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A Journal of Science, Technology, Health, Monitoring and Policy

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# ENVIRONMENT INTERNATIONAL

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# ENVIRONMENT INTERNATIONAL

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## EDITORIAL

### A FURTHER STATEMENT ON SENSORY EFFECTS IN BUILDING ENVIRONMENT CONTROL

The Stockholm Statement (*Environment International*, Vol. 18, pp. 115-116, 1992) contained several recommendations on building-related allergic and other adverse effects on human health and comfort. It outlined short-term and long-term strategies, including those for prevention and remedial actions, and specific recommendations for implementation of these strategies. This statement was produced at a scientific meeting in Stockholm, June 17-18, 1992, organized by the Folksam Research Department and it supplements the Stockholm Statement.

(1) A number of parameters contribute to indoor air pollution. These include inadequate ventilation, poor choice of building materials, insufficient attention to chemicals used in buildings and poor maintenance of heating, air conditioning, and ventilation systems. Because the contribution of these parameters in specific cases is normally unknown, preventive measures in all cases are usually necessary. In particular, improved air ventilation is often a reasonable preventive measure.

(2) The non-industrial indoor air contains low concentrations of chemicals and the adverse effects of deteriorated indoor air are largely demonstrated by human sensory irritation and other perceptions. These are psychological phenomena. Therefore, it is highly desirable that tests according to psychological models be developed which have an adequate bearing on indoor environmental conditions.

(3) Whereas toxicological tests are available for assessment of respiratory irritants at high concentrations, there are no toxicological tests for these agents at levels of concern to non-industrial indoor air quality. Therefore, it is highly desirable to develop appropriate and acceptable animal and other models to test the irritation impact of chemicals before they enter the indoor air. It is essential that the outcomes of the applications of these models can be related meaningfully to human reactions in real-life exposure situations.

(4) Controlled low-level exposure and field studies of reversible sensory effects using human subjects provide valuable information for avoidance of adverse effects resulting from poor indoor air quality. Such studies should be conducted fully consistent with ethical requirements. Considering the scarcity of human data on the effects of indoor air pollution, the editors of scientific journals are urged to consider a rapid publication of human data.

(5) Sensory effects now have to be looked upon as important end-points in the health assessment of indoor air pollution. They may cause adverse effects on sensory systems such as environmentally-induced sensory dysfunctions. They may be precursors of diseases such as triggering of hypersensitivity reactions. When asthmatics experience an increased frequency of asthma attacks when they are exposed to certain agents above a certain concentration, some of this effect may be mediated by sensory stimulation. Increased frequency of flu and other respiratory infectious diseases has also been observed in spaces where the air exchange rate is inadequate or when the air is recirculated. It is probable that this is a combined effect of mucosal irritation or damages, and microbiological load. Such combined effects require increased attention.

(6) Sensory effects are also important parameters in indoor air quality control. They serve as warnings of many of the indoor contaminants, and usually provide an integrated measure of the sensory load on the indoor space. Since long, sensory effects have been used for dimensioning and checking the functioning of the ventilation system, for example, by the use of the odor criterion. A majority of the volatile organic compounds indoors have odor or are mucosal irritants and, thus, the general performance of the ventilation system is easily checked by measuring these sensory effects. Improvements with respect to the sensory effects very often are accompanied by a reduction of the overall amount of air

pollutants. Of course, some toxic chemicals, like radon, need to be controlled for by specific methods because they cannot be perceived and they may not be linked to other perceivable pollutants.

It is hoped that the international interest envisioned by this and the Stockholm Statement will result in an enhanced scientific database and the development of better theoretical and practical models for understanding and controlling environmentally-induced sensory effects.

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# LUNG CANCER IN RELATION TO AIRBORNE RADIATION LEVELS

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A 1986 aeroradiometric survey of the eastern two-thirds of Washington County, Maryland provided an opportunity to study lung cancers in relation to gamma radiation levels. In the first approach, lung cancer deaths between 1963 and 1975 in four areas of the county categorized as low, moderately low, moderately high, and high showed relative risks of 1.00, 0.93, 1.01, and 1.43, respectively, after adjustment of sex, age, and smoking. A second approach used lung cancer cases diagnosed between 1975 and 1989, controls matched to cases by race, sex, and age, and aerometric radiation readings above the individual residences. In four levels of increasing gamma radiation, odds ratios adjusted for smoking were 1.00, 0.84, 0.90, and 0.92, respectively. No differences were statistically significant.

## INTRODUCTION

Most of the published data relating lung cancer to radon comes from studies of miners (NRC 1988) and indicate a risk roughly proportional to the exposure in level  $\times$  time. Efforts to calculate the risk for non-miners living in houses with much lower radon levels than that found in mines involve the extrapolation of miners' data and are at best controversial. Until some case-control or prospective studies involving radon measurements in a large number of houses are completed, estimates of the lung cancer risk to non-mining populations must be assessed using surrogate measures. A number of approaches have been reported, for example:

- (1) Lung cancer rates among residents of houses of differing construction were reported by Axelson et al. (1979) and Simpson and Comstock (1983), each finding somewhat higher lung cancer rates among residents of houses whose construction was presumed to be indicative of higher radon levels.
- (2) Lung cancer rates in areas of differing natural background radiation have been reported and findings are not consistent. Allwright et al. (1983) found no relationship between total cancer rates and natural background radiation levels in the eastern section of the Republic of Ireland. Archer (1987), on the other hand, found lung cancer rates to be higher in counties of New York, Pennsylvania and New Jersey that were

directly over the Reading Prong granite deposits which tend to contain more uranium and radon than other, non-mineralized, rock; he did not, however, report any radiation measurements. Another study, by Hess et al. (1983), measured radon levels in water samples from over 2 000 public and private wells in Maine and categorized the probable radon levels in houses of the counties. He found a significant association between lung cancer in women and radon levels in the water by county, but the association among men was only suggestive.

(3) Lung cancer mortality and 14 000 indoor radon concentrations in houses in 18 Canadian cities were compared by Letourneau et al. (1983), who found no detectable association between radon daughter concentrations and lung cancer mortality rates, with or without adjustment for differences in smoking habits between cities.

(4) A Guangdong, China study spanning 14 years in two areas of high and low background radon levels could find no variations in lung cancer rates below a cumulative exposure of 15 working level months (WLM) (Hofmann et al. 1986).

(5) Cohen (1989), who probably has more indoor radon measurements than anyone else in the U.S., found that mean radon levels in houses sampled from counties with the highest lung cancer rates were lower than radon levels in houses sampled from the counties with lowest lung cancer rates. He found, incidentally, that mean radon levels in Washington County, Maryland houses were high, averaging 5.1 pCi/L.

## FIRST STUDY

### *Materials and methods*

In 1956, an aeroradiometric survey of the eastern two-thirds of Washington County, Maryland was conducted, using a DC-3 aircraft carrying a six-crystal scintillation detector gated to respond to a minimum gamma-ray energy of about 50 keV. Starting at the eastern boundary of the county, the surveys were made at a level of 500 feet above terrain along 44 lines at half-mile intervals (Moxham 1963). A map based on the survey showed areas of the county with six levels of aeroradioactivity: <200-400, 400-600, 600-800, 800-1000, and >1000 counts per second (CPS), electronically corrected for cosmic components. Chemical analyses of soil collected by Helen Cannon and Moxham showed aerial and sur-

face radiometric data to be highly correlated, with variation coefficients ranging from 17% at 100 to 4% at 1000 CPS.

In 1963, a private census obtained information on 91 909 individuals, an estimated 98% of the residents of Washington County (Comstock et al. 1970). Data of primary interest for this study were the race, sex, age, and smoking history of each adult in the household. In 1971, a follow-up of a 5% sample of the 1963 census households provided information on the characteristics of the individuals still residing in Washington County. Age, sex, education level and frequency of church attendance were the only factors significantly associated with still living in Washington County in 1971. Therefore, these factors were used to establish for each adult aged 25 and over in the 1963 census a probability of remaining in the county eight years later, which was entered on the census tape. These probabilities allow the population remaining in the county to be estimated for any point in the eight-year period; for this study, the mid-point population was calculated as of 1969. Total losses from death and migration averaged only 3% per year. Since only about 2% of the non-institutionalized population was black, this study is confined to whites.

All death certificates of Washington County residents who died between July 15, 1963 and July 14, 1975 have been coded as to primary, contributing and underlying cause of death using the 7th revision of the International Classification of Diseases (ICD). For this study, we used only deaths with underlying cause lung cancer (ICD 162).

For this part of the study, the map of airborne radiation levels measured in 1956 was superimposed on a map of Enumeration Districts used in the 1963 census, and the Enumeration Districts then categorized as to probable mean radiation levels to residents: high, moderately high, moderately low, and low, having airborne radiation levels averaging over 700, about 600, about 500, and under 300 counts per second respectively of gamma radiation.

### *Results*

Table 1 shows the distribution of the mid-point population among the four areas of varying radiation levels. In 1963, nearly two-thirds of the adult population resided in the areas of relatively low airborne radiation, and the percentage of cigarette smokers was higher in this group than in the other three; age distribution also differed considerably between the four areas defined. It was therefore considered desirable to adjust for these variables as well as for



Table 1. Relative risks of lung cancer mortality for white adults residing in four areas of increasing aerial gamma radiation levels, Washington County, Maryland, 1963-1975.

Estimated Mean Radiation Levels Counts/Second	<300	500	600	>700
Mid-point (1969) Population	20,797	5,631	9,735	1,776
Lung Cancer Deaths	139	28	51	16
Adjusted* Rate	6.09	5.65	6.16	8.70
Relative Risk	1.00	0.93	1.01	1.43
95% Confidence Limits	REF	0.62-1.39	0.73-1.39	0.85-2.39

\* Rate per 1000 in 12 years, adjusted by binary variable multiple adjustment procedure for sex, age in 1963 and smoking in 1963.

sex when calculating lung cancer death rates in the four areas. The adjustment procedure used was the binary variable multiple adjustment procedure of Feldstein (1966).

Table 1 also shows the number of lung cancer deaths that occurred in each of the four areas of radiation level during the 12 years, and the adjusted lung cancer death rates for the four areas of varying radiation levels. There are no statistically significant differences in death rates, and the relative risks in all cases include 1.0 in the confidence intervals. It is especially noteworthy that there is no consistent trend with increase in radiation level as would be expected if the dose-response was a linear relationship.

## SECOND STUDY

### Materials and methods

Another health census of Washington County conducted in 1975 provided an opportunity to take a different approach to the relationship, based on the levels of airborne radiation above the individual residences of lung cancer cases and matched controls. Excluded were the city of Hagerstown and the contiguous suburb of Halfway for which household locations were not mapped out in the 1975 census, thus eliminating the most densely populated areas of the county.

Cases were selected from the county cancer register according to the following criteria: an eighth revision ICD code of 162; diagnosis date between August 1, 1975 and December 31, 1989; white; residence in the study area.

For each case, two controls were chosen from the birthdate listing compiled from the census data. One younger and one older person was selected, matched to the case by sex and race. Virtually all the controls were within a month of the case's age, and controls were determined to be alive and without diagnosis of cancer (except non-melanomatous skin cancer) at the time of the case's diagnosis.

For mapping purposes, each subject was assigned a number, and a pin with the number inserted at the residence location in a large-scale map of Washington County on the wall. An overhead transparency of the radiometric survey results was then projected, superimposed on the wall map, and the radiation level was recorded for each household location, without knowledge of case-control status.

The radiation data were then linked with the 1975 census data for each case and control. Census data utilized in this study, in addition to the matching factors of race, sex and age, were self-reported cigarette smoking, education, drinking water source, cooking fuel, and housing status.

Table 2. Odds ratios of lung cancer incidence, 1975-1989, with cases and controls residing in areas of differing aerometric gamma radiation, Washington County, Maryland.

Mean Gamma Radiation Counts/Sec	< 200	200-399	400-599	600-799	800-999	1000 +
-----						
Lung Cancer						
Cases	0	11	99	140	11	2
Controls *	2	16	214	269	11	14
	----- -----				----- -----	
Odds Ratios:						
Crude		1.00	0.76	0.85	0.85	
Adjusted **		1.00	0.84	0.90	0.92	
95% Confidence Interval		REF	0.38-1.84	0.41-1.96	0.34-2.52	
* Controls matched to cases by race, sex and age.						
** Adjusted for smoking by direct adjustment.						

## Results

Table 2 shows the number of cases and controls in each of the six radiation levels covered by the aeroradiometric survey. As the numbers in the extremely high and low radiation areas were small, the two lower and the two higher groups were combined for data analysis. Since the only census factor that showed an independent association with case/control status was smoking, this was the only factor used to calculate adjusted odds ratios.

There was no consistent increase in either crude or adjusted odds ratios with increase in radiation level at the household location.

## DISCUSSION

As with all studies, this one is subject to some difficulties. We did not include information on the length of residence in the particular area in which each individual lived, nor do the airborne radiation levels necessarily reflect the radon levels inside the houses in a given area. Nevertheless, this study does have the advantage of having obtained information on smoking habits before individuals became ill—and for the part of the study based on the 1963 census, before the Surgeon-General released the results of studies showing that smoking was hazardous to health.

The findings of this study are consistent with those of Allwright (1983) and Letourneau (1983), namely that moderately low levels of radiation may have

little if any measurable effect on lung cancer death rates in a general population.

Any attempt to compare these findings with the BEIR-IV estimate of 0.5-4.0% increase in lung cancers per WLM exposure is hazardous because it is impossible to relate directly the airborne radiation levels with the probable indoor radon exposure to residents. However, if we assume that on the average, residents in the lowest areas of airborne radiation receive a lifetime radon exposure of 10 WLM and that those in the highest areas receive about 35 WLM, the difference of 25 WLM should result in excess lung cancers somewhere between 12.5% and 100%, giving a relative risk of 1.13 to 2.0. Although this study's findings show little or no increase in lung cancer risks with higher airborne radiation levels, the 95% confidence limits do not completely rule out the possibility of agreement with BEIR-IV.

It is clear that many more studies are needed before definite conclusions can be drawn. Actual radiation measurements should be made inside homes and lung cancer rates in the homes of high- and low-level measurements compared. Certainly, extrapolations from data based on mine exposures, where radiation exposures are complicated by dust, fumes, and deep breathing, may justifiably be questioned as to their relevance to the household environment.

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## CALIBRATION OF THE HUND TYNDALLO-METER FOR MEASURING ENVIRONMENTAL TOBACCO SMOKE IN A TEST DUCT

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The Hund tyndallometer (Type TM Digital  $\mu\text{P}$ ) has recently been used to study the air quality in SAS aircraft. This publication contains an evaluation of its performance in measuring environmental tobacco smoke (ETS), comparing its output with the gravimetric method which was used for measuring respirable dust concentration (van der Wal 1988). The influence of several parameters, such as the age of ETS, the type of smoke (main stream or side stream), and the relative humidity was investigated. The Hund measurements of ETS generated by a smoking machine (ISO 7210) in a test duct were calibrated against gravimetric measurements of respirable dust trapped on teflon membrane filters. An accurate calibration was, however, not possible for outputs of less than 1 mV. This is 0.1% of the electrical output range (1V) and corresponds to  $100\mu\text{g}/\text{m}^3$  of respirable dust. For times of up to 7 min and relative humidity (RH) levels between 8 and 72%, the ageing of ETS did not appear to influence the output of the Hund. RH levels in this range also did not clearly influence the instrument response factors. Higher RH levels did, however, affect the response factor, probably due to condensation of water vapour on the particles. The average response factors for three types of cigarettes were respectively 177, 173, and  $138\text{ mg m}^{-3}\text{ V}^{-1}$ . The average response factor of the Hund for measurements taken during SAS flights was  $131\text{ mg m}^{-3}\text{ V}^{-1}$ , with a relative standard deviation of 24%. As some of the measurements did not meet the Hund preconditions (output  $<1\text{ mV}$ , gravimetric concentration  $<100\mu\text{g m}^{-3}$ ), they were excluded from this evaluation. The Hund tyndallometer can be a useful instrument to collect additional information on real-time peak concentrations under limiting operating conditions.

### INTRODUCTION

A study of components of the environment of the passenger cabins aboard 48 selected European Scandinavian Airlines Systems flights was conducted during the period of 14-30 September, 1988 (van der Wal 1988; Malmfors et al. 1989). The measurements were performed with a portable air sampler designed by TNO (van der Wal 1989; van der Wal and van de Belt 1990). One of the components measured was the respirable dust concentration. The time-weighted average concentration was measured by

sampling on a Teflon membrane filter (Millipore,  $1\mu\text{m}$ ) equipped with a cyclone to separate the coarse dust particles from the respirable particles (Casella Ltd). Simultaneously, the respirable dust concentration was monitored with a Hund tyndallometer type TM Digital  $\mu\text{P}$ , which measures only small respirable particles.

The characteristic of the cyclone of the Casella sampler is defined according to the Johannesburg Convention. The curve indicated by ACGIH is the characteristic according to the American Conference

of Governmental Industrial Hygienists (ACGIH). In practice, the Johannesburg Convention and the ACGIH curves give almost the same results; both curves are accepted by ISO (1981).

The measuring range of the tyndallometer is 0-100 mg/m<sup>3</sup>, corresponding to an output signal of 0-1 Volt. The signal is dependent on many particle parameters: particle size distribution, index of refraction, shape, and colour.

The Hund tyndallometer measures the light scattered by particles in the detection volume; thus, in principle, the surface area is measured, and not the particle volume (= mass).

One of the main sources of respirable particles in an aircraft is environmental tobacco smoke (ETS). The particle size distribution of ETS shows some variance, sometimes growing by coagulation and adsorption of water vapour, and sometimes shrinking by evaporation of water and other volatile compounds. Thus, the age of the ETS and the air humidity affect the particle size distribution and the output of the tyndallometer.

The main stream and the side stream of cigarette smoke have many different characteristics. Generally, the particle size distribution changes rapidly after generation, yet slows after several minutes (Keith and Derrick 1960).

Combination of the response curve and the curves of Keith and Derrick (1960) indicates that the response of the Hund tyndallometer will decrease about 40% in the period of 30-240 s after generation. The number of smaller particles (0.10-0.25 µm) decreases and the number of larger particles increases; however, this latter effect is insufficient to compensate for the decrease of the signal, due to the reduced number of small particles.

There was a poor correlation between the average output of the Hund tyndallometer (mV) and the average respirable dust concentration determined gravimetrically by Teflon filters (mg m<sup>-3</sup>) during the SAS flights.

According to the manufacturer, the ratio of the gravimetric respirable dust concentration to the output of the instrument is 100 mg m<sup>-3</sup> V<sup>-1</sup> (calibration with road dust). For the measurements taken aboard SAS aircraft, this ratio can be approximated with a log normal distribution. A log normal distribution is characterized by the geometric mean (GM) and the geometric standard deviation (GSD), which were calculated for four different classes (Table 1).

Although the GMs of the various classes did not vary much (from 120 to 150), the individual values showed great variation.

The aim of this investigation was to evaluate the relation between influencing parameters and the output signal of the Hund tyndallometer, especially for conditions common in aircraft cabins during flights. The gravimetric method is regarded as reference model for determining the respirable dust concentration.

#### TEST SET-UP

The parameters to be investigated were:

#### Ageing of ETS

ETS was generated at one end of an aluminium tube with a 30-m length and a 40-cm diameter. Air samples were taken from the tube at various distances from the generator. Air velocity was set at between 0.07 and 0.14 m/s, so that the age of the ETS was about 7 min at the other end of the tube for the lower speed.

Table 1. Respirable dust concentrations and ratio (response factor) of tyndallometer.

Class	Respirable dust concentration, µg m <sup>-3</sup>				Ratio, mg m <sup>-3</sup> V <sup>-1</sup>			
	GM	GSD	n	range	GM	GSD	n	range
T/NS	50	2.15	47	15-440	133	1.81	45	31-424
T/S	187	1.90	48	11-600	147	1.74	47	24-942
B/NS	155	1.67	48	45-560	119	2.60	40	9-2066
B/S	187	1.71	47	45-500	130	1.40	44	58-304

T = tourist section

B = business section

NS = non smoking

S = smoking

n = number of measurements

GM = geometric mean

GSD = geometric standard deviation

*Relative humidity (RH)*

The air supplied to the test tube was conditioned for temperature and relative humidity. The temperature was kept constant at 20°C ± 2°C, with varying relative humidity levels.

*Type of smoke*

Main stream smoke, side stream smoke, and a combination of the two.

*Smoke source*

Three types of cigarettes were used to generate the smoke.

Type A: low-tar cigarettes, channel ventilation;

Type B: ultralight cigarettes, conventional filter ventilation;

Type C: light cigarettes, conventional filter.

The parameters measured were:

Average gravimetric respirable dust concentration, measured by filters;

Response of the Hund tyndallometer;

Respirable dust concentration measured with a monitor on the basis of attenuation of β-radiation by the collected dust;

CO-concentration;

Temperature;

Relative humidity;

Air velocity.

INSTRUMENTATION

*The cigarette smoke generator*

According to the guidelines of ISO standard 7210, "Smoking machines for tobacco and tobacco products—non-routine test methods" (ISO 1983), a cigarette smoke generator was constructed in the TNO machine shop. The generator supplies one 35-mL puff of cigarette smoke per minute; cigarettes are automatically positioned in the airstream, lit, smoked (one puff of two seconds duration per minute), removed after seven or eight puffs, and replaced by the next cigarette. Details are described in the ISO standard 7210.

*The test duct and the sampling equipment*

The test duct consisted of an aluminium tube with a length of 30 m and a diameter of 0.4 m. At different points (Fig. 1), sampling points were connected to the Hund tyndallometer, and to a filter holder which was equipped with a Teflon membrane filter, and a cyclone with a characteristic according to the Johannesburg Convention. The air passing through the detection volume of the tyndallometer also passed through the filter holder, assuring that both devices take the same air sample. The air flow was controlled by a critical flow orifice and a pump, resulting in an air flow rate of 1.9 L/min.

