money, 864 **Oil in water** unit for separating, 905 Oklahoma permits for solid waste disposal 677 Oregon agrees with feds over water permits, 774 will prohibit open burning by 1975, 397 Organic compounds in water characterization of materials in drinking water, 1036 coprecipitation with iron salts, 1031 Organic materials in air chemical and physical properties of, 633 Overview Corp. programs and approach, 309 **Oxygen** atoms reactions with reactive hydrocar-bons, 742 Ozone effect on corn pollen, 427 Ozone in the air effects on navel oranges, 1014 P Paper plastic composite of, 1085 Paper industry impact of legislation on control spending, 205 Parathion containers decontamination technique for, 826 Particle size determination conifuge use for, 1091 glow discharge device for, 1117 by glow discharge perturbations, 1117 Particulate emissions control by wet scrubbing, 994 stricter New Jersey standards, 485 Particulate matter fluorescence analytical methods for, 922 health effects of small particles, 863 particle size determination for, 1105 removal by Pentapure Impinger, 865 removed by small precipitator. 587 size distribution data from nation-al surveillance network, 1019 size distribution from lignite combustion, 929 Patent Office, U.S. antipollution patent processing, 307 Peabody Engineering Corp. SO₂ removal process, 689 Peabody-Galion Corp. role in water and waste water treatment business, 974 Peerless Instruments Corp. stack monitor, 397 Pennsylvania air pollution rules, 1061 enforces sanitary landfill regulations, 396 xperiments with biological pest control, 586 experiments phases out dumps, 677 sediment control regulation, 1061 statewide air monitoring system, 586 strip mining regulation, 107 sues U.S. Army for pollution, 1061 Pennwalt Corp. role in water and waste water treatment business, 974 The Permutit Co. tube settler design, 312 Peroxyacetyl nitrate (PAN) effect on tobacco leaves, 71

Pesticide containers decontamination technique for, 826 Pesticides see also names of specific compounds adsorption by lake bottoms, 538 biological control of gypsy moth, 586 EPA bans DDT, 675 EPA takes testimony on DDT, bans mercury-containing pesticides, 395 federal legislation passes, 1059 herbicide spray controller, 203 in lake trout, 451 metabolism by marine organisms, 629 removal from the soil by volatilization of degradation products, 910 volatilization of degradation products from soils, 910 Petroleum industry costs of pollution abatement, 307 fate of waste lube oils, 25 Philadelphia fines owners of smoky autos, 11 Phosphate removal use of activated sludge for, 280 Phosphorus in lakes, soluble portion and bio-logical activity, 431 proton activation analysis for water samples, 79 Photochemical reactions in the air effect of homogeneous mixing on, 812 Photochemical smog effect of hydrocarbon and nitrogen oxides on, 253 hydrocarbon and NO_x role in for-mation of, 253 mechanism for, 47 reactive hydrocarbons direct measurement in, 742 role of humidity in formation, 487 role of singlet molecular oxygen in. 365 role of SO2 in, 423 Photochemistry degradation of iron: NTA complex, 367 of Los Angeles atmospheric sam-ples, 342 Photooxidation of sulfur dioxide, 72 Physical-chemical treatment in new Cleveland plant, 782 for removing nitrogen from waste waters, 622 Phytoplankton methods for determining growth rate of, 1096 Plants cadmium uptake by, 1005 Plastics for solid waste composite formation, 1085 **Polarographic analysis** for heavy metals in water sam-ples, 452 **Pollution control business** assessment of water and waste water control market, 974 Research-Cottrell Co., 16 views of Frank Zurn, 495 Pollution control expenditures by chemical industry, 398 financing m plants, 406 municipal treatment as result of proposed legislation, 205 Pollution Control Industries, Inc. role in water and waste water treatment business, 974 Polychlorinated biphenyls U.S. task force recommends against complete ban, 395 Polymers for removing mercury as mercuric chloride from aqueous solu-