Both the first and last sampling point were connected to a CO monitor, while the first point was also connected to a Frieseke/Hoepfner (FH) beta dust monitor with cyclone, which monitored the respirable

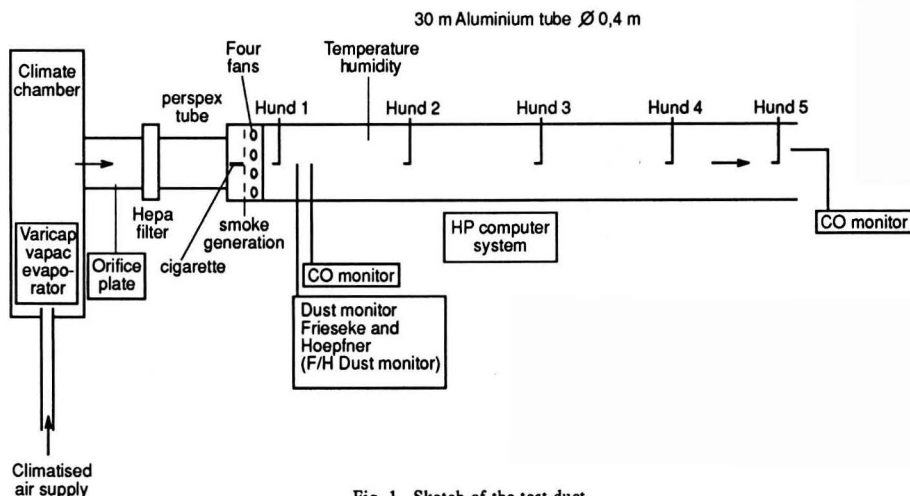


Fig. 1. Sketch of the test duct.

Sample point no.	1	2	3	4	5
Distance from smoke generator, m	1	8.5	16	24	30
Average residence time in s at 0.07 m/s	14	121	230	340	430
Hund tyndallometer	+	+	+	+	+
Teflon filter	+	+	+	+	+
FH beta dust monitor	+				
CO monitor	+				+

dust concentration. The characteristic of the cyclone follows the Johannesburg Convention. The sampling flow rate was 17 L/min ( $1 \text{ m}^3/\text{h}$ ).

The supply air, conditioned at a constant temperature and humidity, passed an HEPA (High Efficiency Particulate Air) filter in front of the smoke generator.

The Teflon filters were weighed in a conditioned room before and after the sampling. The room temperature was  $21.0^\circ\text{C} \pm 0.5^\circ\text{C}$  and the relative humidity  $58 \pm 2\%$ . Each filter was weighed three times. The average was used for calculating the weight of the collected dust. The deviation of any single mass measurement from the average was less than  $5 \mu\text{g}$ , and usually less than  $2 \mu\text{g}$ .

The CO-concentration was measured with a Maihak Unor 6 N infrared gas analyser. The detection was based on absorption of infrared radiation (nondispersive infrared). The lower detection limit was  $0.1 \text{ ppm}$  ( $0.12 \text{ mg m}^{-3}$ ); the range was  $0\text{-}50 \text{ ppm}$  ( $0\text{-}58 \text{ mg m}^{-3}$ ).

In order to obtain real time information about the dust load, the respirable dust concentration was monitored gravimetrically with a Frieseke-Hoepfner beta dust monitor, Type FH 62IT1.

A multichannel analyser (Hewlett-Packard computer system) collected the electrical signals from the tyndallometer, the CO-monitors, the beta-dust monitor, the temperature and humidity probes, and the orifice for measuring the air flow rate.

## RESULTS

The experiments with Type A cigarettes were preliminary experiments; six experiments were performed. Most experiments (18) were performed with Type B cigarettes. Four experiments were performed with Type C cigarettes, but only at conditions which are representative for flights in aircraft.

The results are summarized in Table 2.

Table 2. Summary of results of experiments with ETS from cigarettes.

Experiment no.	1	2	3	4	5	6
Type cigarettes	A	A	A	A	A	A
Type ETS*	M+S	M+S	M+S	M+S	M+S	M+S
RH, % (av)	38	45	5.5	1.7	88	67
air speed, $\text{m s}^{-1}$ (av)	0.07	0.17	0.17	0.07	0.07	0.07
Average for five sample points (relative standard deviation, %)						
Filter, $\text{mg m}^{-3}$	1.92 (4.6)	0.91 (1.3)	0.82 (3.6)	1.63 (4.2)	2.07 (6.2)	0.73 (5.7)
Hund, mV	13.6 (4.5)	5.3 (2.8)	6.3 (3.6)	15.3 (3.2)	19.8 (6.6)	6.00 (3.4)
Response factor, $\text{mg m}^{-3} \text{ V}^{-1}$	142 (2.5)	171 (3.6)	131 (2.6)	106 (3.6)	105 (7.6)	122 (3.9)
CO, ppm (n=2)	7.41	3.40	3.1	7.36	6.31	2.45
Av.ppm CO/av $\text{mg/m}^3$ (filter)	3.9	3.7	3.8	4.5	3.0	3.4

\* M = mainstream; S = sidestream



Table 2. Continued.

Experiment no.	7	8	9	10	11	12
Type cigarettes	B	B	B	B	B	B
Type ETS*	M+S	M+S	S	M+S	M+S	M+S
RH, % (av)	53	49.5	44	49	50	77
air speed, m s <sup>-1</sup> (av)	0.085	0.083	0.168	0.168	0.168	0.169
Average for five sample points (relative standard deviation, %)						
Filter, mg m <sup>-3</sup>	1.46 (3.6)	1.23 (6.8)	0.90 (0.9)	0.99 (8.0)	0.88 (6.7)	1.08 (5.9)
Hund, mV	8.11 (4.4)	7.16 (5.8)	4.62 (4.9)	5.83 (4.1)	4.59 (3.5)	10.1 (34)
Response factor, mg m <sup>-3</sup> V <sup>-1</sup>	180 (7.5)	172 (10)	192 (3.2)	171 (9.6)	191 (9.9)	117 (31)
CO, ppm (n=2)	5.54	5.44	4.18	4.22	2.93	4.51
Av.ppm CO/av mg/m <sup>3</sup> (filter)	3.8	4.4	4.6	4.3	3.3	4.2

\* M = mainstream; S = sidestream

Experiment no.	13	14	15	16	17	18
Type cigarettes	B	B	B	B	B	B
Type ETS*	S	S	S	M+S	M	S
RH, % (av)	72	79	76.5	76.5	21	20
air speed, m s <sup>-1</sup> (av)	0.169	0.088	0.168	0.168	0.168	0.169
Average for five sample points (relative standard deviation, %)						
Filter, mg m <sup>-3</sup>	0.99 (8.9)	1.90 (5.2)	1.82 (1.8)	1.79 (4.7)	0.022 (30)	0.49 (9.2)
Hund, mV	5.90 (7.0)	16.5 (17)	13.3 (15)	15.1 (17)	0.13 (23)	3.02 (34)
Response factor, mg m <sup>-3</sup> V <sup>-1</sup>	168 ( 4.8)	135 (13)	139 (13)	121 (15)	170 (24)	165 (13)
CO, ppm (n=2)	4.24	7.52	7.61	7.43	0.063	2.47
Av.ppm CO/av mg/m <sup>3</sup> (filter)	4.3	4.0	4.2	4.1	3.3	5.0

\* M = mainstream; S = sidestream

## DISCUSSION

The respirable dust concentration determined from filter samples was constant during the transportation through the 30-m tube. This means that there was no significant loss of ETS. Possible differences caused by humidity changes in the dust samples were not detected because the filters had been conditioned in the room with the microbalance before being weighed. The standard deviation of the five simultaneous measurements was less than 10% of the mean, with the exception of the experiments with the lower concentrations (<0.2 mg/m<sup>3</sup>). These exceptions were mostly found in experiments with mainstream smoke. It

was impossible to evaluate the mainstream smoke in this test set-up; because of the small amount of mainstream smoke in the total smoke, the experiments with the total smoke and the sidestream smoke could be considered essentially comparable. A respirable dust concentration of 0.2 mg/m<sup>3</sup> corresponded with about 20 µg of dust on the filter. The detection limit of the microbalance was about 5 µg.

The actual dust concentration could increase at high relative humidity levels by absorption of water vapour; the continued increase of the response of the Hund with ageing of ETS (>72%, Experiments 12, 14, 15, and 16) indicated an increasing particle diameter.

Table 2. Continued.

Experiment no.	19	20	21	22	23	24
Type cigarettes	B	B	B	B	B	B
Type ETS*	S	S	M+S	M	M	M+S
RH, % (av)	23.5	24	30.5	29	33	8.6
air speed, m s <sup>-1</sup> (av)	0.171	0.172	0.172	0.173	0.076	0.172
Average for five sample points (relative standard deviation, %)						
Filter, mg m <sup>-3</sup>	0.32 (5.7)	0.098 (8.0)	0.101 (15)	0.040 (22)	0.232 (4.6)	0.308 (5.0)
Hund, mV	1.96 (7.7)	1.41 (9.8)	1.57 (16)	0.18 (9.8)	1.13 (5.0)	1.92 (5.2)
Response factor, mg m <sup>-3</sup> V <sup>-1</sup>	162 (9.4)	71 (15)	66 (23)	226 (20)	226 (7.1)	161 (4.2)
CO, ppm (n=2)	1.67	1.15	-	-	-	1.28
Av.ppm CO/av mg/m <sup>3</sup> (filter)	5.2	11.7	-	-	-	2.4

\* M = mainstream; S = sidestream

Experiment no.	25	26	27	28
Type cigarettes	C	C	C	C
Type ETS*	M+S	M+S	M+S	M+S
RH, % (av)	18.2	19.3	16.4	17.5
air speed, m s <sup>-1</sup> (av)	0.184	0.184	0.184	0.184
Average for five sample points (relative standard deviation, %)				
Filter, mg m <sup>-3</sup>	0.411 (5.6)	0.511 (8.1)	0.487 (5.0)	0.436 (7.7)
Hund, mV	2.33 (5.1)	2.53 (8.3)	2.74 (5.8)	3.22 (5.7)
Response factor, mg m <sup>-3</sup> V <sup>-1</sup>	177 (7.2)	204 (5.2)	177 (4.4)	135 (6.9)

\* M = mainstream; S = sidestream

The response and the response factor of the Hund ( $\text{mg m}^{-3} \text{V}^{-1}$ ) was constant at different ages of the ETS (standard deviation less than 10%) in the relative humidity level range of 8-72% and at respirable dust concentrations of not less than  $0.2 \text{ mg/m}^3$ .

At humidities >72%, the response increased with age and the response factor decreased as ETS aged. This could be due to ETS particles growing as a result of condensation of water vapour. Some examples are given in Table 3.

If the experiments with Type B cigarettes with respirable dust concentrations  $<0.2 \text{ mg/m}^3$  and/or with relative humidity levels >72% were excluded, ten experiments remained for evaluation of the influence of the RH.

The average response factor ( $n = 10$ ) was  $177 \text{ mg m}^{-3} \text{V}^{-1}$  with a standard deviation of  $14.5 \text{ mg m}^{-3} \text{V}^{-1}$  (8.2%). The observed range was  $162\text{-}205 \text{ mg m}^{-3} \text{V}^{-1}$  (Fig. 2).

There was a moderate agreement between the gravimetric results and the results of the FH  $\beta$ -dust monitor. Deviations of up to  $\pm 30\%$  were found (exceptional result of Experiment 20 excluded). There was no relation between deviations and RH level. The CO-concentrations at both sides of the test tube corresponded closely.

For the measurements in the passenger cabin of an MD-80 aircraft during flight, as part of the study on improving the air quality in passenger cabins of aircrafts (Crommelin and Dubbeld 1990), Type C

Table 3. Influence of relative humidity on response of the Hund (examples). Experiments with Type B cigarettes.

Experiment no.	7	14	11	12
RH, %	53	79	50	77
air velocity, $\text{m s}^{-1}$ (av)	0.085	0.088	0.168	0.169
response Hund, mV (av)				
sample point 1	7.85	12.1	4.38	6.63
2	8.36	13.9	4.79	7.77
3	7.70	14.8	4.60	8.62
4	8.09	15.6	4.70	14.3
5	8.56	16.9	4.50	13.3
response factor Hund (av), $\text{mg m}^{-3} \text{V}^{-1}$				
sample point 1	194	161	212	164
2	172	137	173	129
3	196	123	183	129
4	173	115	177	73
5	167	139	211	88

) M = mainstream  
S = sidestream

cigarettes were used to calibrate the Hund tyndallometer. Four experiments were performed in the test duct with this type of cigarettes. The relative humidity level was kept at about 20%, which is representative for the flight.

The average response factor of the four experiments was  $173 \text{ mg m}^{-3} \text{V}^{-1}$  with a relative standard deviation of 16.5%. This is relatively high, in comparison with Type A and B cigarettes. The number of experiments, however, was limited ( $n = 4$ ).

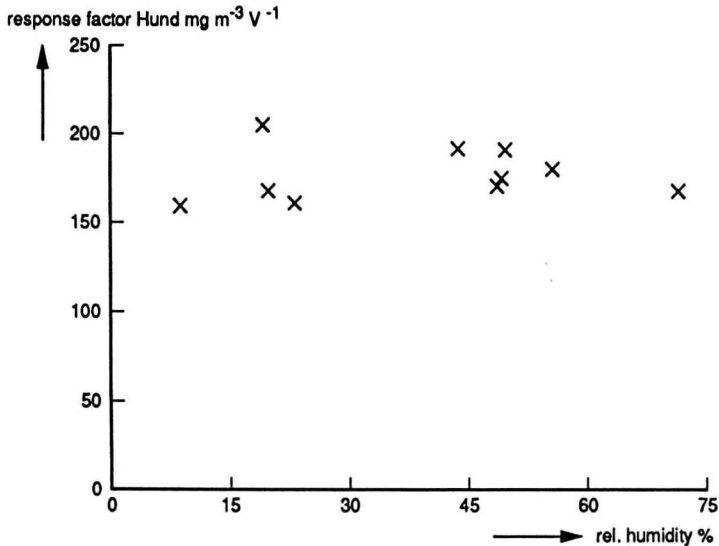


Fig. 2. Average response factors of Hund tyndallometer for relative humidities <72% and Type B cigarettes.

### EVALUATION OF THE RESULTS OF MEASUREMENTS TAKEN DURING SAS FLIGHTS

Laboratory research pointed out that it was impossible to achieve reliable results with extremely low respirable dust concentrations. The gravimetric dust concentration should be at least  $200 \mu\text{g}/\text{m}^3$  for a one-hour sampling period with the minimum amount of collected dust of  $20 \mu\text{g}$  (sampling flow rate  $1.9 \text{ L}/\text{min} = 114 \text{ L}/\text{h}$ ) (cyclone requires flow rate of  $1.9 \text{ L}/\text{min}$  for correct operation). The response of the Hund should be not less than  $1 \text{ mV}$ , corresponding to about  $100\text{-}200 \mu\text{g}/\text{m}^3$ .

Many of the measurements of the respirable dust concentration performed during the SAS flights gave results of less than  $20 \mu\text{g}$  of collected dust on the filters and less than  $1 \text{ mV}$  as average output of the Hund tyndallometer. Moreover, the zero calibration of the Hund with a Millipore filter in the inlet showed

sufficient variations that a correction should be applied.

These corrections were applied by considering the base line of the Hund signal as the zero signal. These corrections varied between  $-1 \text{ mV}$  to  $+1 \text{ mV}$ . All results with an average output of less than  $1 \text{ mV}$  were rejected. Also the results with gravimetric dust concentrations of less than  $100 \mu\text{g}/\text{m}^3$  were rejected. The average flight period was about two hours, so that a dust concentration of  $100 \mu\text{g}/\text{m}^3$  corresponded to a dust load on the filter of about  $20 \mu\text{g}$ .

After rejecting these results, 100 pairs remained with a dust concentration range between  $100$  and  $600 \mu\text{g}/\text{m}^3$  and an average output of the Hund between  $1\text{-}4.2 \text{ mV}$ . The average calculated response factor for these 100 pairs of measurements was  $131 \text{ mg m}^{-3} \text{ V}^{-1}$  with a standard deviation of  $32 \text{ mg m}^{-3} \text{ V}^{-1}$ . The coefficient of variation is  $24\%$ . The range is  $58\text{-}227 \text{ mg m}^{-3} \text{ V}^{-1}$  (see Fig. 3 for the

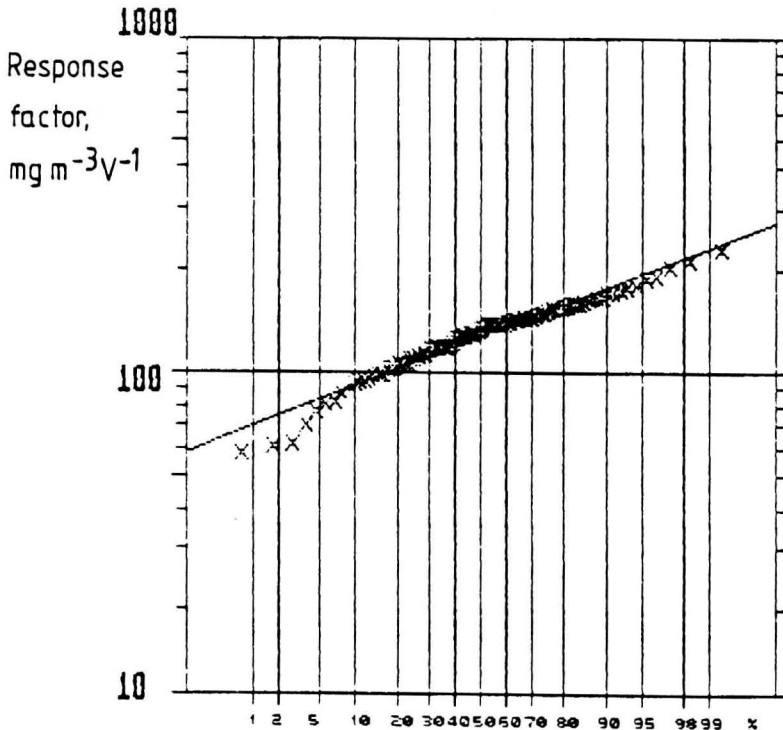


Fig. 3. Cumulative frequency distribution of the response factor of the Hund tyndallometer.

GM =  $127 \text{ mg m}^{-3} \text{ V}^{-1}$ ; GSD = 1.29; AM =  $131 \text{ mg m}^{-3} \text{ V}^{-1}$ ; STD =  $32 \text{ mg m}^{-3} \text{ V}^{-1}$  (24%).

cumulative frequency distribution of the response factor.)

Although the Hund tyndallometer was not intended to be used as an absolute instrument for measuring respirable dust concentrations, but rather to be indicative of smoking activities in aircrafts (example Fig. 4), it was possible to make an estimation of the respirable dust concentration with this instrument. The respirable dust concentration can be estimated with an accuracy of  $\pm 50\%$  within a 95% confidence interval. This holds only for concentrations of more than  $100 \mu\text{g}/\text{m}^3$ ; during the SAS flights 100 out of 190 measurements (53%) fell within this range. Almost all measurements in the business/no-smoking class had to be rejected because of this requirement.

The average response factor of the SAS-flight measurements ( $131 \text{ mg m}^{-3} \text{ V}^{-1}$ ) was less than the average response factor for Type B and Type C cigarettes ( $177$  and  $173 \text{ mg m}^{-3} \text{ V}^{-1}$ ). The average response factor for Type A cigarettes ( $138 \text{ mg m}^{-3} \text{ V}^{-1}$ ) corresponded well. Possibly, the particle size distribution of the Types B and C filter cigarettes is different from that resulting from cigarettes smoked in the aircraft.

## CONCLUSIONS

The Hund tyndallometer has been calibrated with ETS in a test duct with a smoking machine according to ISO Standard 7210 against the gravimetric respirable dust concentration using Teflon membrane filters. An accurate calibration was impossible when the output was less than 1 mV. This is 0.1% of the range (1 V) or and corresponds to a particle concentration of  $100 \mu\text{g}/\text{m}^3$ .

Experiments with mainstream smoke alone produced too low a signal to obtain reliable results. The results of side stream alone and the mixture of mainstream and sidestream smoke could be considered comparable.

There was no effect of ageing ETS for periods up to 7 min as measured by the output of the Hund tyndallometer at relative humidities between 8 and 72%.

The average response factor for RH in range 8-72% was  $138 \text{ mg m}^{-3} \text{ V}^{-1}$  for Type A cigarettes ( $n=4$ , coefficient of variation  $\text{CV} = 20\%$ );  $177 \text{ mg m}^{-3} \text{ V}^{-1}$  for Type B ( $n=10$ ,  $\text{CV} = 8.2\%$ ), and  $173 \text{ mg m}^{-3} \text{ V}^{-1}$  for Type C ( $n=4$ ,  $\text{CV} = 16.5\%$ ).

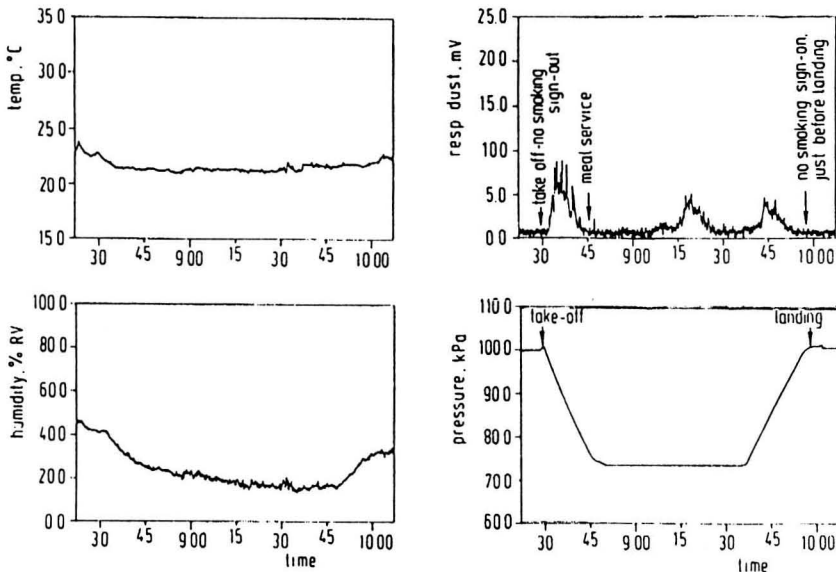


Fig. 4. Output of a data logger during one flight. The average gravimetric dust concentration was  $133 \mu\text{g m}^{-3}$ .

There was no well-defined effect of the relative humidity level on the response factor in the range between 8 and 72% RH.

At relative humidity levels over 72%, the response increased and the response factor decreased with increasing age; this could be due to a growth of the particle by condensation of water vapour.

The average response factor of the Hund for the measurements during the SAS flights was  $131 \text{ mg m}^{-3} \text{ V}^{-1}$ , after rejecting unreliable results which did not meet the Hund preconditions (gravimetric respirable dust concentration  $100 \text{ } \mu\text{g/m}^3$ ; output of the Hund 1 mV) with a coefficient of variation (CV) of 24%. The results of Type A cigarettes in the test duct agreed well with this result. The results of Type B and Type C cigarettes led to a higher response factor. The type of cigarette may affect the response factor of the Hund.

The Hund tyndallometer could be a useful instrument in collecting additional information on peak concentrations in real time and could give a good estimation of the gravimetric respirable dust concentration at levels higher than  $100 \text{ } \mu\text{g/m}^3$ , provided the instrument is calibrated in situ against the gravimetric method and RH levels are lower than 72%.

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# THE BACKGROUND ADJUSTMENT IN RISK ASSESSMENT OF ENVIRONMENTAL TOBACCO SMOKE AND LUNG CANCER

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Most epidemiologic studies of environmental tobacco smoke (ETS) and lung cancer in nonsmokers have used marriage to a smoker as the ETS exposure index. Several risk assessments of ETS and lung cancer have calculated a summary relative risk from a meta-analysis of epidemiologic studies, and have increased that relative risk by applying a "background" adjustment. The aim of the background adjustment is to compensate for the ETS exposure received by nominally unexposed subjects, namely persons married to nonsmokers. Estimates of the ratio of ETS exposure of subjects married to smokers to that of subjects married to nonsmokers have been based on measurements of cotinine, a metabolite of nicotine, in body fluids. The validity of those estimates of relative ETS exposure is questionable, because of analytical problems and the possibility of exposure to nicotine from sources other than ETS. Attempts to explain differences between relative risks reported in U.S. and foreign studies of ETS and lung cancer on the basis of lower U.S. background exposure ratios, or higher foreign ETS exposures, are not supported by the available evidence.

## INTRODUCTION

In recent years, there has been considerable interest in a possible relationship between exposure to environmental tobacco smoke (ETS) and lung cancer. Reports have been published of a number of epidemiologic studies that address this issue, and several reviews and summary estimates of risk have appeared, including one by the National Research Council (NRC 1986). More recently, the U.S. Environmental Protection Agency (EPA) issued a draft report of an ETS-lung cancer risk assessment (EPA 1990). The EPA report calculated a summary ETS-lung cancer relative risk for women of 1.41 from a meta-analysis of 22 epidemiologic studies. A correction for bias arising from misclassification of smoking status reduced the summary relative risk to 1.28. The EPA report then adjusted the summary relative

risk upwards to 1.48, to correct for "background" exposure of the "unexposed" comparison groups in the ETS studies, which for most studies consisted of women married to nonsmokers.

The rationale for a background correction is that the nominally unexposed subjects, namely women married to nonsmokers, may be exposed to ETS outside the home or by smokers in the home other than the spouse. If ETS exposure in fact produced an increased risk of lung cancer, then the relative risk for women married to smokers compared to women with no ETS exposure would be higher than the relative risk when the comparison group is women who are married to nonsmokers but who may receive some ETS exposure from other sources.

This paper describes the calculation of the background adjustment, its underlying assumptions, and

the logical basis for its use. It also examines the proposition that different degrees of background exposure bias might account for the fact that ETS-lung cancer associations reported in foreign studies are generally higher than those reported in U.S. studies. Since background adjustments have typically been based on a comparison of cotinine concentrations in body fluids of subjects married to smokers and of subjects married to nonsmokers, the paper reviews the relevant cotinine studies and assesses the reliability of their results for the purpose of background adjustment.

#### CALCULATION OF THE BACKGROUND ADJUSTMENT

The logic of the adjustment calculation is as follows. Suppose with the EPA report that ETS exposure is causally related to lung cancer risk, and that the lung cancer relative risk of ETS is linearly related to exposure, that is,  $RR = 1 + \beta d$ , where  $d$  is a measure of ETS dose and  $\beta$  is a coefficient with a value greater than zero. Suppose also that women married to smokers receive  $r$  times as much ETS exposure as women married to nonsmokers, and that  $r$  is greater than 1. Then, if the exposure of women with non-smoking husbands is  $d_{mns}$  (assumed greater than zero), the exposure of women with smoking husbands is  $d_{ms} = r d_{mns}$  ( $mns$  denotes marriage to a nonsmoker and  $ms$  denotes marriage to a smoker). Since  $r$  is the ratio of the ETS exposure of a woman married to a smoker to the exposure of a woman married to a nonsmoker, it is referred to here as the spousal smoking ratio or the background ETS exposure ratio.

Based on the above assumptions, the relative risk for women with smoking husbands compared to women with no ETS exposure is

$$RR_{ms} = 1 + \beta r d_{mns}, \quad (1)$$

while the relative risk for women with nonsmoking husbands compared to those with no ETS exposure is

$$RR_{mns} = 1 + \beta d_{mns}. \quad (2)$$

Consequently, the relative risk of women with smoking spouses compared to those with nonsmoking spouses is

$$RR_{ms:mns} = (1 + \beta r d_{mns}) / (1 + \beta d_{mns}). \quad (3)$$

Note that under the assumption  $r > 1$ ,  $RR_{ms:mns} < r$ , since it is equal to

$$r - (r - 1) / (1 + \beta d_{mns}).$$

Given the values of  $RR_{ms:mns}$  and  $r$ , we can solve Equation (3) to get the value of  $\beta d_{mns}$ . That is,

$$\beta d_{mns} = (RR_{ms:mns} - 1) / (r - RR_{ms:mns}). \quad (4)$$

Substituting in Equation (1), we get

$$RR_{ms} = 1 + r(RR_{ms:mns} - 1) / (r - RR_{ms:mns}), \quad (5)$$

while from Equation (2) we get

$$RR_{mns} = 1 + (RR_{ms:mns} - 1) / (r - RR_{ms:mns}). \quad (6)$$

The analysis above is in terms of "true" relative risks, not reported relative risks. If in a particular study or meta-analysis the reported  $RR_{ms:mns}$  is smaller than the value assumed for  $r$ , Equations (5) and (6) can be applied to the reported  $RR_{ms:mns}$  to derive estimated values for  $RR_{ms}$  and  $RR_{mns}$ . However, though it is not possible under the assumptions of the model for the true  $RR_{ms:mns}$  to be greater than the true  $r$ , a reported  $RR_{ms:mns}$  may be greater than an estimated  $r$ . In this case, Equations (5) and (6) cannot be applied. Such an event could be due to one or more of three circumstances:

- (1) the assumed linear dose-response model is wrong;
- (2) the estimated value of  $r$  is smaller than the true value;
- (3) the reported value of  $RR_{ms:mns}$  is greater than the true value.

Figure 1 illustrates the effect of the background adjustment for values of the background exposure ratio,  $r$ , ranging from 1.75 to 3.0. The graph shows that the adjusted relative risk is quite sensitive to the value assumed for  $r$ . If the reported relative risk is 1.20 for example, then the adjusted relative risk is 1.64 for  $r = 1.75$ , but is only 1.33 for  $r = 3$ . However, if the reported relative risk is close to 1.0, the background adjustment can have only a small effect even if the assumed value of  $r$  is relatively small. For instance, with  $r = 1.75$ , a reported relative risk of 1.02 becomes, after adjustment, 1.05.

The assumption that exposure to ETS is causally related to lung cancer is essential for the validity of the background adjustment. If ETS exposure does not increase the risk of lung cancer, then the true relative risk of lung cancer in nonsmoking women due to spousal ETS exposure is 1, that is,  $RR_{ms:mns} = 1$ , and of course  $RR_{ms}$  is also 1, irrespective of the value of  $r$ , the background ETS exposure ratio. It follows that



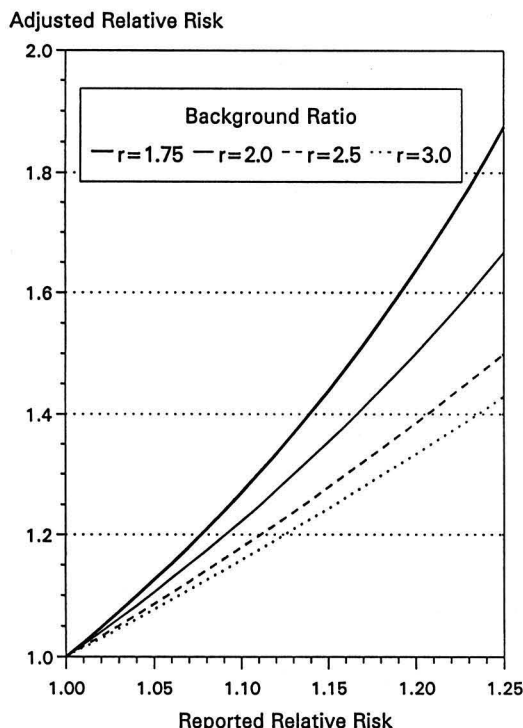


Fig. 1. Relation between reported relative risk and adjusted relative risk for values of  $r$  ranging from 1.75 to 3.0.

applying a background adjustment to an observed relative risk, and then using the higher adjusted relative risk to support a causal conclusion, would constitute an invalid circular argument. A causal judgment must be made before a background adjustment can validly be applied. For this reason, it would be inappropriate to apply a background adjustment to a summary relative risk from a meta-analysis, if the relative risk in question is not significantly greater than 1. In this case a causal inference is not supported by the data.

The EPA report assumed that  $r = 3$ , on the basis of data on urinary cotinine concentrations in subjects married to nonsmokers and to smokers (this choice is discussed later). Since the reported value of  $RR_{ms:mns}$  from the meta-analysis of the epidemiologic studies was 1.28, after adjustment for smoker misclassification bias, the report's estimated value of  $RR_{ms}$  was

$$1 + 3(1.28 - 1)/(3 - 1.28) = 1.48.$$

Similarly, the estimated value of  $RR_{mns}$  was

$$1 + (1.28 - 1)/(3 - 1.28) = 1.16.$$

The EPA report's background adjustment involves three assumptions:

- (1) lung cancer risk is linearly related to ETS dose;
- (2) relative ETS dose in nonsmokers can be accurately estimated from concentrations of cotinine (a metabolite of nicotine) in body fluids;
- (3) current ETS exposure patterns reflect past exposures among lifetime nonsmokers.

If the hypothesis that ETS is causally related to lung cancer is accepted, and these assumptions are at least approximately correct, then the background adjustment process is a reasonable way to estimate the risk of ETS exposure relative to zero exposure. However, all three assumptions are open to question. The second assumption is particularly dubious, for the reasons given below in the review of the cotinine studies.

## DIFFERENCES BETWEEN U.S. AND FOREIGN RELATIVE RISKS

Relative risks reported in epidemiologic studies of ETS and lung cancer in the U.S. are generally lower than those reported in foreign studies. For example, Brown (1990) calculated summary relative risks for women, adjusted for smoker misclassification bias, of 1.02 from nine U.S. case-control studies, 1.41 from ten Asian case-control studies, and 1.37 from four European case-control studies. The summary relative risk for the 14 Asian and European studies is 1.38, which is significantly different from the relative risk of 1.02 for the nine U.S. studies ( $p = 0.02$ ). Inconsistency between reported risks for the U.S. and for other countries is important for two reasons. First, if it cannot be explained it increases the likelihood that the weak reported associations are the result of bias and confounding. Second, it indicates that it is invalid to combine non-U.S. data with U.S. data in order to estimate U.S. risk.

In a discussion of differences between U.S. and non-U.S. results, the EPA report (1990) suggested that spousal smoking ETS exposure among Japanese wives may be higher than among U.S. wives, and that the non-spousal (background) ETS exposure of Japanese wives may be lower than that of U.S. wives. If so, the background ratio,  $r$ , would be lower in the U.S. than in Japan, which might explain lower reported lung cancer relative risks from ETS in the U.S. studies. Differences in housing, employment, and social customs between the two countries were mentioned as possible reasons for variations in ETS exposure patterns, but no hard evidence was offered. The EPA report also suggested that similar differences in spousal and background ETS exposures may prevail in other Asian and in European countries.

Could realistic assumptions about background ETS exposure ratios explain the differences in the relative risks calculated by Brown (1990)? The cotinine studies reviewed below do not suggest that the background ratio for U.S. women is below 1.5 or that the ratio for foreign women is above 3.0. Assuming an  $r$  of 1.5 for the U.S. and of 3.0 for other countries would give an estimated  $RR_{ms}$  of 1.06 for the nine U.S. studies, of 1.77 for the ten Asian studies, and 1.68 for the four European studies. The magnitude of the differences in relative risk estimates is increased by these background adjustments. For the adjusted U.S. relative risk to be comparable to the foreign adjusted relative risk, the assumed U.S. value of  $r$  would have to be about 1.05, which is implausibly low.

An alternative explanation for the U.S. and foreign estimated risk differences might be that ETS exposures are higher in foreign populations. Consider the adjusted relative risks of 1.06, 1.77, and 1.68 for the U.S., Asian, and European studies, respectively, calculated in the previous paragraph on the basis of  $r$  values of 1.5 and 3.0 for the U.S. and foreign countries respectively. Assuming again, along with the EPA report, that lung cancer relative risk is linearly related to ETS exposure, that is,  $RR = 1 + \beta d$ , the estimated background-adjusted U.S. relative risk, 1.06, can be expressed as  $1 + \beta d_{ms,U.S.}$ , and the estimated Asian relative risk, 1.77, can be expressed as  $1 + \beta d_{ms,Asian}$ . (Note that  $d_{ms}$  represents exposure from all sources, background and spousal smoking.) Therefore, the ratio of the ETS exposure of Asian women married to smokers to that of U.S. women married to smokers,  $d_{ms,Asian}/d_{ms,U.S.}$ , is estimated as  $0.77/0.06 = 12.83$ . A similar calculation for the European studies gives a ratio of 11.33. In other words, to account for the U.S. versus foreign risk difference, the total ETS exposure (including background exposure) of foreign women married to smokers would have to be 11 to 13 times greater than the exposure of U.S. women married to smokers, an implausible scenario that is inconsistent with the results of the cotinine studies discussed below.

## COTININE STUDIES

The EPA report's choice of  $r = 3$  for the ETS exposure ratio of women with and without smoking husbands was the same as that used in the National Research Council (NRC 1986) report on ETS, and was based on a study by Wald and Ritchie (1984) of urinary cotinine concentrations in a sample of 121 nonsmoking married men in England. Lee (1987) found a similar ratio for salivary cotinine concentrations in a sample of several hundred men and women, also in England. The results of these and other cotinine studies are summarized in Table 1. The background ratios shown in Table 1 exhibit considerable variability. The ratio for women in Lee's study is substantially higher than those for women in the U.K. studies of Proctor et al. (1991) and Thompson et al. (1990), and is also higher than the female ratio in the Cummings et al. (1990) U.S. study. On the other hand, Lee's ratio is similar to those for the Riboli et al. (1990) ten-country study. Several of these studies are reviewed below, to see what light they shed on differences between ETS background exposures and ratios in the U.S. and foreign countries.

Cummings (1990) reported background ETS exposure ratios of 1.55 and 1.27 for nonsmoking mar-

Table 1. Cotinine in nonsmokers.

Study	Country	Subjects	Average Cotinine <sup>1</sup>			Av. Cotinine <sup>1</sup> Zero ETS <sup>2</sup>
			Spouse nonsmkr.	Spouse smkr.	Ratio (r)	
Cummings (1990)	US	225 f	6.8	10.5	1.55	6.2
		241 m	8.6	10.9	1.27	
		162 f+m				
Lee (1987)	UK	340 f	0.8 <sup>3</sup>	2.2 <sup>3</sup>	2.91	
		448 m	1.2 <sup>3</sup>	3.8 <sup>3</sup>	3.10	
Proctor <i>et al.</i> (1991)	UK	24 empl. f	1.3 <sup>3</sup>	1.7 <sup>3</sup>	1.31	1.0 <sup>3</sup>
		28 unempl. f	1.1 <sup>3</sup>	2.2 <sup>3</sup>	2.00	
		15 f				
Riboli <i>et al.</i> (1990)	10 countries	643 empl. f	4.5 <sup>4</sup>	10.1 <sup>4</sup>	2.24	3.0 <sup>4</sup>
		679 unempl. f	3.1 <sup>4</sup>	9.5 <sup>4</sup>	3.06	
		334 f				
Thompson <i>et al.</i> (1990)	UK	99 f	7.4	11.0	1.49	5.6
		82 m	8.3	23.3	2.81	
		51 f+m				
Wald & Ritchie (1984)	UK	121 m	8.5	25.2	2.96	

<sup>1</sup> Urinary cotinine concentration in *ng/mL*, except as otherwise noted.

<sup>2</sup> Self-reported zero exposure to ETS.

<sup>3</sup> Salivary cotinine concentration in *ng/mL*.

<sup>4</sup> Urinary cotinine standardized for creatinine, in *ng/mg* creatinine.

ried women and men, respectively, based on a sample of 225 women and 241 men in Buffalo, N. Y. (the ratio for men and women combined was 1.38). These 466 married subjects were part of a sample of 663 nonsmokers whose urinary cotinine levels were measured by Cummings *et al.* (1990). For the 663 subjects, the ratio of urinary cotinine in those reporting ETS exposure in the preceding four days to those reporting no ETS exposure was 1.56, very little higher than the spousal smoking ratio of 1.38 for the 466 male and female married subjects. This similarity was largely due to the fact that the mean cotinine level of 6.22 *ng/mL* for the 162 subjects reporting no ETS exposure in the previous four days was not much less than the mean level of 7.75 *ng/mL* for the 372 subjects married to nonsmokers. The fact that the mean cotinine level of persons reporting no exposure was so close to that of persons married to nonsmokers suggests that in both groups these levels may be largely due to factors other than ETS exposure, such as dietary sources of nicotine (Idle 1990) and

artifacts of the cotinine bioassay. The accuracy of both the gas chromatography and the radioimmunoassay techniques for measuring low levels of cotinine in body fluids is uncertain (Biber *et al.* 1987; Letzel *et al.* 1987; Anderson *et al.* 1991). Cotinine levels were similarly close for these two groups of subjects (self-reported zero ETS exposure and married to nonsmokers) in the studies of Thompson *et al.* (1990), Proctor *et al.* (1991), and Riboli *et al.* (1990), which are discussed below.

The difference between Cummings *et al.*'s spousal smoking ratio for men (1.27) and that of Wald and Ritchie (1984) (2.96) is due to the fact that the mean urinary cotinine for nonsmoking men married to smokers differed substantially between the two studies: 10.9 *ng/mL* for the Cummings study and 25.2 *ng/mL* for the Wald and Ritchie study. The mean levels for men married to nonsmokers were almost the same in the two studies: 8.6 *ng/mL* and 8.5 *ng/mL*, respectively.

Thompson et al. (1990) studied urinary cotinine levels in 184 self-reported nonsmoking men and women attending a health screening center in London. Excluding three subjects who had cotinine levels over 100 ng/mL, the ratios of mean cotinine levels for those living with a smoker to those living with a nonsmoker were  $23.3/8.3 = 2.81$  for men and  $11.0/7.4 = 1.49$  for women. The mean levels in women of 11.0 ng/mL and 7.4 ng/mL, for those with smoking and nonsmoking spouses or partners, respectively, were almost the same as the levels reported by Cummings (1990) (10.5 ng/mL and 6.8 ng/mL). However, the mean level for men living with smokers, 23.3 ng/mL, was similar to that found by Wald and Ritchie and is much higher than the level of 10.9 ng/mL reported by Cummings. The mean cotinine levels for all subjects reporting no exposure to ETS in the previous seven days was 5.6 ng/mL, which compares with the mean level of 7.9 ng/mL for all subjects living with nonsmokers. As in the Cummings et al. study, the similarity of those levels suggests that they may be both largely due to factors other than ETS exposure.

In another English study, Proctor et al. (1991) measured salivary cotinine and personal exposures to nicotine in 52 nonsmoking women, and questioned them about their exposure to ETS in the previous 24 h. For working women, the ratio of mean cotinine in women living in smoking households to mean cotinine in those living in nonsmoking households was 1.3. For nonworking women the ratio was 2.0. If the measure of exposure to ETS is taken as personal nicotine exposure rather than salivary cotinine level, then the ratios become quite different. For example, the mean nicotine exposure in nonworking women with smoking spouses was  $7.4 \text{ ng/m}^3$ , while in nonworking women with nonsmoking spouses it was  $0.5 \text{ ng/m}^3$ , giving a ratio of 14.8. The authors noted that 15 women who reported no ETS exposure (and had no detectable nicotine exposure) had detectable salivary cotinine, with levels overlapping cases for whom ETS exposure was reported. In 10 of these 15 women, salivary cotinine was higher than the level at the beginning of the 24-h study period, so that ETS exposure before the start of that period could not explain the apparent inconsistency. This led the authors to express doubts about the validity of salivary cotinine information at low levels. It should be noted that detectable airborne nicotine exposure does not necessarily imply ETS exposure. Airborne nicotine was detected in buses in the city of Southampton, U.K., some six months after smoking on buses was banned (Proctor 1990).