Potomac River goals of river basin commission, 1074 pollution by sewage, 4 **Power generation** basis of new source performance standards, 885 costs of pollution abatement, 307 plants proposed in Wyoming, 864 use of wet scrubbers, 994 warm water discharges, 224 Presidential announcements and messages environmental message to Congress, 199 J. F. Pritchard & Co. SO2 removal process, 1062 Procon/UOP SO₂ removal process, 689 Procter & Gamble Co. withdraws detergents sale from Dade Co., Fla., 11 Protein formation from cellulose waste, 715 Proton activation analysis for trace metals in water samples. 79 Pulp and paper industry cleanup at Washington and Ore-gon mills, 307 costs of pollution abatement, 307 kenaf as raw material source, 306 mill converts to recycling, 1063 paper recycling, 700 praised on cleanup program, 776 Ravcycle process, 596 spray irrigation with waste water, 587 **Purity Corp** particulate removal device, 865 Pyrolysis of solid wastes 417 Union Carbide process, 203 Questor Corp. automotive exhaust control sys-tem, 1062 R Radiation effects on newborn children, 199 EPA assesses relative role of sources, 965 Illinois incident response team. 305 uranium concentrations in surface air, 368 **Radioactive discharges** release from Piketon nuclear gaseous diffusion plant in Ohio, 368 underground storage of wastes, 13 Raytheon Co. incinerator residue project, 971 Recycling conversion of paper mill, 1063 at fertilizer plant, 692 glass containers, 988 at Kaiser steel mill, 588 secondary effluent study in Cali-fornia, 677 the secondary materials industry, 700 sludge and sewage effluent, 871 **Remote sensing** ERTS satellite is transmitting, 775 Republic Steel Corp. clean coking process, 1063 **Research-Cottrell Co.** interview with president, 16 SO2 removal process, 689 Reserve Mining Co. sued over tailings discharged to Lake Superior, 305 Resources alternatives to trees as fiber sources, 876

tions, 745

Resource recovery EPA funds Delaware plant, 1061 EPA lets three contracts, 971

planned site in Tampa, Fla., 586 of solid wastes, 419 Rex Chainbelt, Inc. enters air pollution control field, 866 role in water and waste water

treatment business, 974 Reynolds Metals Co. aluminum remelting process, 306

Rhodia, Inc. H₂S removal process, 487

Rollins International, Inc.

role in water and waste water treatment business, 974 Ryckman, Edgerly, Tomlinson &

Assoc. to investigate hazardous spills, 1063

S

Saltzman technique for NO_x critical evaluation of, 348 Sanitary landfill design and operation, 408 Savannah Electric & Power Co. SO₂ removal process, 397 Securities and Exchange Commission environmental information for investors, 303 Selenium in water fluorometric analysis for, 621 Severn estuary DDT fate in sediments of, 914 Sewage measurement of suspended solids in, 43 Sewage sludge bactericidal properties of digested sludge, 448 dangers as fertilizer, 397 toxic agent in the liquid phase of digested sludge, 448 Sewage treatment algae growth project, 587 Cleveland upgrades old plant, 782 disinfection with on-site hypochlorite, 201 financing municipal plants, 406 high rate treatment is possible, merits of turn-key plant construction, 20 MES services in Maryland, 324 methanol aids anaerobic decomposition, 775 PhoStrip process tests, 15 physical-chemical plant in Minnesota, 306 status of physical-chemical pro-cesses, 984 tube settlers, 312 underground plant in New York City, 107 use of oxygen, 109 water machine," 397 Silicon alloys for removing heavy metals from water, 1109 Singlet oxygen production from photoexcited ni-trogen dioxide, 837 role in photochemical smog for-mations, 365, 837 Sludge encapsulation process, 874 land disposal, 871 materials recovery from phos-phate, 1062 production of biopolymers from, 161 useful biological materials in, 161 Small Business Administration, U.S. pollution effects on business, 123 Smoke Kodak suppression process, 489 Sodium analysis proton activation analysis for water samples, 79

Soil effect of chemical structure on degradation of aliphatic acids in, 732 Soil samples presence of 11 metals in indus-trial areas in Michigan, 560 Solid wastes Alaska enforces law, 774 aluminum can remelting, 306 bagasse used for bank checks, 588 building bricks from waste, 502 burned for power, 587 California bans ocean sludge dumping, 677 Chemfix sludge fixing process. 203 Colorado embraces utility con cept, 677 composting in N.Y. City, 971 future of municipal incineration. 992 Glasphalt pavement at airport, glass reclamation plant onstream, 1063 heat of combustion, formula for .1119 incineration of scrap tires, 775 MES services in Maryland, 324 mill recycles waste paper, 1063 new directions for federal program, 318 on-stream resource recovery. 1078 Pa. enforces landfill regulations, 396 Pennsylvania, Oklahoma, require sanitary landfills, 677 plastic composites of, 1085 reclaiming peanut hulls, 592 recycled plastic as concrete component, 13 recycling plastic foam, 679 recycling problems and opportuni-ties, 700 recycling projects supported by New Jersey, 485 resource recovery projects, 971 sanitary landfill design, 408 sanitary landfill operations, 408 sewage sludge fertilizer hazards, 397 shredding as adjunct to landfilling 866 status of glass recycling pro-grams, 988 tax incentive bill in Congress, 681 technology and programs, 1078 trash burned as energy source, 780 treatment technology, 412 25% of steel in cans is recycled, 681 Union Carbide pyrolysis process 203 use of plastic composites, 1085 uses for straw, 397 Solid waste combustion formula for heat output determination, 1119 Solvent extraction for removing mercury from indus-trial waste waters, 525 Solvents vapors and air pollution, 4 Spectrometer use to predict performance of air cleaning equipment, 614 Spectroscopic techniques for manganese concentrations in water samples, 642 Steel industry abatement program at Illinois mill, 107 balks at cleanup expense, 971 clean coking process, 1063 coke oven emission control, 118 costs of pollution abatement, 307 impact of legislation on control spending, 205 Kaiser plant wins industrial waste award, 588