A study by Matsukura et al. (1984) reported urinary cotinine:creatinine ratios in men and women in western Japan that were two orders of magnitude higher in nonsmokers and one order of magnitude higher in smokers than those reported by other workers (for example, Muranaka et al. 1988). This study has been criticized on methodological grounds (Adlkofer 1985), but it is worth noting that the reported ratio of urinary cotinine in nonsmokers who lived with smokers ( $n = 272$ ) to urinary cotinine in nonsmokers with no smokers in the home ( $n = 200$ ) was 1.55. This ratio is likely to be close to the ratio for subjects married to smokers versus those married to nonsmokers, so this study does not support the belief that the ETS background ratio in Japan is higher than in the U.S.

The ten-country collaborative IARC study of urinary cotinine levels and self-reported ETS exposure in nonsmokers (Riboli et al. 1990) is a potentially valuable source of information on ETS background exposures and ratios in different countries, including the U.S. and Japan, but unfortunately the investigators have not yet published explicit results pertaining to this issue. Data were collected from 1 369 nonsmoking women at 13 locations in 10 countries: Canada (Toronto), People's Republic of China (Shanghai), Greece (Athens), Federal Republic of Germany (Bremen), Hong Kong, India (Chandigarh), Italy (Turin and Trieste), Japan (Sendai, Miyagi prefecture), and the U.S. (Honolulu, Los Angeles, and New Orleans). An attempt was made to recruit about 100 women at each center, of whom 50% were to be married to a smoker and 50% married to a nonsmoker or unmarried. Within each of these categories, 50% were to be employed outside the home. In most centers, the proportion of women married to smokers was about 50%; there was more variation in the proportion of women employed, from 29% in Los Angeles to 70% in Toronto and 88% in Trieste.

Forty-seven women (3.4% of the total sample) were excluded from analysis because they had cotinine levels relative to creatinine above 50 ng/mg, which suggested that they may have been active smokers. Table 2 shows the age-adjusted mean urinary cotinine levels by center (data read from graph). These means were apparently not adjusted for differences in the sampling proportions of employment status and spousal smoking status. There are three factors that might explain the variation in cotinine means between centers:

(1) differences between centers in the average duration and intensity of exposure to ETS;

Table 2. Cotinine in nonsmokers: the IARC ten-country study (Riboli et al. 1990)

Center	Average Urinary Cotinine (ng/mg creatinine)
Shanghai	2.55
Honolulu	3.75
Chandigarh	4.60
Sendai	4.95
New Orleans	5.15
Turin	6.00
Toronto	6.70
Bremen	7.05
Hong Kong	7.75
Athens	8.05
Warsaw	8.40
Los Angeles	8.50
Trieste	13.75

(2) differences between centers in the relationships between reported duration and intensity of ETS exposure and measured urinary cotinine (that is, differences in regression slopes);

(3) differences between centers in the average "no reported exposure" level of measured urinary cotinine (that is, differences in regression intercepts).

According to the authors, there were no statistically significant differences between the relationships referred to in Item 2. This result seems contrary to the notion that U.S. wives with smoking husbands have lower spousal ETS exposures than do non-US wives because of differences in housing conditions. When ETS exposure duration and intensity were taken into account in a regression analysis, the differences between center cotinine averages were reduced from those shown in Table 2, but were not eliminated. In other words, differences in "no reported exposure" cotinine levels (Item 3) remained.

A question that is pertinent to the present inquiry is how background cotinine levels, as defined by spousal smoking status, varied by country in the IARC study. Although explicit information on this point is not yet available, it appears from the discussion in the previous paragraph that the mean cotinine levels in Table 2 are likely to provide a rough indication of the relative ordering of background cotinine

levels between centers. Los Angeles is at the high end of the list of means in Table 2, but both Honolulu and New Orleans are towards the low end, so there is little to suggest in these data that U.S. women have higher nonspousal ETS exposures than women in other countries. An explicit comparison between countries of cotinine ratios for women married to smokers and to nonsmokers cannot yet be made for the IARC study, but it does not seem likely that the results for the various centers, when available, would suggest that the ratio for U.S. women is very much different from those for women in other countries.

The Riboli et al. paper does provide cotinine information by spousal smoking status for all centers combined. For non-employed women, the ratio of mean cotinine levels for those married to smokers and those married to nonsmokers was  $9.5/3.1 = 3.06$ . For employed women, the ratio was  $10.1/4.5 = 2.24$ . For women reporting no exposure to ETS from any source in the previous four days, the mean cotinine level was 3.0 ng/mg creatinine, which compares with means of 3.1 ng/mg and 4.5 ng/mg for women who were non-employed and employed, respectively, and who were married to nonsmokers. Again, these results suggest that a substantial part of the measured cotinine levels may not be a consequence of ETS exposure.

## OTHER INFORMATION CONCERNING BACKGROUND ETS EXPOSURE

It was noted above that the EPA report's suggestion that Japanese women married to nonsmoking men have a relatively low background ETS exposure was not supported by any hard evidence. It is true that the smoking prevalence among Japanese women (about 15%) is lower than among U.S. women, so that Japanese wives may receive less ETS exposure from other women than do U.S. wives. However, the smoking prevalence among Japanese men (84% in 1966 and 61% in 1988, according to Hirayama 1990) is considerably higher than among U.S. men, so Japanese wives may well receive more ETS exposure from men other than their husbands, including relatives living with them, than do U.S. wives. Such exposure could occur in the home, in the workplace, in public transportation, and in other public places. Matsukura et al. (1984) noted that because of the high proportion of male smokers in Japan, exposure to ETS "is often almost unavoidable in public places."

A substantial proportion of Japanese women are employed outside the home. In the Hirayama Japanese cohort study (Hirayama 1990), 69% of the female subjects were employed in 1966, 44% in agricultural occupations and 25% in other occupations. Although women in agricultural occupations may receive less ETS exposure from fellow-workers than do those in other occupations, there is no reason to suppose that they receive none. In a logistic regression analysis of data from the Hirayama study (Hirayama 1984) subdivided by husband's occupation, there was no difference in the estimates of spousal smoking and lung cancer relative risk for rural and other occupations. If there were a real association between ETS exposure and lung cancer, and if wives of nonsmoking rural workers, many of whom were themselves employed in rural occupations, had a lower background ETS exposure than wives of men in other occupations, one would expect to find a higher lung cancer relative risk in the rural wives of smokers.

Other Japanese studies of ETS and lung cancer also reported a high proportion of employed women. In the Akiba et al. (1986) case-control study in Hiroshima and Nagasaki, 55% of the nonsmoking control women were employed. In the Shimizu et al. (1988) case-control study in the Japanese city of Nagoya, 35% of the nonsmoking control women were exposed to ETS in the workplace.

The argument that Japanese wives with smoking husbands receive more ETS exposure from their husbands than do U.S. wives because they live in smaller

homes cannot be accepted as fact without evidence to substantiate it. As noted above, the results of the IARC study do not appear to support this contention. In any case, considerations other than the size of housing are relevant, particularly the amount of smoking by husbands in the presence of their wives, both in terms of time and numbers of cigarettes smoked.

As in the case of Japan, there are no convincing data to suggest that background ETS exposure is lower in other Asian and European countries than in the U.S., or that the ETS exposure of women married to smoking husbands is higher.

## CONCLUSIONS

The EPA report (1990) used the ETS background adjustment issue as a rationale for the difference between the results of U.S. studies and of foreign studies, on the basis that the background ETS exposure ratio may be lower in the U.S. than in other countries. However, there is little if any evidence available to substantiate such a variation in background ratios. Information from existing cotinine studies is inconsistent, and its reliability as an indicator of ETS exposure is questionable. In any case, no plausible difference in background ratios could explain observed differences between relative risks from U.S. and foreign studies. The unexplained inconsistency between the U.S. and foreign estimates of ETS and lung cancer relative risk argues against a causal interpretation of reported associations, and makes the use of foreign data to estimate U.S. risk unjustified.

The use of a background adjustment is not valid unless a causal link between ETS exposure and lung cancer has been inferred. In particular, if the relative risk from a meta-analysis of ETS and lung cancer studies is not significantly elevated, a causal inference is not justified and a background adjustment cannot validly be applied.

Even if an association between ETS exposure and lung cancer is assumed, the validity of background adjustments based on relative cotinine levels is open to question. The similarity in several studies of the average cotinine levels for persons reporting no ETS exposure and for persons living with nonsmokers suggests that background cotinine levels may be substantially due to factors other than ETS exposure, such as dietary nicotine and analytical artifact. If so, background ratios derived from cotinine measurements underestimate the ETS exposure of persons married to smokers relative to the exposure of per-

sons married to nonsmokers, and background adjustments using those ratios are excessive.

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# THE USE OF REACTIVE COATINGS ON FILTERS FOR THE REMOVAL OF INDOOR GASEOUS POLLUTANTS

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We have shown that formaldehyde can be removed from the air when the fibre-glass filters are coated with the polymer polyethylenimine, mixed with glycerol to keep it from hardening. By increasing the amount of polymer applied to the filter, it has been possible to extend the life of the coating to almost four months during which time the level of formaldehyde in the house is well within acceptable levels. It was also shown that in the process of air-conditioning a house or dehumidifying the air, the formation of water of condensation will remove part of the water soluble pollutants in the air.

## INTRODUCTION

The issue of indoor air quality is of increasing concern. Many problems have been identified and solutions have been proposed which vary in efficiency and costs (Indoor Air '90). The recent problem of urea formaldehyde foam insulation (UFFI) installed in Canadian homes and then removed due to the release of formaldehyde has promoted much research on indoor air (Tataryn 1983). A majority of Canadian homes have a forced-air heating system in which a furnace fan circulates the air in the house over a heat-exchanger. A filter is usually present between the fan and the heat-exchanger. Indoor air levels of formaldehyde can be reduced by coating the particle filter (usually fiberglass) in the furnace with a reactive polymer which chemically reacts with the formaldehyde as the air is circulated by the furnace fan (Gesser 1984). Initial results showed the polyethylenimine coating to be effective for only two weeks. The hard polymer surface became coated with

reaction products which remained on the filter surface and prevented further reaction.

We have recently shown (Gesser 1990) that it was possible to extend the useful life of the reactive coating to at least one month by plasticizing the polyethylenimine coating with glycerol. Glycerol keeps the coating soft and tacky and allows the exchange between the surface and bulk material. Since the furnace filter in the house is meant to be changed every month, a coating which can last a month fulfills the normal domestic need.

We showed also (Gesser 1990) that since the polyethylenimine is basic, it can react with and neutralize acidic gases such as  $\text{NO}_x$ ,  $\text{SO}_x$ ,  $\text{H}_2\text{S}$ , and hydrogen halides.

This experimental procedure tested the removal of other gases and the possible commercial application of thicker coatings with the view of further extending the lifetime of the coatings to a few months.

## EXPERIMENTAL

A one-storey bungalow with basement (total volume approx. 1000 m<sup>3</sup>) was selected for testing. The forced air furnace had two 2.5 cm thick fiberglass filters (51 × 63.5 cm and 41 × 63.5 cm). Formaldehyde was generated by heating paraformaldehyde in a weighable vial (at a constant temperature) placed near the air intake located in a basement room. The loss of paraformaldehyde could be determined daily and compared with the concentration of formaldehyde in the house. After several trials it was possible to adjust the temperature to give a formaldehyde concentration of about 75 µg/m<sup>3</sup> corresponding to a weight loss in paraformaldehyde of about

60 mg/h. The normal exchange rate of air in the house was determined to be 0.83 air changes/h.

Formaldehyde was analysed by the pararosaniline method previously described (Gesser 1984).

The polyethylenimine used was Corcat 600 (Hoechst Celanese) and was mixed with glycerol (U.S.P.) in a ratio of two parts of polyethylenimine to one part of glycerol and applied to the filter as a fine spray from a 5% PEI aqueous solution. The filter was air dried before use. The mass of polyethylenimine/glycerol on the two filters was 46 g, compared to 6.2 g polyethylenimine on a 2.5 × 41 × 51 cm fiberglass filter used previously (Gesser 1990).

Table 1. The change in formaldehyde levels in test house due to polyethylenimine coated filter. A = in; B = out.

Date	[H <sub>2</sub> CO] µg/m <sup>3</sup>	Date	[H <sub>2</sub> CO] µg/m <sup>3</sup>	Date	[H <sub>2</sub> CO] µg/m <sup>3</sup>
B		A			
Oct 1	79	Nov 2	24	Dec 16	12
2	62	3	7	17	10
3	80	4	0	19	22
4	70	5	4	22	25
5	75	6	24	23	19
A		8	28	24	0
6	38	10	19	26	8
7	2	12	42	Jan 8	32
8	4	14	28	9	30
9	24	16	20	10	4
10	70	18	1	12	8
12	20	20	5	14	16
14	0	22	24	17	25
17	5	24	0	18	23
18	0	26	17	19	38
19	0	28	4	20	35
20	2	30	8	21	54
21	18	Dec 1	2	22	101
22	16	2	0	23	91
23	0	3	2	25	130
B		4	29	27	72
24	67	6	20	28	73
25	85	7	8	B	
28	107	8	19	Feb 1	64
29	84	9	0	2	42
29	78	10	7	3	75
30	97	12	8	4	85
31	88	14	0	5	103
Nov 1	60				

Laboratory tests were conducted to determine the effectiveness of the polyethylenimine/glycerol coating on the removal of toluenediisocyanate from air. The apparatus used was similar to that previously described for the test performed on acidic gases (Gesser 1990). Reticulated polyurethane foam 2.5 cm thick and 7.3 cm in diameter with 45 ppi was used as the support for the coatings (16 g of a 2:1 mixture of polyethylenimine/glycerol). Air was flowed at 80 mL/min over a toluenediisocyanate reservoir kept at constant temperature to give a concentration of about  $140 \text{ mg/m}^3$  of toluenediisocyanate which then passes through the coated filters. The toluenediisocyanate in air was analysed by gas chromatography before and after passing through the filter.

## RESULTS AND DISCUSSION

Initial results showed that the concentration of formaldehyde in the house fluctuated considerably ( $25 \mu\text{g/m}^3$  to  $70 \mu\text{g/m}^3$ ). It was eventually shown that the major cause of the variations, under sealed conditions with a constant input of formaldehyde, was the operation of the air conditioner and its dehumidifying function. Examination of the water of condensation showed it to contain from 0.4 mg/L to 2.5 mg/L of formaldehyde and depended on the amount of water collected (from 50 mL to 300 mL/h)

which in turn depended on the use of the dishwasher (which vented indoors) and the frequency of showering that occurred as well as the outside temperature.

When air conditioning was stopped, indoor concentration of formaldehyde became reasonably constant at about  $75 \mu\text{g/m}^3$ . When the coated filters were installed, the concentration dropped to low concentrations (0 to about  $20 \mu\text{g/m}^3$ ) and increased again when the coated filters were removed (Table 1).

When the same coated filters were reinstalled in the furnace, the concentration of formaldehyde in the house remained low for an additional two and a half months (Nov. 2 to Jan. 22) and then rose indicating that the coating had been exhausted, i.e., completely reacted with the formaldehyde. These results are plotted in Figure 1. Thus, the greater coating of polyethylenimine on the filter can extend the effective life of the coated filter in removing formaldehyde. When the filters were removed, it was noted that they were covered with a continuous layer of dust. This places a time limit on the usefulness of the coated filter unless dust and particulate matter are removed prior to the coatings. An electrostatic filters could serve to reduce the interference of dust. With a suitable design, it should be possible to extend the effective life of the reactive coating to six months. The coating, as well as the coating after reaction, is

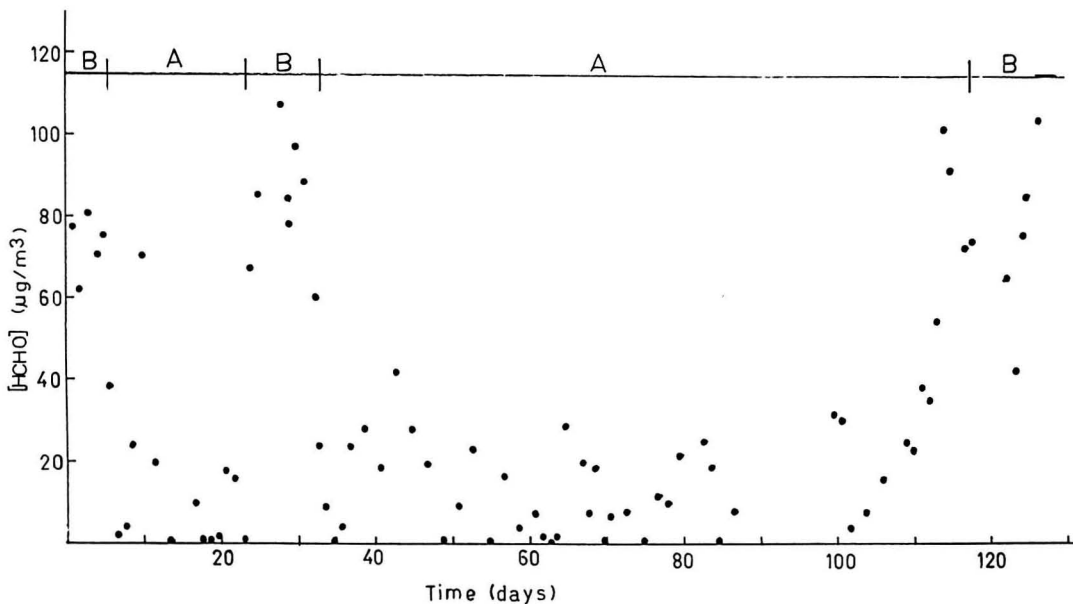


Fig. 1. A plot of the formaldehyde concentration in the house as a function of time. A represents the time when the coated filter is in use. B represents the time when the coated filter is not in use.

harmless and can be readily disposed of since it is not classified as a hazardous waste.

The laboratory tests showed that toluenediisocyanate was removed by both components of the coatings, the polyethylenimine as well as the glycerol used to plasticize the coatings. The effectiveness of the coating to remove such high levels of toluenediisocyanate (reduced to below the STEL of  $150 \mu\text{g}/\text{m}^3$  or the TWA of  $40 \mu\text{g}/\text{m}^3$  (NIOSH 1990) seemed to decrease with exposure. However, when the filter was allowed to rest, it recovered and became highly effective again. This may be related to the diffusion of the polyethylenimine/glyceroltoluenediisocyanate reaction products from the air-coating interface and would be less important at the lower concentrations of toluenediisocyanate usually encountered.

Preliminary tests have shown that basic gases such as ammonia can be readily removed from air by passing the air through a filter coated with the acidic polymer, polyacrylic acid plasticized with glycerol.

An acid/base indicator can be used to show when the coating has been completely reacted.

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## EXHAUST EMISSIONS FROM WORK MACHINERY IN FINLAND

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This study mapped work machines having internal combustion engines that are used in Finland. The aim of the study was to clarify the number and average engine power of machines in use, their annual operating hours, and their loading properties, in order to calculate the exhaust emissions from these machines. The numerical data were gathered from sales figures, vehicle registration, and statistics on production, import, and export. The average load factor was evaluated on the basis of the real fuel consumption in comparison to nominal fuel consumption. Data on the duration operation are based on the number of working hours and the results of earlier studies. There were about 400 000 diesel engine machines in Finland in 1990. The most common types of machines are agricultural tractors, construction machines, and combine harvesters. The total fuel consumption of diesel machinery was 780 000 m<sup>3</sup> accounting for about 30% of the total amount of diesel fuel consumed in traffic. Nitrogen oxide emissions came to 41 million kg, which is 15% of all nitrogen oxide emissions in Finland. Other studied compounds—unburned hydrocarbons, carbon monoxide, and particulate matter—accounted for 5-10% of all emissions in Finland.

### INTRODUCTION

Environmental protection has been an urgent issue in all industrialized countries. The Government of Finland has granted high priority to national and international environmental protection efforts with regard to future energy policy decisions.

Since the late 1970s, the Energy Department of Finland's Ministry of Trade and Industry has funded research on the supply of energy and the environment. A current research and development program, called SIHTI, is studying the emissions of various energy sources at all stages of the energy production chain. The aims are to devise new methods and equipment that would reduce the environmental impact and to organize the collection, analysis, and dissemination

of information on environmental technology transmitting of international knowledge and to promote other forms of cooperation.

The research areas of this program include energy production, fuel chains and their environmental loading, traffic emissions, and alternative fuels. This study of exhaust emissions from work machinery is one of the projects conducted in the research area of traffic emissions.

Traffic emissions have been found to be a major environmental loading factor for carbon monoxide and for oxides of nitrogen. Decisions and practical steps have been taken to reduce traffic emissions (IEA 1991). Emissions arising from work machinery have been ignored because they have been assumed

to be of much less importance in comparison to emissions from cars. The reduction measures which have been implemented or planned deal mostly with cars and trucks. Yet even though emissions from road traffic will be cut in the near future, the relative importance of other emissions will increase.

Little research work has been done on the exhaust emissions from work machinery. A Swedish study reported that the emissions of NO<sub>x</sub> and particulate matter from diesel-operated work machinery are quantitatively similar to the emissions from heavy trucks (Karlson 1990). A Dutch study showed that the diesel fuel consumption of moving work machinery equalled the consumption of heavy truck traffic, and that work machinery produced 10-20% of the total NO<sub>x</sub> and particle emissions as well as 5-10% of the total hydrocarbons and carbon monoxide emissions (Achten 1990). Some basic studies on lowering emissions and on the contribution to total emissions have been carried out in Sweden (Hedbohm 1990; Karlson 1990). The exhaust emissions from work machinery have not been studied in Finland before. Some estimations, based on total energy consumption, have previously been made.

In Finland, a work machine is defined an engine-powered machine or an equipment which as such or with some complementary devices has been designed for a specific task but which is neither a car nor a truck. A work machine may either move by its own energy or it may be transportable.

Data on the number of work machinery and of their use are not as readily available as that for cars, data of which are kept by the authorities in official registers. This may be one reason why work machines have been overlooked as a source of emissions.

The aim of this study was to determine the total number of work machinery used in Finland and to estimate their total amount of emissions.

## MATERIALS AND METHODS

The figures used to characterize each type of machinery here are the total number of machines (N), the average nominal engine power (P), the load factor (k), and the average duration of operation (T, in h/y). The load factor is defined as the relation between operating power and the rated engine power, in percent; it can vary considerably even within a specific machine type. The annual energy consumption (E) can then be evaluated as:

$$E = \frac{NPkT}{100} \quad (1)$$

Further, the amount of exhaust emission components (H<sub>i</sub>) are calculated with nominal emission factors (e<sub>i</sub>):

$$H_i = Ee_i \quad (2)$$

where "i" denotes a specific compound of emission. The emission compounds included in this study were carbon monoxide, hydro-carbons, nitrogen oxides, particulate matter, sulphur dioxide, and carbon dioxide. The specific emission factors used here are similar to a Swedish study (Stenström 1989, 1990), and are given in Table 1. The machines were classified according to the type of fuel consumed.

The total fuel consumption (Q) can be calculated from the nominal fuel consumption (q):

Table 1. Specific emission factors (Stenström 1989, 1990).

COMPONENT	DIESEL ENGINE (g/kWh)	PETROL ENGINE (g/kWh)	
		2-stroke	4-stroke
Hydrocarbons	1-2.5	125-150	10-15
Carbon monoxide	4-5	200	150
Oxides of nitrogen	14-16	2-5	5-10
Particulate Matter	1-1.7	5	0.5
Sulphur dioxide	1	0.1-0.2	0.1-0.2
Carbon dioxide	740-900	1100-1400	1200-1500

$$Q = Eq \tag{3}$$

Equation (3) gives consumption in mass units. To convert this into capacity units, the result must be divided by density. A density of 750 000 g/m<sup>3</sup> for gasoline and 850 000 g/m<sup>3</sup> for diesel fuel was used in this study.

Total fuel consumption enables comparison with other statistics, thus, enabling verification of the results. It is also a good indicator of the significance of a specific type of machines with respect to emissions.

The first finding was that there is no uniform classification for work machines. Several classifications are in use in Finland, but most of them were not suitable for this study. The classification used by the Finnish customs authorities was found to be useful for the present purpose because it is based on an international classification, covers all machines in use, and is also used for production statistics, which is useful when making comparisons against sales figures.

The number of agricultural tractors and combine harvesters was calculated from statistics kept by the State Research Institute of Engineering in Agriculture and Forestry. In the case of forest machinery, the data are based on the knowledge of the Research and

Development Unit of the Forest Industries in Finland. The figures for construction machines, compressors, lawn mowers, etc. were calculated from statistics kept by the Finnish Association of Technical Traders.

It is obvious that all machines which have been sold are not in use. For this reason, only years representing the total duration of operation were taken into consideration.

Import, export, and production statistics were used if sales figures were not available. The vehicle register kept by the Central Statistical Office of Finland and based on the Motor Vehicles Register Centre does not cover all work machines (Central Statistical Office of Finland 1991). Off-road machines, e.g. agricultural tractors, forest tractors, and wheel-based excavators, are usually recorded in the registers. The data from insurance companies proved not to be specific enough because the same type of insurance policy is used to insure different types of machines. The total number of insurance policies can be used only as an estimate of the total number of drivable work machines. Data from some earlier Finnish studies were also used (Alapartanen 1988; Kallberg 1984).

The average annual operating time of work machines decreases as a function of time, so it is useful to classify machines into two groups (Fig. 1).

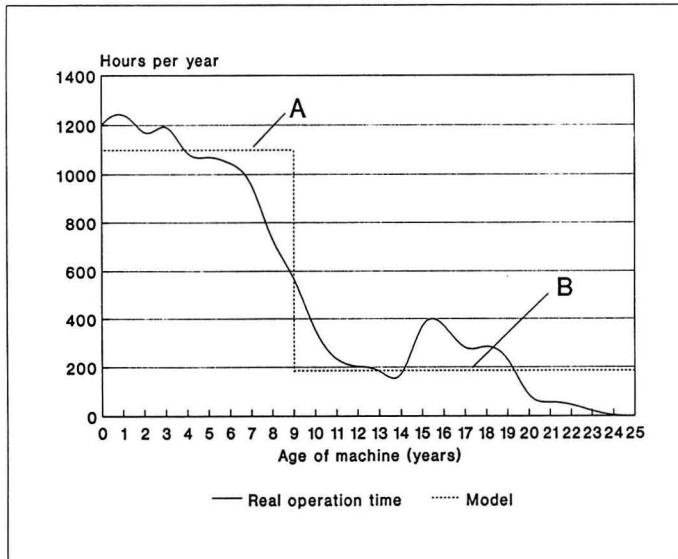


Fig. 1. Model for estimating the average duration of operation (h/y).

New machines are mostly operated by contractors or used in industry, so their annual operating time is high, but as they grow older their operating time decreases. The age at which the annual operating time decreases depends on the type of machine, being about five years for construction machines and about ten years for agricultural tractors.

For forest machinery, the annual operating time is based on working hours recorded by the Association of Machine Entrepreneurs in Finland. In the case of earth-moving machinery, the annual operation time was determined in a similar way, but since no specific statistical data were available, the estimate is based on the average operation rate of the machines (Alapartanen 1988).

The load factor was calculated from the real fuel consumption compared to the nominal fuel consumption. Thus, it does not matter how this load is distributed; it may be steady or varying.

## RESULTS

The number of diesel engine machines in Finland totalled about 390 000 in 1990. It should be noted that at the same time, Finland had a total of 1.9 million passenger cars, 207 000 vans, 9 000 buses and 75 000 trucks, and special road vehicles. Thus, the proportion of off-road machinery is considerable. Drivable work machines account for half of all diesel

engine vehicles (Fig 2). The data on machine types are shown in Table 2.

### *Agricultural machines*

Agricultural tractors constitute the most significant work machine type in Finland. Tractors account for two out of three diesel-powered vehicles. Some very old tractors are still in use, because the annual operation time is quite low, averaging 400 h/y for new tractors and 200 h for tractors more than ten years old. The total number of farm tractors is about 250 000. Some 20 000 tractors are used by contractors or in industry; they are used more actively than agricultural tractors, usually 700-900 h/y. New tractors in Finland have an average engine power of 55 kW; but, older tractors are not so powerful. The load factor is 65% for ploughing and forage harvesting and 80% for harrowing. But, because tractors are also used for many lighter tasks, the average load is about 25% of the nominal power. Agricultural tractors account for one third of all the fuel consumed by machines with diesel engines.

There are about 40 000 combine harvesters in Finland, the annual operating time of which is usually less than 100 h. Their average nominal power is 55 kW and their engine load is steady, with a load factor of 65%.

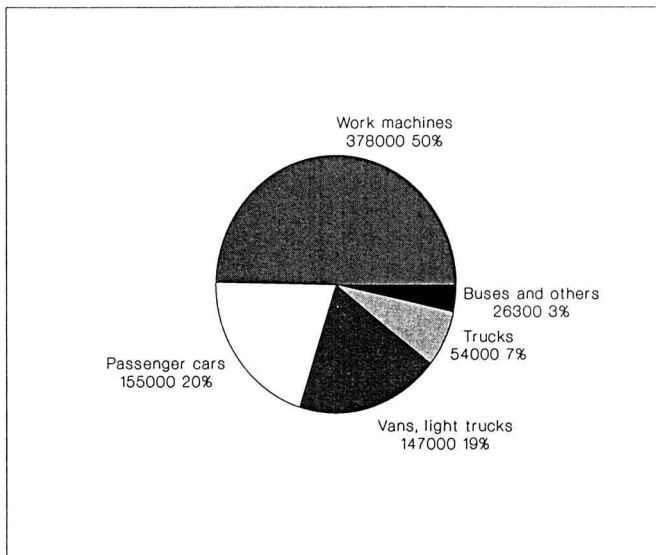


Fig. 2. Number of diesel engine vehicles in Finland in 1990.



Table 2. Machine data.

	ENGINE POWER kW	AVERAGE LOAD FACTOR %	NUMBER	AVERAGE USE h/a	FUEL CON- SUMPTION g/kWh
<b><u>MACHINES WITH A DIESEL ENGINE</u></b>					
Cranes	130	25	900	900	240
Other industrial trucks	30	30	2000	400	250
Fork-lift trucks > 12 t	170	30	500	2000	240
Fork-lift trucks < 12 t A	50	30	2000	2000	250
Fork-lift trucks < 12 t B	30	30	4000	600	260
Bulldozers	110	40	700	700	260
Blade graders	150	40	1300	800	250
Rollers	45	30	1000	500	250
Wheel loaders A	110	35	2400	1400	240
Wheel loaders B	100	35	6600	450	250
Tractor backhoes A	65	40	3200	1300	245
Tractor backhoes B	60	40	5000	400	255
Excavators, crawler A	110	50	2300	1500	230
Excavators, crawler B	100	50	2000	600	240
Excavators, wheel A	100	50	900	1500	230
Excavators, wheel B	90	50	1700	600	240
Combine harvesters	55	65	40000	70	245
Minitractors	10	30	5000	200	280
Railroad machinery	150	30	200	900	250
Agricultural tractors A	55	25	93000	400	245
Agricultural tractors B	45	25	140000	200	255
Tractors, industry	55	30	11000	700	250
Tractors, maintenance	50	25	8000	900	245
Other tractors	45	20	40000	100	265
Forest harvesters	75	70	1700	2000	230
Forest tractors	75	45	2000	1600	230
Haulers	150	30	1000	1400	230
<b>TOTAL</b>			<b>378400</b>		
Compressors	70	60	900	900	250
Towed rollers	35	70	50	100	270
Man-steered rollers	9	70	100	200	300
Vibration plates	6	70	2200	250	300
Generators	35	50	6700	1000	270
<b>TOTAL</b>			<b>9950</b>		
<b><u>MACHINES WITH A GASOLINE ENGINE</u></b>					
Fork-lift trucks	30	30	5000	400	350
Lawn mowers, drivable	10	50	38000	200	350
Agricultural tractors	25	25	20000	50	360
Skidoos, professional	20	30	5000	400	380
Skidoos, other	15	25	65000	20	380
<b>TOTAL</b>			<b>133000</b>		
Vibration plates	3.5	60	4300	200	380
Lawn mowers, non-drivable	3	40	900000	20	420
Chain saws, professional	2.5	60	11000	900	480
Chais saws, other	1.5	30	300000	10	500
Clearing saws	2	40	50000	200	480
Generators	3	60	42000	50	360
Two-wheel tractors	3	50	70000	10	380
Snow blowers	4	30	20000	10	400
<b>TOTAL</b>			<b>1397300</b>		

### Construction machines

All earth-moving machinery accounts for one third of the total fuel consumption. This group includes excavators, tractor backhoes, front-end bucket loaders, bulldozers, rollers, blade graders, and haulers. Excavators are either crawler- or wheel-based, whereas almost all bucket loaders are wheel-based. The total number of construction machines is about 30 000; this figure includes tractors that are put to similar uses. New machines are usually put to very active use, from 1 300 - 1 500 h/y. After about five years, their annual operation time decreases, and the operation time can vary considerably. Some very old machines are still in use. Typical annual operation times for older vehicles are from 300 - 600 h. The average engine power is about 100 kW, excluding tractor backhoes which have engines of 60 to 65 kW. The load factor is 50% of the nominal power for hydraulic operating excavators and from 35 to 45% of the nominal power for loaders. Rear-dump haulers and blade graders are much more effective; their nominal power is 150 kW, the load factor being 30%.

### Industrial machines

Most machines used in industry do loading work. Industrial trucks and tractors account for one eighth of all diesel fuel consumed by work machines. Trucks

can be classified into two categories. One group is used as part of a factory process, and the other group is operated for various kinds of loading tasks, e.g. in warehouses. The annual operating time in the first group averages from 2 000 - 4 000 h, but the second group may operate for only a few hundred hours. About 2 500 trucks are in heavy use; 500 of them are big trucks used in harbors or by the forest industry. Their average engine power is 170 kW, and their lifting capacity is over 12 000 kg. Smaller trucks have an engine power of 50 kW when in active use, and other trucks 30 kW. The load factor for all trucks is 30%. In industry, tractors are used similarly to trucks. Their nominal power is 55 kW, and the load factor is 30%. Tractors used by contractors and in maintenance are also included here.

### Forest machines

Forest tractors and harvesters are in heavy use. Their annual operating time is from 1 500 - 2 200 h. These machines have high load factors, up to 70% for harvesters and 45% for tractors. Both groups have an average engine power of 75 kW. Forest machinery accounts for one tenth of all fuel consumed by work machines. One must also remember that a great deal of wood is transported by normal agricultural tractors, which are not included here.

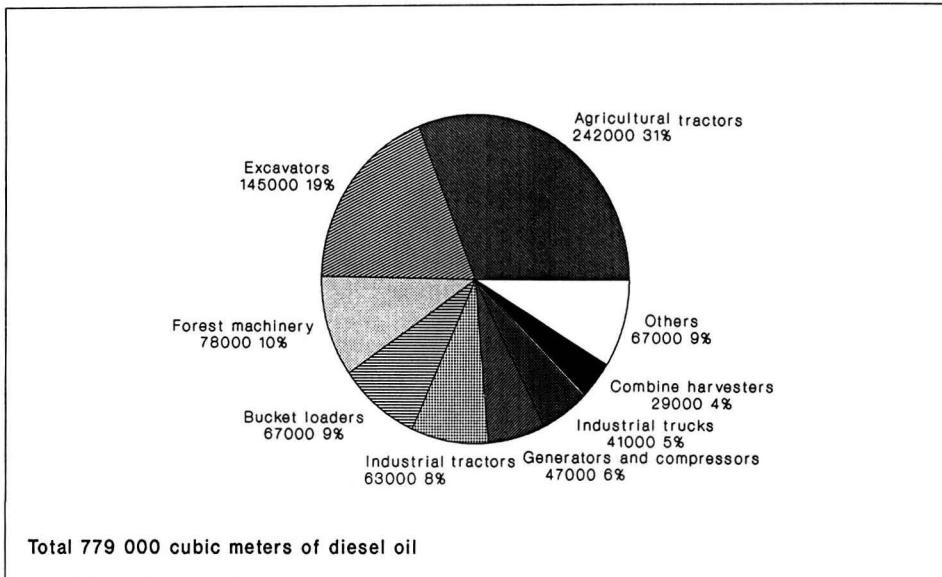


Fig. 3. Total amount of diesel oil consumed by work machinery (m<sup>3</sup>/y) in Finland in 1990.

Table 3. Total energy consumed by work machinery in Finland in 1990 and specific emission factors.

	TOTAL ENERGY (GWh/a)	SPECIFIC EMISSION FACTORS (g/kWh)					
		HC	CO	NOx	PART	SO <sub>2</sub>	CO <sub>2</sub>
<b><u>MACHINES WITH A DIESEL ENGINE</u></b>							
Cranes	26	1.2	4	15	1.0	1.0	770
Other industrial trucks	7	1.2	5	15	1.2	1.0	800
Fork-lift trucks > 12 t	51	1.0	3	14	1.0	1.0	770
Fork-lift trucks < 12 t A	60	1.5	4	15	1.2	1.0	800
Fork-lift trucks < 12 t B	22	1.8	5	15	1.5	1.0	830
Bulldozers	22	1.6	5	15	1.3	1.0	830
Blade graders	62	1.4	4	15	1.3	1.0	800
Rollers	7	1.6	5	15	1.3	1.0	800
Wheel loaders A	129	1.6	4	16	1.0	1.0	770
Wheel loaders B	104	2.0	5	15	1.5	1.0	800
Tractor backhoes A	108	1.2	4	15	1.0	1.0	780
Tractor backhoes B	48	1.8	5	15	1.5	1.0	820
Excavators, crawler A	190	1.6	5	15	1.3	1.0	740
Excavators, crawler B	60	2.0	5	15	1.5	1.0	770
Excavators, wheel A	68	1.6	5	14	1.3	1.0	740
Excavators, wheel B	46	2.0	5	15	1.5	1.0	770
Combine harvesters	100	2.0	5	15	1.7	1.0	780
Minitractors	3	1.6	4	14	1.2	1.0	900
Railroad machinery	8	1.4	4	15	1.2	1.0	800
Agricultural tractors A	512	1.5	4	15	1.5	1.0	780
Agricultural tractors B	315	2.5	5	15	1.7	1.0	820
Tractors, industry	127	1.8	5	15	1.5	1.0	800
Tractors, maintenance	90	1.8	5	15	1.0	1.0	780
Other tractors	36	2.0	5	15	1.5	1.0	850
Forest harvesters	179	1.2	4	15	1.0	1.0	740
Forest tractors	108	1.2	4	15	1.0	1.0	740
Haulers	63	1.2	4	15	1.0	1.0	740
<b>TOTAL</b>	<b>2550</b>						
Compressors	34	1.4	4	12	1.2	1.0	860
Towed rollers	0	1.8	4	12	1.2	1.0	800
Man-steered rollers	0	1.8	5	15	1.5	1.0	960
Vibration plates	2	1.8	5	15	1.5	1.0	960
Generators	117	1.6	4	12	1.0	1.0	860
<b>TOTAL</b>	<b>153</b>						
<b><u>MACHINES WITH A GASOLINE ENGINE</u></b>							
Fork-lift trucks	18	15	150	10	0.5	0.1	1100
Lawn mowers, drivable	38	10	100	10	0.5	0.1	1100
Agricultural tractors	6	15	150	10	0.5	0.1	1100
Skidoos, professional	12	150	200	5	5.0	0.1	1200
Skidoos, other	5	150	200	2	5.0	0.1	1200
<b>TOTAL</b>	<b>79</b>						
Vibration plates	2	15	150	5	0.5	0.2	1200
Lawn mowers, non-drivable	22	15	150	5	0.5	0.2	1300
Chain saws, professional	15	125	200	2	5.0	0.2	1500
Chain saws, other	1	150	200	2	5.0	0.2	1550
Clearing saws	8	150	200	2	5.0	0.2	1500
Generators	4	15	150	5	0.5	0.2	1100
Two-wheel tractors	1	15	150	5	0.5	0.2	1200
Snow blowers	0	15	150	5	0.5	0.2	1200
<b>TOTAL</b>	<b>53</b>						

Table 4. Total emissions from work machinery and total fuel consumption by work machinery in Finland in 1990.

	TOTAL EMISSIONS (1000 kg/a)						TOTAL FUEL CONSUMPTION m <sup>3</sup>
	HC	CO	NOx	PART.	SO <sub>2</sub>	CO <sub>2</sub>	
<b><u>MACHINES WITH A DIESEL ENGINE</u></b>							
Cranes	32	105	395	26	26	20270	7433
Other industrial trucks	9	36	108	9	7	5760	2118
Fork-lift trucks > 12 t	51	153	714	51	51	39270	14400
Fork-lift trucks < 12 t A	90	240	900	72	60	48000	17647
Fork lift trucks < 12 t B	39	108	324	32	22	17928	6607
Bulldozers	34	108	323	28	22	17895	6595
Blade graders	87	250	936	81	62	49920	18353
Rollers	11	34	101	9	7	5400	1985
Wheel loaders A	207	517	2070	129	129	99607	36525
Wheel loaders B	208	520	1559	156	104	83160	30574
Tractor backhoes A	130	433	1622	108	108	84365	31176
Tractor backhoes B	86	240	720	72	48	39360	14400
Excavators, crawler A	304	949	2846	247	190	140415	51344
Excavators, crawler B	120	300	900	90	60	46200	16941
Excavators, wheel A	108	338	945	88	68	49950	18265
Excavators, wheel B	92	230	689	69	46	35343	12960
Combine harvesters	200	501	1502	170	100	78078	28852
Mintractors	5	12	42	4	3	2700	988
Railroad machinery	11	32	122	10	8	6480	2382
Agricultural tractors A	767	2046	7673	767	512	398970	147432
Agricultural tractors B	788	1575	4725	536	315	258300	94500
Tractors, industry	229	635	1906	191	127	101640	37368
Tractors, maintainance	162	450	1350	90	90	70200	25941
Other tractors	72	180	540	54	36	30600	11224
Forest harvesters	214	714	2678	179	179	132090	48300
Forest tractors	130	432	1620	108	108	79920	29224
Haulers	76	252	945	63	63	46620	17047
<b>TOTAL</b>	<b>4260</b>	<b>11388</b>	<b>38253</b>	<b>3437</b>	<b>2550</b>	<b>1988441</b>	<b>730580</b>
Compressors	48	136	408	41	34	29257	10006
Towed rollers	0	0	1	0	0	98	39
Man-steered rollers	0	1	2	0	0	121	44
Vibration plates	4	12	35	3	2	2218	815
Generators	188	469	1407	117	117	100835	37244
<b>TOTAL</b>	<b>240</b>	<b>618</b>	<b>1853</b>	<b>162</b>	<b>154</b>	<b>132529</b>	<b>48149</b>
<b><u>MACHINES WITH A GASOLINE ENGINE</u></b>							
Fork-lift trucks	270	2700	180	9	2	19800	8400
Lawn mowers, drivable	380	3800	380	19	4	41800	17733
Agricultural tractors	94	938	63	3	1	6875	3000
Skidoos, professional	1800	2400	60	60	1	14400	6080
Skidoos, other	731	975	10	24	0	5850	2470
<b>TOTAL</b>	<b>3275</b>	<b>10813</b>	<b>692</b>	<b>116</b>	<b>8</b>	<b>88725</b>	<b>37683</b>
Vibration plates	27	271	9	1	0	2167	915
Lawn mowers, non-drivable	324	3240	108	11	4	28080	12096
Chain saws, professional	1856	2970	30	74	3	22275	9504
Chain saws, other	203	270	3	7	0	2093	900
Clearing saws	1200	1600	16	40	2	12000	5120
Generators	57	567	19	2	1	4158	1814
Two-wheel tractors	16	158	5	1	0	1260	532
Snow blowers	4	36	1	0	0	288	128
<b>TOTAL</b>	<b>3686</b>	<b>9111</b>	<b>191</b>	<b>135</b>	<b>11</b>	<b>72321</b>	<b>31009</b>

*Other machines*

It was found that other machines are not important with regard to exhaust emissions. The number of small gasoline-fueled machines is enormous, about 1.5 million, but their engines are small and their annual operation time is low.