steel cans contain 25% recycled | Toxic materials material 681 bischloromethyl ether in the air, use of wet scrubbers, 994 930 Stone & Webster/Ionics SO₂ removal process, 689 Sulfides in drinking water kinetics of oxidation of disagreeable constituents, 529 Sulfur analysis proton activation analysis for water samples, 79 Sulfur dioxide (SO₂) Arizona smelter controls, 485 control evaluation of 11 materials for stack gas controls, 350 control at sulfuric acid plants, 110 Cu smelting process features complete recovery, 13 emissions and control from ammunition plants, 986 oxidation by corona discharge, 895 photooxidation of, 72 in power plant plumes, attenua-tion as determined by instrumented aircraft, 172 removal from smelter off-gases, 396 removal from stack gases, 397, 959, 969 removal from sulfuric acid plant off gases, 1062 removal via homogeneous path-ways in the atmosphere, 72 review of technology for removal from stack gases, 688 role in photochemical smog for-mation, 423 stack gas removal process, 306 in stack gases, sorption on mag-nesium oxide, 268 Sulfuric acid plants basis of new source performance standards, 885 Sulfur isotope ratios use as air and water pollution index in cities, 338 Supreme Court, U.S. accepts water pollution case, 483 Suspended solids depolarization method for measurement, 43 Sybron Corp. role in water and waste water treatment business, 974 т **Technology transfer** EPA program launched, 105 Tekology Corp. building bricks from waste, 502 Tennessee Valley Authority (TVA) cost of pollution control, 489 to help southern states with rural waste collection, 864 long-term coal contract, 585 SO2 removal process, 689 strip-mining regulations, 15 Texas activities of the GCWDA, 402 **Textile industry** water use and waste treatment. 36 Thermal pollution beneficial uses of warm water 232 Utah British approach, 670 power station water intakes, 859 review of ecological effects, 224 use of waste heat, 389 Tobacco smoke condensate presence of metals in, 740 Tom's Foods Ltd. peanut processing, 592 Toxicology blood lead measurement, 969 information service readied, 489 quick blood test for lead, 775 toxic substance control consid-ered in Congress, 788

disposal of nerve gas in the ocean, 928 emergency evacuation procedure in Colorado, 677 targets of OSHA, 684 Toxic metals in cigarette smoke, 830 Trace analysis proton activation analysis for water samples, 79 Trace metals in the air filter papers advantage over glass filters, 617 Trace elements in the atmosphere analysis of 35 materials by neuactivation spectroscopy, tron 441 Trace elements in water analytical differential voltammetry method for, 243 Transportation Transpo '72 reviewed, 588 Trimex Corp. SO₂ removal process, 397 Tritium analysis of, in air, 1062 Tritium in water biochemical fate in algae metabolism, 638 **Turn-key construction** implications for consultants, 110 pros and cons, 20 used by Canadian steel firm, 398 views of Frank Zurn, 496 U Ultrafiltration heavy metals removal, 521 Ultraviolet cascade instrument for measurement of aerocolloids. 143 Union Carbide Corp. industrial use of Unox process, 878 molecular sieving for gas removal, 110 refuse pyrolysis system, 203 SO₂ removal process, 1062 United Kingdom government environmental department, 1076 United Nations (U.N.) Conference on Human Environ-ment, 199, 303, 675, 682 UOP/Procon SO₂ removal process, 689 The Upjohn Co. plastic foam recycling, 679 U.S. Army modernization and pollution control programs, 986 U.S. Geological Survey (USGS) authority over groundwater, 213 increases environmental effort, 965 water storage in aquifers, 199 U.S. Navy will study fluidized-bed incinera-tion, 1063 U.S.S.R. environmental agreement with U.S., 585 given EPA planning grant, 863 v Vandium in polluted atmospheres, 68 Ventilation systems characterization of systems in buildings, 609 Virginia air enforcement actions, 480 diesel bus injection system, 305 registers stationary air pollution sources, 396 Virgin Islands, U.S.