**TOTAL EMISSIONS**

The total amount of diesel fuel consumed by work machines was 780 000 m<sup>3</sup> in 1990 (Fig. 3), which is about 30% of all traffic consumption in Finland, whereas the amount of gasoline consumed, 69 000 m<sup>3</sup>, was only 3% of all traffic consumption. The energy consumption and specific emission factors for each type of work machines are presented in Table 3, their emissions and the fuel consumed in Table 4.

Nitrogen oxide emissions account for 15% of all such emissions in Finland. This is one fourth of all nitrogen oxide emissions of traffic origin. As to particulate matter, carbon monoxide, carbon dioxide, and hydrocarbons, the work machines account for from 4% to 10% of all emissions, respectively. This is still a considerable proportion, especially when compared against road traffic emissions. Sulphur dioxide emissions seem to be less important.

**DISCUSSION**

The total emissions from work machinery were calculated on the basis of data on the number and duration of operation of work machines and specific emission factors. Emissions from off-road diesel machines, in particular, were found to be significant in comparison to those from other sources. The results were similar to those of the Dutch study, whereas work machines are even more important in Sweden because Swedish work machine emissions equalled those of heavy road traffic (Achten 1990; Karlson 1990). One reason might be that the load factor for tractors in Sweden is much larger. It should also be remembered that many more tractors are sold annually in Finland than in Sweden, although the total area of agricultural land use is smaller. It seems, that in Finland tractors have more engine power than would be necessary. This might cause inefficient use of the machines. Modern diesel engines are much better than older ones.

Emissions were evaluated on the basis of one figure only. This method does not take into account differences in operation, e.g., how steady or varying the load is. It is obvious, however, that load variations increase some components of emissions while

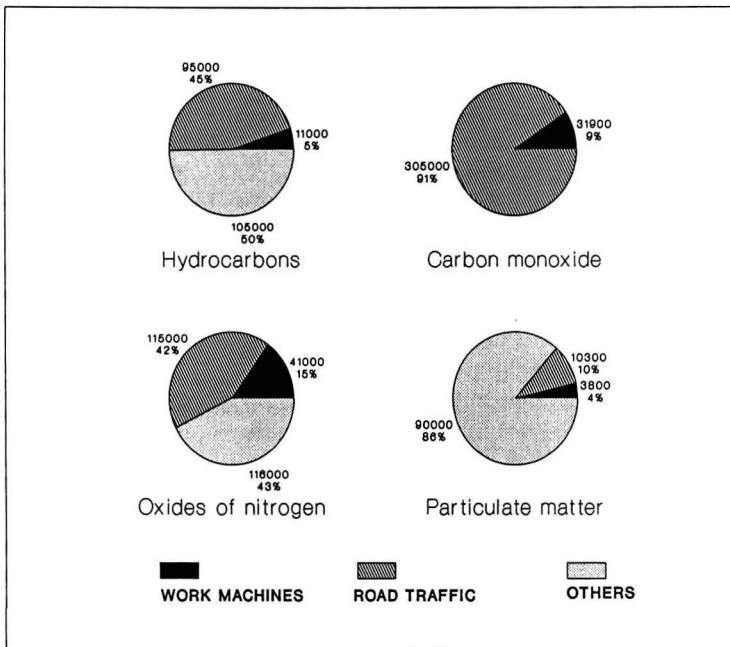


Fig. 4. Emissions from work machinery in Finland (1000 kg/y) in 1990 compared to emissions from other sources.

decrease others. Therefore, it would be important to study the operation of the most important types of work machines in more details, as more precise knowledge would make it possible to define proper strategies for lowering emissions.

Strict standards have been set for road traffic, which means that the importance of off-road machinery will increase (Steeg 1991). Because the work performed by off-road machines is important and difficult to compensate, lowering emissions is not an easy task. Yet, the machine driver can find ways to use a machine that minimizes both emissions and fuel consumption. Another possibility would be to use better fuels, or even alternative fuels. These would have an effect on all machines, whereas engine modifications effect new machines only.

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# **PATTERN OF POLYNUCLEAR AROMATIC HYDROCARBONS IN INDOOR AIR: EXPLORATORY PRINCIPAL COMPONENT ANALYSIS\***

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Principal component analysis (PCA) was used to study polynuclear aromatic hydrocarbon (PAH) profiles in indoor air. Fifteen PAHs were measured in ten different homes in Columbus (Ohio) which had different indoor emission characteristics such as gas utilities, wood-burning fireplaces, and cigarette smokers. Different PAH concentration patterns emerged depending upon the emission sources present in the different homes. Of these, cigarette smoking appeared to have the greatest impact on the indoor PAH concentrations. The PCA allowed convenient displays of the multidimensional data set from which the PAH concentration characteristics could be elucidated. The interrelationship between the different PAHs was also studied by correlation analysis.

## **INTRODUCTION**

Polynuclear aromatic hydrocarbons (PAHs) are formed during combustion processes such as burning of vegetation and fossil fuels, and there is always a background concentration of PAHs in ambient air. In

residences, several combustion sources such as gas cooking and heating, wood-burning fireplaces, and cigarette smokers may be present. Thus, additional PAHs can be generated within a home and indoor air may contain higher concentrations of PAH than the outside air. If several emission sources are present in the same home, a convolution of different PAH patterns is seen at the receptor site. When several PAHs are measured simultaneously, each PAH becomes a variable (or a dimension) and the resulting multi-dimensional data are difficult to interpret. The problem is compounded by the fact that different sources may emit some of the same PAHs. Choosing a par-

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ticular PAH to represent a multitude of others may simplify the problem, but may also lead to erroneous conclusions. To obtain useful information from a multidimensional data set, one has to identify specific patterns of PAHs rather than studying one PAH at a time.

Multivariate statistical techniques can be used to interpret multidimensional data sets. Several such applications to a variety of environmental problems have been reported in the past few years. Techniques such as pattern recognition and hierarchical cluster analysis have been used to study urban aerosols (Hopke et al. 1976; Roscoe et al. 1982). Pattern recognition has also been used to develop a learning machine that can predict undesirable air pollution characteristics (Roeloff et al. 1986). Principal component analysis has been used to study polynuclear aromatic hydrocarbons in Welsh soil (Jones et al. 1989) and also in soil and air using SIMCA principal component analysis (Vogt et al. 1987).

The objective of this study is to interpret a data set of PAH concentrations in indoor air using multivariate techniques such as principal component analysis (PCA). An advantage of PCA is that it allows multi-dimensional data to be projected onto two or three dimensions in a way that much of the information of the original data is retained. So, visual display of the multi-dimensional data is possible using two or three dimensional plots. In this study, PCA was used to graphically represent the different homes and to determine the contributions of different emission sources to the indoor PAH level. The interrelationships among the different PAHs were also studied by correlation analysis.

#### *Principal component analysis*

Principal component analysis is a way of projecting multi-dimensional data onto two or three dimensions while preserving most of the variance in the data set. The original variables are transformed into new sets of variables, or principal components (PCs). The transformation is done in such a way that the first PC contains most of the variance of the data set. The successive PCs contain decreasing amounts of the residual variance. The PCs are formed by linear combinations of the original variables and are orthogonal to each other.

Usually, the first two or three PCs account for a large fraction of the total variance and can be used to represent the original data set. This reduces the dimensionality of the problem with the PCs as the new dimensions. Each original data point (referred to as an object) is represented in two or three dimen-

sions as a function of the PCs and is a point in the new data space. Its coordinates are called PC scores. For  $n$ -dimensional measurements of  $y$  objects, the  $x$ th PC score of the  $y$ th object  $Z_{xy}$  is expressed as follows (Smyers et al. 1984):

$$Z_{xy} = \sum_{j=1}^n a_{xj} V_{yj}$$

where  $a_{xj}$  is the loading of the  $j$ th variable on the  $x$ th component and for the  $y$ th observation, and  $V_{yj}$  is the  $j$ th variable. The loadings  $a_{xj}$  are a measure of the correlation of the corresponding variable along the particular PC, that is, they measure the contribution of the original variable to the PC  $Z_{xy}$ . Mathematical details of PCA are available in the literature and are omitted for brevity.

#### DESCRIPTION OF DATA

Several PAH monitoring studies were carried out by Battelle Columbus (Columbus, Ohio) under contract from the U. S. EPA (Chuang et al. 1986, 1988, 1991; Wilson et al. 1992). The data used in this study were obtained from the study of indoor and outdoor air at eight homes and two background homes in Columbus, Ohio (Chuang et al. 1986; Wilson et al. 1991). Stratification variables in the study were the following combustion sources: cigarette smoking, use of wood-burning fireplaces, and heating and cooking by natural gas or electricity. The sampling was done consecutively in the kitchen (7 am to 3 pm), living room (3 pm to 11 pm), and bedroom (11 pm to 7 am). Simultaneously, a 24-h outdoor sample was taken. The sampling head incorporated a quartz filter to collect particles, which was followed by a polyurethane foam (PUF) adsorbent to collect vapor-phase PAHs. At the end of sampling, the PAHs that were collected on the PUF and the filter were recovered by Soxhlet extraction. The extracts were combined, concentrated, and analyzed by GC/MS. The air exchange rate was determined by injecting SF<sub>6</sub> into the home and measuring its decreasing concentration with time. Details of the sampling and analysis procedures have been published elsewhere (Chuang et al. 1986). The 15 PAHs measured are listed in Table 1 and the average concentrations in different types of homes is also listed.



Table 1. Average concentrations of PAHs in different types of homes and outdoors.

PAH	Home Type A ng/m <sup>3</sup>	Home Type B ng/m <sup>3</sup>	Home Type C ng/m <sup>3</sup>	Home Type D ng/m <sup>3</sup>	Home Type E ng/m <sup>3</sup>	Outdoor Concentrations ng/m <sup>3</sup>
1 Quinoline	4.0	18.2	1.52	12.85	1.85	0.54
2 Isoquinoline	5.97	19.9	1.11	5.62	0.67	0.72
3 Phenanthrene	69.9	71.6	68.32	122.6	40.22	37.37
4 Anthracene	21.79	7.1	3.41	8.11	2.14	2.72
5 Fluoranthene	17.40	18.5	19.17	27.47	10.93	13.44
6 Pyrene	8.55	8.40	8.24	12.97	5.04	7.67
7 Cyclopenta (c, d) pyrene	0.73	1.50	0.21	0.77	0.14	0.68
8 Benzo(a)anthracene	0.87	2.35	0.67	1.66	0.42	1.28
9 Chrysene	1.55	5.5	1.3	3.29	0.70	2.36
10 Benzofluoranthenes	1.73	4.27	1.42	3.15	0.48	3.0
11 Benzo (e) pyrene	1.02	2.06	0.83	1.84	0.23	1.67
12 Benzo (a) pyrene	0.91	2.75	0.80	1.82	0.07	1.38
13 Indeno (1, 2, 3-c, d)pyrene	0.90	2.25	0.66	1.65	0.24	1.27
14 Benzo (g, h, i) perylene	1.90	2.26	1.01	2.23	0.36	2.38
15 Coronene	1.68	1.66	0.60	1.49	0.13	1.44

Home type A - homes with gas utilities

Home type B - homes with gas utilities and smokers

Home type C - homes with gas utilities and fire place

Home type D - homes with gas utilities, smokers and fire place

Home type E - homes with electrical utilities

## DATA ANALYSIS

The data matrix was formed by taking each PAH as a variable. Each variable was standardized so that it had a mean of zero and unit variance. The PCs were computed using the method of eigen-value decomposition of the covariance matrix, and a Varimax rotation was applied (Jolliffe 1986). The rotation maximizes the number of loadings that are either high or near zero and minimizes the loadings that have intermediate values. Thus, the rotation enhances the interpretability and the classification of the data (Hopke et al. 1976).

The PC analysis provides useful information through the variable loadings and the PC score plot. The loading of a variable determines its importance in the PC. A high value (0.5-1.0) implies that the variable is significant for this PC. In the PC score plot, the objects are plotted in the plane of the first two PCs. To study some of the stratification vari-

ables, separate analysis of subsets was carried out. The univariate Pearson correlation coefficients between the PAHs were also calculated and presented. For data analysis, the statistical package Statgraphics (Statgraphics 1986) was used.

## RESULTS AND DISCUSSION

### *Effects of indoor combustion sources*

The results of the PC analysis using the kitchen and living rooms (which are more dependent on the home characteristics than the bedrooms) are presented for all the homes in Table 2 and Fig. 1. The first three PCs account for 82.8% of the variance in the data set and the loading for the different PAHs are presented in Table 2. PC1 is mainly comprised of PAH1 and 7 to 14, PAH 3, 5, and 6 account for PC2 and PAH 2, 4, 13, and 14 make up PC3. Not enough is known about the PAH emission characteristics of the dif-

Table 2. Variable loading for the indoor air data set (kitchen and living room).

PAH	PC1 (61.8%)	PC2 (11.9%)	PC3 (9.1%)
1 Quinoline	0.71	0.21	0.20
2 Isoquinoline	0.34	-0.17	0.68
3 Phenanthrene	0.26	0.90	0.24
4 Anthracene	-0.10	0.13	0.68
5 Fluoranthene	0.31	0.93	0.02
6 Pyrene	0.35	0.91	0.02
7 Cyclopenta(c,d)pyrene	0.82	0.07	0.25
8 Benzo(a)anthracene	0.90	0.27	0.12
9 Chrysene	0.95	0.22	0.04
10 Benzofluoranthenes	0.88	0.37	0.11
11 Benzo(e)pyrene	0.80	0.50	0.24
12 Benzo(a)pyrene	0.93	0.32	0.11
13 Indeno(1,2,3-c,d)pyrene	0.87	0.30	0.28
14 Benzo(g,h,i)perylene	0.61	0.42	0.57
15 Coronene	0.33	0.17	0.72

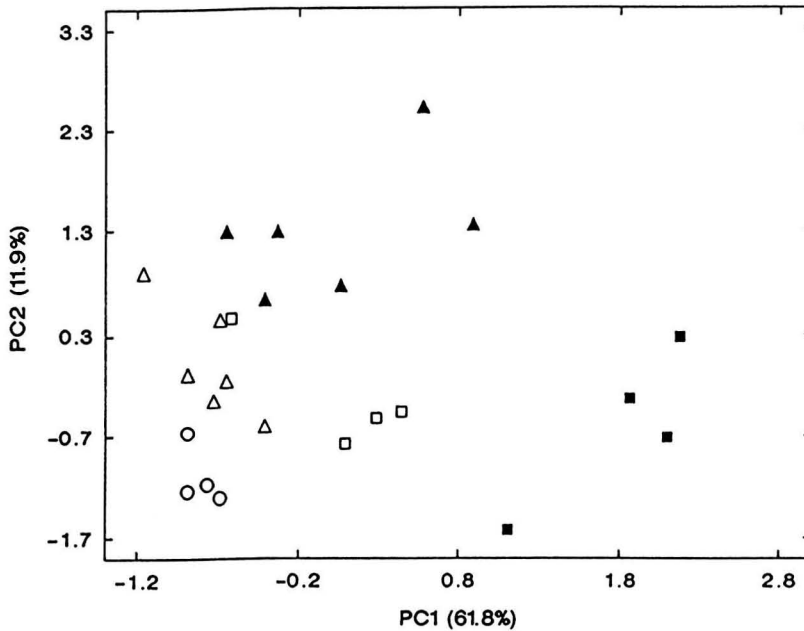


Fig. 1. Principal component analysis of PAHs in indoor air samples (kitchens and living rooms). Principal component score plot: □, homes with gas utilities (Type A); ■, homes with gas utilities and smoking (Type B); △, homes with gas utilities and fire places in use (Type C); ▲, homes with gas utilities, smoking and fireplaces in use (Type D); ○, all electric homes (Type E).

ferent sources present here. So, we could not attribute each PC to a certain source. Except for PAH1, the rest of the compounds exhibiting high loading along PC1 are 4-5 ring compounds and would be found mainly in the particle phase. Some of these compounds are known to be given off by cigarette smoke (Hoffman and Wynder 1986). The PAHs associated with PC2 are mainly found in the vapor phase and can be expected to be given off by cigarette smoke (Hoffman and Wynder 1986) as well as wood smoke (EPA 1989). In PC3, not much can be said about the contributions of PAH 2 and 4, as they can practically come from any source. However, PAH 14 and 15 have been associated with auto exhaust (NRC 1983) as well as tobacco smoke (Hoffman and Wynder 1986).

The correlation analysis complemented the PC loadings. In the correlation matrix presented in Table 3B, we see strong correlations between the concentrations of PAH 3, 5, and 6, evidenced by correlation coefficients close to 0.90. These PAHs were mainly responsible for PC2. Fair correlations also exist among PAH 7 to 14, which correlated strongly along PC1. Benzo[a]pyrene correlated

strongly with PAH 7 to 14 (correlation coefficient close to 0.80). Concentrations of benzo[g,h,i]perylene (PAH 14) and coronene (PAH 15) were well correlated. Because the collection and retention efficiencies of PUF for anthracene are relatively poor (Chuang et al. 1987), losses during sampling and possible losses during evaporative concentration of the sample may account for the poor correlation of anthracene (PAH 4) with PAH 3, 5, and 6.

In Fig. 1, each room (or object) was plotted in the plane of the first two PCs. Here, we see clustering for different types of homes. The all-electric homes (Type E) are characterized by low values on both axes, reflecting low concentrations of all PAHs. Clustering of homes with gas utilities and wood-burning fireplaces (Type C), homes with gas utilities only (Type A), the smokers' homes with gas utilities (Type B), and smokers' homes with gas utilities and fire place (Type D) is also evident. The average concentration of each PAH in different home types is presented in Table 1. The homes in this study were in different neighborhoods and were of different ages and floor plans. The sampling was done on different days under varying ambient temperatures and air

Table 3. Univariate correlation between the PAHs;

(A) (upper right hand): non-smokers' homes only (Type A and C); (B) (lower left hand): all indoor samples (Type A, B, C, D, and E).

PAH	(A)														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1 Quinoline		0.23	-0.13	0.00	-0.21	-0.08	0.64	0.32	0.49	0.38	0.44	0.46	0.58	0.77	0.75
2 Isoquinoline	0.56		0.53	0.95	0.07	0.15	0.36	0.66	0.46	0.21	0.15	-0.10	0.08	0.28	0.29
3 Phenanthrene	0.41	0.10		0.54	0.78	0.78	-0.12	0.27	0.36	-0.02	0.04	-0.34	-0.36	-0.22	-0.27
4 Anthracene	0.01	0.26	0.24		0.06	0.12	0.31	0.61	0.28	0.22	0.17	-0.09	0.10	0.19	0.20
5 Fluoranthene	0.46	0.06	0.90	0.08		0.93	-0.42	-0.09	0.28	-0.07	-0.19	-0.36	-0.48	-0.45	-0.52
6 Pyrene	0.47	0.02	0.90	0.11	0.96		-0.18	0.03	0.40	0.06	-0.03	-0.18	-0.32	-0.22	-0.29
7 Cyclopenta(cd)pyrene	0.48	0.25	0.38	0.23	0.27	0.37		0.40	0.29	0.58	0.61	0.69	0.84	0.94	0.94
8 Benzo(a)anthracene	0.62	0.27	0.54	0.17	0.50	0.56	0.88		0.66	0.62	0.46	0.45	0.34	0.52	0.52
9 Chrysene	0.63	0.25	0.45	0.07	0.48	0.53	0.86	0.97		0.61	0.38	0.23	0.16	0.40	0.39
10 Benzo(a)fluoranthene	0.68	0.30	0.55	0.08	0.63	0.65	0.73	0.91	0.95		0.69	0.77	0.74	0.70	0.67
11 Benzo(e)Pyrene	0.74	0.39	0.68	0.11	0.72	0.74	0.67	0.86	0.85	0.95		0.66	0.65	0.67	0.66
12 Benzo(a)Pyrene	0.70	0.31	0.53	0.06	0.57	0.62	0.81	0.94	0.96	0.97	0.94		0.87	0.82	0.78
13 Indeno (1, 2, 3-c, d) pyrene	0.82	0.49	0.58	0.11	0.56	0.57	0.76	0.88	0.86	0.89	0.92	0.92		0.90	0.88
14 Benzo(g, h, i)perylene	0.61	0.43	0.66	0.22	0.57	0.60	0.66	0.69	0.65	0.74	0.84	0.76	0.83		0.99
15 Coronene	0.26	0.39	0.40	0.22	0.24	0.26	0.50	0.39	0.37	0.44	0.53	0.45	0.48	0.85	

(B)

quality. Considering all these uncertainties, the class separations for the homes with different stratification variables are fairly good. There is little overlap for the different homes, and the general trends seem to be well represented in Fig. 1. When the PC analysis was done by including the bedrooms, results quite similar to those in Fig. 1 were obtained. Thus, they are not presented here.

It is evident from Fig. 1, that both gas utilities and fireplaces affect the PAH concentration in indoor air. Homes A and C show increased scores on both PC axes compared to the all-electric homes E. This implies that there is a net increase of all PAHs associated with PC1 and PC2. In Fig. 1, the gas-heated homes that have fireplaces (C) show relatively lower loadings on PC1 than the ones that do not have fireplaces (A). This lower PAH concentration may be due to higher intrusion of relatively cleaner outdoor air when the fireplaces were in operation. An increase in the concentration of pollutants correlated with PC2 is reasonable, because these more volatile PAHs are generated in greater quantities than are the larger PAHs during combustion of wood (EPA 1989). However, without more detailed air exchange meas-

urements as a function of fireplace operation, a definite statement cannot be made. Similar inferences can also be drawn by looking at the average concentrations of PAH associated with PC1 and PC2 in home types A and C (Table 1).

In Fig. 1, the most dramatic effect is seen for homes with smokers (Type B and D). Smoking resulted in higher values for both PC1 and PC2, that is, it generated all PAH compounds and its effects were more significant than those of the other stratification variables. In Table 1 also we see that the concentrations of PAH associated with PC1 and PC2 are higher for smokers' homes (B and D) than non-smokers' homes (A and C). Other studies have also suggested that cigarette smoke produces a wide range of PAH including the four/five ring ones (Hoffman and Wydner 1986).

Separate PC analysis of the subsets of the smokers' homes (Type B and D) and non-smoker's homes (Type A and C) yielded interesting results. The PC loadings for the smokers and non-smokers are presented in Table 5A and B, respectively. The first three PC account for more than 80% of the variance in both cases. Due to matrix inversion problem, one

Table 4. (A): univariate correlation between the PAHs in smokers' home (Type B and D).

(B): univariate correlation between the mutagenicity of indoor air particle extracts and selected PAHs (7 to 9, 11 to 15).

		(A)														
PAH		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1	Quinoline															
2	Isoquinoline	0.43														
3	Phenanthrene	0.41	0.12													
4	Anthracene	0.05	0.19	0.27												
5	Fluoranthene	0.45	0.10	0.89	0.15											
6	Pyrene	0.47	0.03	0.90	0.18	0.96										
7	Cyclopenta (c, d) pyrene	0.51	0.12	0.27	0.23	0.19	0.29									
8	Benzo(a) anthracene	0.65	0.20	0.53	0.23	0.52	0.58	0.83								
9	Chrysene	0.65	0.19	0.46	0.13	0.51	0.56	0.78	0.96							
10	Benzo(a)fluoranthene	0.71	0.25	0.49	0.13	0.56	0.59	0.75	0.90	0.91						
11	Benzo (e) pyrene	0.74	0.30	0.58	0.14	0.62	0.65	0.71	0.85	0.82	0.95					
12	Benzo (a) pyrene	0.72	0.24	0.44	0.10	0.47	0.53	0.83	0.91	0.90	0.97	0.94				
13	Indeno (1, 2, 3-c, d) pyrene	0.76	0.33	0.41	0.11	0.38	0.42	0.79	0.77	0.72	0.88	0.89	0.91			
14	Benzo (g, h, i) perylene	0.54	0.24	0.34	0.15	0.26	0.32	0.71	0.56	0.48	0.72	0.77	0.76	0.89		
15	Coronene	0.28	0.02	0.16	0.13	0.03	0.10	0.61	0.35	0.27	0.50	0.54	0.54	0.67	0.90	

(B)

Mutagenicity of particulate extract	*	*	*	*	*	*	0.76	0.83	0.87	*	0.55	0.72	0.49	0.31	0.22
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Table 5. Variable loadings from the PC analysis of the subsets;  
(A): smokers' homes only (Type B and D); (B): non-smokers' homes (Type A and C).

PAH	(A)			(B)		
	PC1 (48.79%)	PC2 (24.89%)	PC3 ( 8.99%)	PC1 (43.49%)	PC2 (26.29%)	PC3 (12.29%)
1 Quinoline	0.02	0.82	0.13	0.70	-0.04	0.08
2 Isoquinoline	-0.38	0.27	-0.44	0.11	0.11	0.97
3 Phenanthrene	-0.02	-0.23	0.93	0.18	0.78	0.49
4 Anthracene	0.59	0.04	0.51	0.04	0.07	0.96
5 Fluoranthene	-0.025	0.17	0.97	-0.26	0.94	-0.03
6 Pyrene	0.15	0.16	0.96	-0.05	0.95	0.03
7 Cyclopenta (c, d) Pyrene	0.92	0.27	-0.20	0.80	-0.25	0.30
8 Benzo (a) anthracene	0.86	0.40	0.20	0.52	0.12	0.66
9 Chrysene	0.82	0.39	0.15	0.52	0.54	0.35
10 Benzofluoranthene	0.62	0.71	0.10	0.85	0.16	0.14
11 Benzo (e) pyrene	0.47	0.82	0.20	0.79	0.05	0.10
12 Benzo (a) pyrene	0.72	0.68	0.02	0.90	-0.16	-0.15
13 Indeno (1, 2, 3-c, d) Pyrene	0.44	0.79	-0.23	0.88	-0.33	0.02
14 Benzo (g, h, i) perylene	0.26	0.61	-0.30	0.92	-0.23	0.20

variable had to be rejected. Since PAH 15 and 14 correlated strongly in both subsets, PAH 15 was rejected. In both cases, PAHs 7 to 14 strongly correlated along PC1. However, the contributions to PC2 in the two cases were quite different. In the smoker's homes, PC2 had large loading of PAH 10 to 14 and moderate loading of PAH 8 and 9. Quinoline (PAH 1) which is a marker of tobacco smoke (Wilson et al. 1990) also has high loading along PC2. Most of the compounds that show high loading along this axis are known to be produced by cigarette smoke (Hoffman and Wynder 1986). In the non-smoking homes, PAH 3, 5, and 6 show high loading on PC2 and PAH 9 shows moderate loading. Thus, along this PC axis, one can distinguish between the smoking and non-smoking homes. The object score plots showed trends similar to that of Fig. 1, so they are not presented here.

A comparison of the correlations between the PAHs is interesting. In Table 2A and Table 3A, the correlation matrices for the nonsmokers' homes and the smokers' homes are presented. The three-ring PAH 3, 5, and 6 are well correlated in both cases. Notable are the correlations among PAHs 7 to 15. In

smokers' homes, there is fairly good correlation between PAHs 7 to 15. PAH 12 had a correlation coefficient greater than 0.80 with all PAHs 7 to 13, and PAH 14 and 15 were strongly correlated. For the nonsmokers' homes, the correlation between PAHs 7 to 15 is much smaller. PAH 12 has high correlation with PAH 13 and 14 only. Quinoline (PAH 1), which is a combustion product of tobacco smoke and whose concentration has been shown to parallel that of the tobacco smoke marker nicotine in indoor air (Wilson et al. 1990) is seen to correlate much better with the less volatile PAHs (7 to 15) in the smokers' than in the non-smokers' homes. This absence of correlation among these PAHs in nonsmokers' homes implies that they were produced from a variety of sources, whereas in smokers' homes they came mainly from the cigarette smoke. This indicates that cigarette smoke is a major contributor of PAHs 7 to 15 in indoor air.

#### *Indoor/outdoor comparison*

A two PC analysis of the outdoor and the indoor (kitchen and living room) PAH concentrations resulted in two PCs containing 71.3% of the total

variance, with the first PC accounting for 57.6% of the total variance. The variable loadings are presented in Table 6A. Compounds 7 to 15 show high correlation along PC1, and PAH 1, 3, 5, and 6 have high loadings along PC2 and compounds 2 and 4 have moderate loadings. Thus, PC1 represents the PAHs that are less volatile and contain four or more rings, whereas PC2 represents the PAHs that are more volatile and contain fewer than four rings. The object scores are presented in Fig. 2. The outdoor samples cluster along PC1, indicating that the PAH concentration pattern outdoors differs from that indoors. The indoor air showed much higher values on PC2, indicating higher concentrations of the smaller PAHs.

All of the compounds in this study are semi-volatile, that is, they are distributed between the vapor and the particulate phases in air. Depending on the ambient conditions, for example, temperature and humidity, and the vapor pressures of the individual PAH, their phase distributions and hence dispersion in air may change. The PAHs corresponding to PC1 are found mainly in the particle phase, while those corresponding to PC2 have a significant fraction in

the vapor phase. The vapors may disperse more readily outdoors, leading to lower outdoor concentrations. Adsorption of particles on the surface of walls, carpets and furniture may have resulted in a decrease in the concentrations of particles in indoor air. This would have reduced the concentrations of the larger PAHs that are associated with the particles. As a result in Fig. 2, it can be seen that the outdoor concentrations of the larger PAHs can be higher than that in indoor air. This can also be deduced by looking at the concentration levels in Table 1. Considering all these factors and with the limited data set, it is difficult to predict the effects of outdoor air on the indoor PAH concentrations.

#### *Effect of air exchange rates*

The PC plots can be used as effective display techniques to study other parameters such as the air exchange rate. Air exchange rates are measured as the number of hours needed to replace the air in the house. The air exchange rate corresponding to each object (or room) was plotted on a PC plot. Though the plot is not shown here, each type of home main-

Table 6. (A): variable loading from the PC analysis of the combined indoor (kitchen and living room) and outdoor data.

(B): variable loadings for the analysis of mutagenicity of indoor air particles.

PAH	(A)		(B)	
	PC1 (57.69%)	PC2 (14.19%)	PC1 (77.79%)	PC2 (14.89%)
1 Quinoline	0.28	0.69	---	---
2 Isoquinoline	0.12	0.45	---	---
3 Phenanthrene	0.12	0.92	---	---
4 Anthracene	0.0	0.38	---	---
5 Fluoranthene	0.24	0.88	---	---
6 Pyrene	0.47	0.72	---	---
7 Cyclopenta (c, d) Pyrene	0.79	0.11	0.74	0.50
8 Benzo (a) anthracene	0.90	0.31	0.96	0.21
9 Chrysene	0.82	0.35	0.98	0.11
10 Benzofluoranthenes	0.92	0.23	---	---
11 Benzo (e) pyrene	0.91	0.27	0.80	0.48
12 Benzo (a) pyrene	0.91	0.33	0.87	0.46
13 Indeno (1, 2, 3-c, d) Pyrene	0.89	0.35	0.68	0.67
14 Benzo(g, h, i) perylene	0.90	0.07	0.39	0.91
15 Coronene	0.77	0.02	0.13	0.90

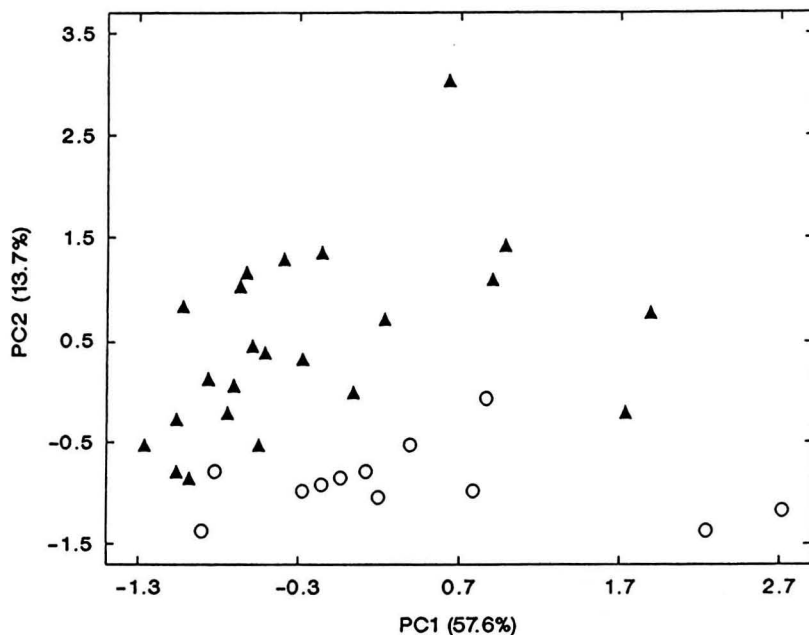


Fig. 2. PAHs in indoor and outdoor air samples. Principal component score plot; ▲, indoor air samples; ○, outdoor air samples.

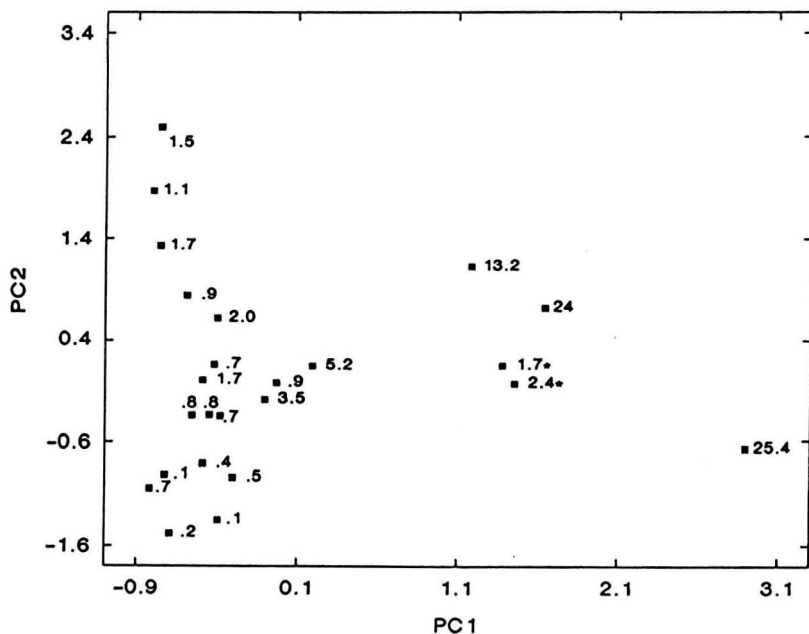


Fig. 3. Principal component analysis of indoor air samples including only those PAHs that are known to be carcinogenic and/or mutagenic; PC score plot. Mutation frequency in revertants  $\times 10^4$  per  $m^3$  of air are given beside the symbols.

tained its PAH concentration pattern despite varying air exchange rates. The dispersion of the PAHs may be expected to depend upon such factors as diffusion coefficients, vapor pressure, humidity, particle characteristics, and surface adsorption or desorption; thus, different PAHs would disperse at different rates. Clearly, the dispersion of pollutants is more complex than just simple air exchange as measured by dispersion of SF<sub>6</sub>, a highly volatile tracer. This would explain why we did not see any distinct effect of air exchange rate on the PAH concentration in the PC plots.

#### *Mutagenicity of indoor air particles*

An attempt was made to see if the mutagenicity of the particles collected in this study correlated with the PAH concentrations and with PCs. Only the PAHs known to be carcinogenic and/or mutagenic were used for this analysis. The PAHs included in the list were 7 to 9 and 11 to 15. It was assumed that these PAHs were mainly in the particle phase and were thus collected on the filter. Mutagenicity of the particle fraction of these samples was studied at the same time and have been published elsewhere (Lewtas et al. 1987). The first PC accounted for 77.7% of the variance and the second for another 14.8%. The variable loadings are shown in Table 6B. The PAHs 7 to 9 and 11 to 13 are mainly responsible for PC1 whereas PAH 14 and 15 are mainly responsible for PC2. The mutation frequency values expressed as revertants per cubic meter of air are presented on the PC score plot of Fig. 3. The univariate Pearson correlation coefficients were also calculated and are presented in Table 4B. High correlation (correlation coefficient 0.70) between mutation frequency and PAH 7 to 9 and 12 was seen. Though it is difficult to predict any trend based on Fig. 3 alone, one can say that the increase in mutagenicity parallel increases in the PC1 score. The rooms with asterisks are exceptions and we are unable to predict the reason behind this. If least-square regression lines between mutation frequency and PAHs 8, 9, and 11 are drawn, the same rooms appear as outliers. It should be noted that these PAHs themselves account for only a small fraction of the total mutagenicity (Lewtas et al. 1987), but may be correlated with other species given off from the same sources that account for the residual mutagenicity. PAH 14 and 15, which account for PC2, seem to contribute less towards the mutagenicity.

## CONCLUSIONS

Principal component analysis was used to display and analyze the concentration patterns in indoor air. It was found that gas utilities, wood burning, as well as cigarette smoke contribute to the PAH level in indoor air. In the PC score plots, the homes with similar emission characteristics were seen to cluster together. Cigarette smoke was found to be the largest contributor of PAH 7 to 15 in indoor air. An active fire place seemed to reduce the concentrations of larger PAHs as it generated a flow of air from outside, but it increased the concentrations of three-ring compounds as they were produced by wood smoke. On the PC score plot, the outdoor PAH concentration pattern seemed to be very different from that of the indoor concentration pattern. The concentration of smaller, volatile PAHs was higher in indoor than in outdoor air. An attempt was also made to correlate the mutagenicity of indoor air particles with the PAH concentration. Muta-genicity correlated strongly with chrysene and on the PC score plot, it was seen to increase along PC1. Although source emission studies have suggested some of these relationships before, use of principal component analysis in a receptor modeling approach to interpretation of indoor air pollution problem has not previously been done, and confirms the inferences of the source studies.

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## **PUBLIC AWARENESS AND ATTITUDES TOWARD VARIOUS USES OF RENOVATED WATER**

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Public awareness and knowledge in the State of Bahrain regarding various aspects of wastewater were assessed along with the public's attitude toward various specific uses of reclaimed water during different conditions. Data were obtained by a survey of 500 respondents. The results indicated that a large percentage of the individuals surveyed were not aware of the basic and simple aspects concerning wastewater. Most respondents were strongly opposed to using reclaimed water regardless of the conditions and were willing to pay more to avoid using it. However, the opposition decreased as one moves from ingestion to bodily contact and finally non-bodily contact. The reasons for the opposition were mainly health risks, psychological repugnance, and religion. Knowledge and attitudes of the respondents were found mainly to be dependent on the level of education and age. Comparing the present finding with previous works, it can be concluded that people in many countries in the world regardless of their differences, have similar attitudes toward the various uses of reclaimed water.

### **INTRODUCTION**

During the last decades, the Arabian Gulf States have expanded tremendously due to the implementation of large-scale developmental plans. This was demonstrated in a steady growth in population and rapid expansion of commercial, industrial, and agricultural activities. These continuous activities posed a significant strain and an ever-increasing demand for the basic facilities, such as water supply. Bahrain, like other Gulf countries, has no surface water resources, and groundwater is the only source of fresh water. Until 1975, all water supply was from groundwater, but due to continually increasing demand and exploitation, combined with degradation of its quality (Madany and Akhter 1990; Raveendran and

Madany 1991), it has become necessary to turn to alternative sources, such as desalination. Other sources, such as treated wastewater, are well recognized worldwide to constitute an increasingly important element of the total available water resources. This resource has been considered in Bahrain, as well as other Gulf States, to be a potential mean of supplementing available supplies (Akhter and Madany 1991). In Bahrain and other Gulf States, all research activities have been directed towards the health and environmental implications of reusing treated wastewater (Abdulraheem 1989; Al-sheryani 1992), but virtually no attention has been given to public opinion, awareness, and attitudes toward the use of reclaimed wastewater. Most of the studies regarding

public perceptions and attitudes have been carried out in developed countries. For example, Athanasiou and Hanke (1970) conducted a random telephone survey of 291 households in Baltimore, Maryland, to test attitudes towards reused wastewater for both potable water supply and recreational uses. Johnson (1971) surveyed 221 persons in five U.S. cities and found that 77% of the interviewees would be willing to drink renovated wastewater. Bruvold and Ward (1972) analyzed public opinion in ten communities in California, and concluded that public acceptance of reclaimed wastewater is associated with the particular use. Gallup (1973) performed a nation-wide survey on attitudes toward water supply and recycled sewage, and found that 54% of the respondents were opposed to drinking recycled waste water. Olson and Pratte (1978) conducted a survey in Irvine, California, to determine attitudes toward expanded uses of reclaimed water, and obtained that respondents were in favor of using such water to supplement existing water supply as long as the uses were not in the home. Bruvold (1979) conducted a study dealing with public assessment of wastewater reclamation and reuse options in ten California cities. The study found that about 600 of the respondents were opposed to direct reuse. Another study interviewed 140 Irvine residents to assess their knowledge and evaluation regarding the use of reclaimed wastewater in their community, and similar conclusions were obtained (Bruvold 1981). A study, carried out in five towns in Puerto Rico (Calas and Rios 1981) on consumer attitudes toward reuse of wastewater, obtained that 28% of the respondents were willing to consume reclaimed water. Although results from one study to another varied some, overall, about 50-60% of the individuals surveyed opposed direct reuse of reclaimed water, and about 25-40% were in favor of direct reuse. The present paper reports results from a study conducted throughout Bahrain, where reclaimed water is used only for irrigation purposes, and has the following aims:

- (1) to assess the awareness and knowledge of individuals regarding wastewater,
- (2) to measure attitudes toward various specific uses of reclaimed water at different situations,
- (3) to investigate personal and socio-demographic variables associated with attitudes toward the use of reclaimed water, and
- (4) to compare the reaction of Bahraini individuals toward wastewater reuse with individuals in other countries.

## METHODS

A total of 500 Bahraini individuals from all areas in Bahrain were interviewed in 1991 by a trained graduate student. Samples were taken from each area in Bahrain and the number selected was based on the population of that area. A pilot questionnaire was designed and used on trial basis. Based on the results of this trial, minor wording and formal changes were made to the questionnaire, and the final version was constructed.

The questionnaire contained three distinct sections. The first which contained eight items, dealt with awareness and knowledge of the respondents about wastewater. The second section consisted of five items and was concerned with the attitudes of the respondents toward the use of treated wastewater at different conditions. The final section of the questionnaire asked for some socio-demographic information, namely age, sex, education, geographic area, and job title (income). The statistical test used in the data analysis was the  $\chi^2$ -test for independence.

## RESULTS

Table 1 presents the demographic characteristics of the study participants. The sample was selected in such a way to be similar in characteristics to the population of Bahrain.

Table 2 summarizes the results for section one of the interview schedule on awareness and knowledge of the individuals regarding wastewater. The data in the table show that only 77.8% of the respondents were aware of the fate of wastewater generated in their houses, and 22.2% gave either wrong answers or said they do not know. A  $\chi^2$ -analysis of the data by sex, age, education, and income, revealed that knowledge of the respondents is dependent only on the level of education ( $\chi^2 = 34.00$ ,  $P < 0.0001$ ) and age ( $\chi^2 = 23.2$ ,  $P < 0.003$ ). Approximately 73.2% of the individuals surveyed in Bahrain were aware of the importance of treating wastewater, whereas a relatively large percentage (26.8%) answered that there is no need for treatment or said 'do not know' (Table 2). The awareness for the need for treatment was significantly related to the education level ( $\chi^2 = 57.2$ ,  $P < 0.0001$ ) and age ( $\chi^2 = 45.6$ ,  $P < 0.0001$ ). Also, only 65.4% of the respondents knew that there is a treatment facility in Bahrain, and this correlates well with the previous question about the need to treat wastewater. The statistical analysis indicated that males are more aware than females of the existence of a treatment plant ( $\chi^2 = 34.4$ ,  $P < 0.0001$ ), and this awareness is also dependent on

Table 1. Characteristics of study subjects.