environmental quality consider-

ations, 1065

Viruses removal technique for waste waters, 438 Volvo

emissions standards requests waiver, 398

W

Washington state

given EPA planning grant, 863

Waste water treatment processes

virus removal technique, 438

Water pollutants organic compounds removal by coprecipitation with iron salts,

1031

Water pollution see more specific entries

Water Pollution Control Advisory, Board

composition, 965

Water pollution control congressional conference on new act, 483

costs of treatment and recycling,

discharge permit issuance barred by court, 105

EPA effluent guidelines, 786

guidelines in new federal law, 1068

importance of runoff, 771 Supreme Court accepts case for review, 483

in textile industry, 36

Water quality monitoring conductivity detects acid mine-water, 587

Enviro Monitor, 510 Mich. Nat. Guard to collect water samples, 967

review of instruments, 130 USGS monitors sediments for PCB's, 965

Water Resources Engineers study four river basins for EPA, 864

Water supply

Canadian excess could supply U.S. in drought, 776 Weather

impact of modification projects, 773

inversion height experiment, 585

Wellman-Lord SO₂ removal process, 689

Westinghouse Corp.

waste control at tube plant, 321

Westinghouse Electric Co. AEC study on saltwater cooling, 1063

automatic monitoring system, 110 Wet scrubbing applications in industry, 994

Wisconsin oil spill contingency plan, 107

water bank program, 586 Wyoming

new power plants proposed, 864

X

X-ray fluorescence for atmospheric lead concentra-tions, 558 X-ray fluorescence spectrometry

for metal determinations in particulate matter, 1105

Zinc analysis uptake by marine algae, 1001 Zinc in water field polarographic technique for, 452 Zurn Industries, Inc. history of the business, 495 role in water and waste water treatment business, 974

z

SO₂ removal process, 689

March 19-23

University of Texas at Austin

Advanced Water Pollution Control: Biological Waste Treatment Short Course Austin, Tex.

Write: Engineering Institutes, P.O. Box K, University of Texas at Austin, Austin, Tex. 78712

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Water Pollution-Law & Regulation

Milwaukee, Wis.

Fee: \$95. Contact: University of Wisconsin—Extension, Dept. of Engineering, 600 W. Kilbourn Ave., Milwaukee, Wis. 53203

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Call for papers

January 1 deadline University of Saskatchewan and others

Waste Water Treatment in Cold Climates International Specialty Symposium

Saskatoon, Saskatchewan, Canada Papers invited dealing with field investigations, theoretical studies, and pilot plant operations for waste water collection, disposal, and treatment in cold climates. Contact: E. Davis, Civil Engineering Dept., University of Saskatchewan, Saskatoon, Saskatchewan, Canada, STN OWO

March 15 deadline American Society of Mechanical Engineers

ASME Winter Annual Meeting

Detroit, Mich.

Papers invited for special session on dry cooling towers for power plant heat rejection. Topics include technological innovations, costs, and problems and solutions connected with dry cooling towers. Write: Ralph L. Webb, The Trane Co., La Crosse, Wis.54601

INDEX TO ADVERTISERS IN THIS ISSUE

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Calgon Corporation. Ketchum, MacLeod & Grove Inc. California Toxicology Service, Inc. Catalytic, Inc. APCL & Inc. Cheme Process Systems Inc. Advertising Design Associates	1133 1134
Chemec Process Systems Inc Advertising Design Associates	1134
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sion Campbell—Mithun, Inc.	1125
Hydrodyne, Inc.	1136
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Research Cottrell Michel-Cather, Inc. Rochester Gas & Electric	1133
Rochester Gas & Electric Hutchins/Darcy Inc.	1082

SERS IN THIS ISSUE	
Ryckman/Edgerley/Tomlinson & As- sociates, Inc.	1133
	1082
Schleicher & Schuell. F. M. Field & Company, Inc. Science Associates, Inc.	
	1134 1136
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	1135
SpectroChem Laboratories, Inc. Spraying Systems Company Tri-State Advertising Company, Inc.	1134 1060
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Ultrachem Corporation	1135
Union Carbide Corporation, Linde	1053
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	1136
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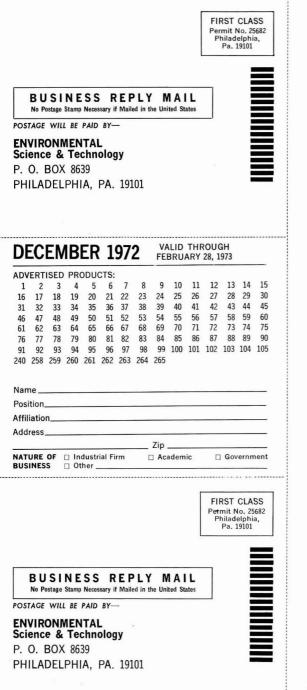
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