Characteristics	Study Subjects (N=500)	
	N	%
<b>Sex</b>		
Male	244	48.8
Female	256	51.2
<b>Age, years</b>		
20-30	198	39.6
31-40	122	24.4
41-50	36	7.2
51-60	40	8.0
>61	104	20.8
<b>Education</b>		
< 6years schooling	163	32.6
11+, non-university	230	46.0
University	107	21.4
<b>Income</b>		
Low	109	21.8
Medium	378	75.6
High	13	2.6

age and level of education ( $\chi^2 = 39.3$ ,  $P < 0.0001$ ). Of those who knew about the treatment plant, only 38% gave a correct answer on its locality. Here also, males were more knowledgeable ( $\chi^2 = 34.4$ ,  $P < 0.0001$ ), and the level of education ( $\chi^2 = 37.4$ ,  $P < 0.0001$ ), was related to the knowledge of the respondents on the locality of the treatment plant.

Knowledge of the wastewater treatment method was poor, as expected, and was dependent on age ( $\chi^2 = 20.2$ ,  $P < 0.009$ ), and education ( $\chi^2 = 20.8$ ,  $P < 0.0003$ ). When the respondents were asked about the possibility of utilizing wastewater, 81.2% responded correctly, and the  $\chi^2$ -analysis showed that the correct answer depends on education level ( $\chi^2 =$

44.0,  $P < 0.0001$ ), age ( $\chi^2 = 30.0$ ,  $P < 0.0001$ ), and income ( $\chi^2 = 12.3$ ,  $P < 0.002$ ). However, only 60.4% were aware of the wastewater reuse program in Bahrain, and 65.4% were aware of reuse in other countries. The statistical analysis showed that awareness of water reuse is dependent on various levels of education, and on age, sex, and income. The final item in section one is concerned with the awareness regarding the importance of wastewater treatment in mitigating coastal pollution. The response rate was about 79% positive, 5% negative, and 15.6% 'do not know', and is related to age ( $\chi^2 = 29.1$ ,  $P < 0.0003$ ) and education ( $\chi^2 = 18.1$ ,  $P < 0.0001$ ).

Table 2. Knowledge and awareness regarding waste water.

Question	Yes	No	Don't know
	(% )		
fate of wastewater generated in the house	77.8	22.2	
need to treat wastewater	73.2	14.5	12.3
existence of a wastewater treatment plant in Bahrain	65.4	8.0	26.6
wastewater treatment method	28.8		71.2
possibility of utilizing wastewater	81.2	18.8	
wastewater reuse projects in Bahrain	60.4	5.0	34.6
Wastewater reuse projects in other countries	65.4	2.4	32.1
treatment of wastewater leads to reduction of coastal pollution	79.4	5.0	15.6

The second section of the questionnaire dealt with attitudes toward the use of treated wastewater at various conditions. Generally, the use of treated wastewater was opposed by 68% of the respondents, and was associated with age ( $\chi^2 = 27.4$ ,  $P < 0.0001$ ), education ( $\chi^2 = 23.1$ ,  $P < 0.0001$ ), income ( $\chi^2 = 14.4$ ,  $P < 0.0007$ ), and sex ( $\chi^2 = 5.86$ ,  $P < 0.01$ ). The most frequent reason mentioned for the stated opposition was health risks (63%), followed by psychological repugnance (27.2%), and religion (9.2%), as shown in Table 3. About 60% of the respondents showed their willingness to pay more for the water supply in order to avoid using treated water. Surprisingly, this willingness was not dependent on income, but it was related to age ( $\chi^2 = 13.6$ ,  $P < 0.008$ ) and sex ( $\chi^2 = 6.5$ ,  $P < 0.01$ ), where females were willing to pay more.

Table 4 summarizes the results on attitudes of respondents toward various specific uses of reclaimed

water at different conditions. The table shows that 96.4% and 94.2% of the individuals surveyed were opposed to using reclaimed water under normal conditions for drinking and cooking purposes, respectively, whereas during drought conditions this opposition decreased, although only slightly. It seems that the respondents were not influenced by the economic factor in their opposition to use reclaimed water for drinking and cooking. So, even if this water was cheaper than conventional water, 95.4% and 92.3% of the respondents were opposed to using it. The opposition of the respondents decreased as the proposed use of reclaimed water was not associated closely with personal contact. For example, under normal conditions, 79.9% and 92.9% of the respondents were in favor of using reclaimed water for industrial operations and irrigation of parks, respectively.

Table 3. Reasons for opposition to uses of treated wastewater.

Reason	N	%
Health risks	220	63.0
Psychologically repugnant	95	27.2
Religious	32	9.2
Environmental	2	0.6

Table 4. Attitudes toward various specific uses of reclaimed wastewater at different conditions.

	Normal Conditions		Drought conditions		Treated wastewater costs less than conventional water	
	Yes (%)	No	Yes (%)	No	Yes (%)	No
Drinking	3.6	96.4	15.5	84.5	4.6	95.4
Cooking	5.8	94.2	19.1	80.9	18.7	92.3
Bathing in home	10.6	89.4	30.7	69.3	19.3	80.7
Swimming pools	20.5	79.5	47.8	52.1	42.6	57.3
Recharging groundwater	29.1	70.9	49.0	50.9	44.0	55.9
Irrigation of vegetables	62.3	37.7	74.6	25.4	71.2	28.8
Industry	79.9	20.1	87.3	12.7	84.3	15.7
Irrigation of Parks	92.9	7.1	96.3	3.7	94.3	5.7

## DISCUSSION

The present investigation represents the first study conducted in the Gulf States regarding public knowledge and awareness about wastewater, and the public's attitude toward the various uses of reclaimed water under different conditions. There is generally a lack of information about the public's

knowledge and attitude toward many issues, because usually public opinion is not studied and not taken into consideration during planning and implementation of most projects. Thus, this research is considered a novel study in the Gulf and perhaps in the Arab region.

The results of the first part of this study on the knowledge and awareness of citizens concerning wastewater revealed generally that their knowledge and awareness is surprisingly low. Relatively high percentages of the public were not able to respond correctly to basic information about wastewater, and were not aware of simple aspects regarding wastewater. For example, 22.2% of the respondents did not know what happens to the wastewater generated in their houses, and 28.8% were not aware of the paramount importance of treating wastewater (Table 2). The above responses are consistent with their unawareness of the existence of wastewater treatment facilities in Bahrain. Similarly, more than 18% of the individuals surveyed were not knowledgeable about the possibility of utilizing wastewater for beneficial applications, and, thus, it was expected to find that 71.2% of the respondents did not have knowledge of wastewater treatment methods. Comparing this finding with that of Bruvold (1981) on the knowledge of Irvine, California citizens about wastewater treatment methods (only 40.7% did not have knowledge), the degree of unawareness of the Bahraini citizens can be seen.

Regarding the knowledge of the Bahraini individuals surveyed about wastewater reuse projects in Bahrain and other countries, 60.4% and 65.4%, respectively, were aware of such projects, whereas 58.2% and only 14.9% of the citizens of Irvine, California were aware of such project in their city and nearby communities, respectively (Olson and Pratte 1978).

It is apparent from statistical analysis that knowledge and awareness of the Bahraini citizens concerning various aspects of wastewater is strongly dependent on age and level of education. Those who were more knowledgeable were in the age group ranging from 20 to 40 years, and this group consists mostly of university graduates and, thus, is better educated.

The results indicate that the public's total knowledge and awareness of various aspects of wastewater is generally low. This strongly suggests that the public is not well informed on wastewater, which shows that there is a need to create an information/education program, and that more effort needs to be put in promoting public awareness.

In Bahrain, 68% of the citizens were opposed to the reuse of renovated water in general, and about 60% of them, regardless of their income, were willing to pay more for water to avoid using reclaimed water. Females were willing to pay more because women, the principal users of water in the home in the Gulf States, are more critical of their household's

water than men. Reasons for opposition were health risks (63%), psychological repugnance (27.2%), religion (9.2%), and environmental adverse effects (0.6%) (Table 3). This is the only study that includes the religious factor as one of the reasons for opposition to reused wastewater. Some Muslims believe wrongly that this water can not be used by them because of its origin.

The above findings are reasonably consistent with the literature. Calas and Rios (1981) found that 72% of the respondents in Puerto Rico opposed consuming reclaimed water, mainly because it is unhealthy (67%), unpleasant (63%), and vulgar (53%). Sixty percent of the citizens of California, U.S.A. opposed the direct reuse of reclaimed water (Bruvold 1979). Bruvold and Ward (1972) reported that psychological repugnance (29.2%), concern over purity (21.5%), and health risks (9.8%) were most frequently mentioned as reasons for the stated opposition to using reclaimed water. Thus, it might be generalized that psychological repugnance and health risks are the two main reasons behind the rejection of using renovated water.

The degree of opposition of the Bahraini citizens, as well as other people in the world to the use of renovated water is associated directly to the proposed use. The data of the present investigation show that the opposition decreases sharply when the suggested application moves away from bodily contact (Table 4). For example, during normal conditions, 96.4% and 94.2% of the respondents were opposed to using reclaimed water for drinking and cooking, respectively, whereas this opposition decreased to 20.1% and 7.1% for the use in industry and irrigation of parks, respectively. These results are consistent with the previous findings on the public's attitude toward various uses of reclaimed water (Baumann and Kasperon 1974; Bruvold and Ongert 1974; Bruvold and Ward 1972; Olson and Pratte 1978; Bruvold 1981). Thus, it can be concluded that people's attitudes toward the different uses of reclaimed water are similar regardless of their cultural and social differences. During drought conditions, respondents' opposition towards the various uses of reclaimed water decreased, although only slightly. This suggests their strong opposition to certain uses, regardless of the condition. This conclusion is further confirmed by the fact that the respondents' attitudes did not significantly change, when the reclaimed water was less expensive than conventional water.



## CONCLUSIONS

In light of the preceding results and discussion, the following is concluded:

(1) Knowledge and awareness of the citizens of Bahrain regarding various aspects of wastewater is very limited. Relatively high percentages of the public are not aware of the basic and simple aspects concerning wastewater. This awareness is dependent mainly on age and level of education.

(2) Most of the citizens oppose using renovated water and are willing to pay more to avoid using it. Females are more than males opposed to using renovated water and are willing to pay more.

(3) There is a clear step-by-step progression of opposition to the use of renovated water as one moves through the hierarchy of uses from non-bodily contact such as in industry and park irrigation, to bodily contact such as swimming and home bathing, and finally to ingestion such as in cooking and drinking.

(4) The reasons for the opposition to using renovated water are mainly health risks, psychological repugnance, and religion.

(5) The citizens of Bahrain oppose strongly certain uses of renovated water, regardless of the condition.

(6) Comparing the results of the present investigation with previous works, it can be concluded that Man's attitudes worldwide toward the various specific uses of reclaimed water are similar.

(7) The data strongly suggest that there is an immediate need in promoting public awareness and knowledge regarding wastewater.

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## LEAD CONTAMINATION IN SURFICIAL SEDIMENTS FROM NEWARK BAY, NEW JERSEY

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Concentrations of lead were determined in surficial sediments collected from 18 locations throughout the Newark Bay, New Jersey estuary. Measured concentrations ranged from 64 mg/kg to 2.5 g/kg (dry weight). In addition, a more intensive sampling effort was conducted in two wetland areas along the Hackensack River. In these areas, concentrations ranged from 25 to 704 mg/kg. The results of these sampling efforts were compared to data from previous studies to characterize temporal trends of lead contamination. Possible sources of lead were identified based on the observed distributions. The potential for adverse effects to fish and shellfish was evaluated based on comparisons to available benchmark sediment quality criteria and toxicity guidelines. The results of this study were found to be similar to previous investigations. Concentrations of lead measured in all reaches of the estuary were found to exceed sediment quality criteria (>250 mg/kg) and predicted toxic effects values (>110 mg/kg). The highest lead concentrations in the estuary were located adjacent to petroleum refineries, paint and pigment formulating plants, and other industrial areas. These results indicate that lead contamination of surficial sediments in Newark Bay may pose a significant threat to aquatic biota.

### INTRODUCTION

The presence of lead in estuarine environments is of importance due to its potential toxicity to aquatic biota and waterbirds. This is particularly relevant in estuaries located within heavily industrialized and populated areas, where significant anthropogenic contributions typically include point and nonpoint sources such as industrial discharges, emissions from automobiles, dustfall, precipitation, combined sewer outfalls (CSO), municipal wastewater and treatment plant effluents, and stormwater runoff (Mytelka et al. 1973; Whipple et al. 1976; Wilber and Hunter 1979a,b; Meyerson et al. 1981; NOAA 1982; Granier et al. 1990; USEPA 1992). Lead is typically reported at concentrations above background levels in aquatic environments proximate these sources (Luoma and Bryan 1978; Eisler 1988).

The adverse effects of heavy metals, including lead, on aquatic ecosystems have been reviewed by several researchers (Greig and McGrath 1977;

Waldhauer et al. 1978; McGreer 1979; Meyerson et al. 1981; NOAA 1984; Malueg et al. 1984; Tietjan and Lee 1984; Haux et al. 1981 1986). Lead frequently is identified as a chemical of concern in aquatic ecosystems due to its demonstrated toxicity to a wide variety of aquatic species (NOAA 1984; 1991a). Lead is readily absorbed by waterbirds, fish, and invertebrates (Waldhauer et al. 1978; DiGiulio and Scanlon 1985; Haux et al. 1986). Exposure to elevated concentrations of lead in contaminated sediments and water has been demonstrated to impair growth and reproduction in aquatic plants and biota, and to effect physiological processes such as respiration, osmoregulation, and metabolism (Leland and Kuwabara 1985; Haux et al. 1986; Eisler 1988; NOAA 1991a). Physiological indicators of chronic lead exposure in fish include spinal curvature, darkening of the dorsal tail region, muscular atrophy, and degradation of the caudal fin (Eisler 1988). Neurological impairments, kidney dysfunction, and anemia have also been ob-

served in aquatic vertebrates as a result of exposure to high concentrations of lead (Leland and Kuwabara 1985).

Newark Bay, New Jersey is an enclosed estuary located in the heavily industrialized and densely populated New York City, New York/Newark, New Jersey metropolitan area. Port Newark and Port Elizabeth, located along the western bank, are two of the largest shipping ports in the world (Meyerson et al. 1981). Newark Bay is included within the New York/New Jersey Harbor Estuary, which has been cited as one of the most chemically polluted estuaries in the United States (NOAA 1984; NOAA 1988). The concentrations of lead and other heavy metals in sediments contribute to the estuary's ranking among the top twenty contaminated coastal sites in the country (NOAA 1988). Numerous studies examining heavy metal contamination in Newark Bay have been conducted (Greig and McGrath 1977; Suszkowski 1978; Waldhauer et al. 1978; Williams et al. 1978; Ellis et al. 1980; USEPA 1981; Meyerson et al. 1981; USEPA 1982; Luther et al. 1987; Ropes 1987; Goeller 1989; HMDC 1989; NOAA 1989, 1991b). However, few studies have been published regarding the impact of this contamination on aquatic species.

As part of a broad investigation of the distribution, flux, and biological effects of chemical pollutants in Newark Bay, the concentrations of lead were determined in surficial sediments collected from locations throughout the estuary. The objectives of this evaluation were to: (a) determine temporal changes in lead levels reported in past studies; and (b) evaluate the potential hazard to aquatic biota. Lead concentrations measured in surficial sediments were compared to results reported in previous investigations. The potential for adverse effects to biota was evaluated through comparisons to available sediment quality criteria and guidelines. In the absence of sediment quality criteria specific to New Jersey waterways, sampling results were compared to values derived from several proposed approaches. A threshold value based on the Equilibrium Partitioning approach (EqP) has been proposed by the United States Environmental Protection Agency (USEPA 1985) and was used here for comparative purposes. Ontario Ministry of the Environment (OME) Draft Sediment Quality Guidelines developed using the Screening Level Concentration (SLC) approach (Persaud et al. 1991) and Washington State Marine Sediment Quality Standards derived for Puget Sound using a combination of the Apparent Effects Threshold (AET) and the EqP methodologies (WADOE 1991) were also

used for comparative purposes. In addition, preliminary values developed by the California State Water Resources Control Board (CSWRCB 1990) based on the AET approach were considered. Data also were compared to median- and lowest-observed sediment toxic effect levels derived by the National Oceanic and Atmospheric Administration (NOAA) from a compilation of the results of acute and chronic bioassays in several aquatic species (NOAA 1991a). Although many of these values are considered site-specific and are not intended for use as regulatory guidelines, they provide useful benchmarks for assessing the potential toxicity of contaminated sediments.

## METHODS

Eighteen surficial sediment samples were collected throughout Newark Bay in February, 1990 at locations where deposition and accumulation of sediment materials were visually evident (Fig. 1). Sampling locations included eleven sites along the Passaic River and two samples from the Hackensack River. Five samples were collected from areas within the Arthur Kill, the Elizabeth River, and the Rahway River. Sediments at each location were collected with a petite Ponar Grab sampler. Samples were taken from the center of each grab sample using an acid-cleaned stainless steel scoop and transferred to pre-cleaned, amber glass containers. Samples were stored at 4°C until laboratory analysis. Lead analyses were conducted by Twin City Testing Corporation (St. Paul, Minnesota) according to the U.S. Environmental Protection Agency (USEPA) Method 6010 (USEPA 1986). The detection limit for the analysis was 5 mg/kg (dry weight basis).

During October and November of 1991, 27 additional sediment samples were collected from two wetlands located along the western bank of the Hackensack River. Specifically, 19 samples were collected from the Hackensack Meadowlands (Area I), located near the western spur of the New Jersey Turnpike and bounded by the Belleville Turnpike and the AmTrak railway (Fig. 2). An additional eight samples were collected from a marsh area (Area II) located east of the New Jersey Turnpike Western spur near Kingsland Creek, where direct industrial inputs were limited (Fig. 3). Sediments were collected at each location using a Lamont dredge. Samples were transferred to pre-cleaned, teflon-lined glass containers and stored at 4°C until laboratory analysis. Lead analyses were conducted by Accutest (Dayton, New Jersey) using USEPA Method 6010 at a detection limit of 5 mg/kg (dry weight basis).

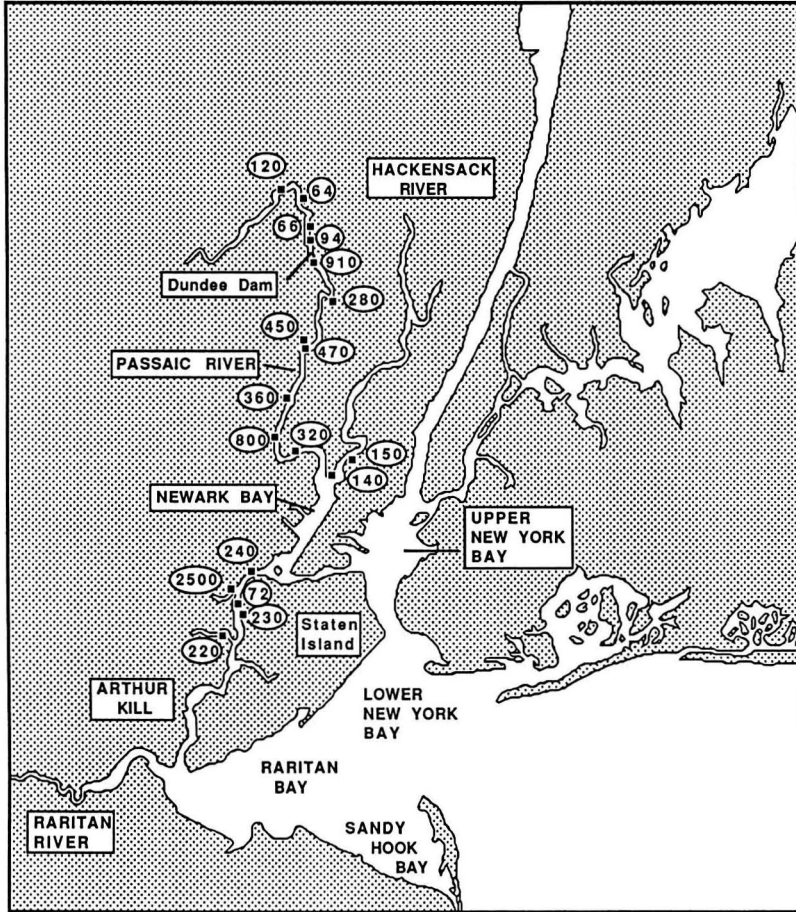


Fig. 1. Lead (Pb) concentrations (mg/kg dry wt.) measured in surficial sediment from various sections of Newark Bay, New Jersey.

## RESULTS

The concentrations of lead measured in the 18 surficial sediment samples collected in February, 1990 are presented in Fig. 1. Lead concentrations ranged from 64 mg/kg to 2.5 g/kg. The highest concentration was found at the confluence of Morses Creek and the Arthur Kill. Lead concentrations in sediments from the Arthur Kill ranged from 72 mg/kg to 2.5 g/kg (median concentration of 230 mg/kg). Sediment concentrations in the Passaic River ranged from 280 to 910 mg/kg (median concentration of 450 mg/kg) below the Dundee Dam and 64 to 120 mg/kg (median of 68 mg/kg) above the dam. The highest lead concentration in the Passaic River was found near a high sedimentation area below the

hydro-electric station at the Dundee Dam (Fig.1). Lead concentrations in sediments were higher than the background lead level of approximately 31 mg/kg estimated by Suszkowski (1978) from measurements in deep sediment cores from Newark Bay.

Lead concentrations measured in sediments from the Hackensack wetlands are presented in Table 1. In Area I, concentrations ranged from 121 to 704 mg/kg, with a median concentration of 280 mg/kg. The highest concentration was measured in surficial sediments collected adjacent to the Belleville Turnpike (Site 6). Concentrations in Area II were lower, ranging from 25 to 122 mg/kg, with a median concentration of 41.5 mg/kg.

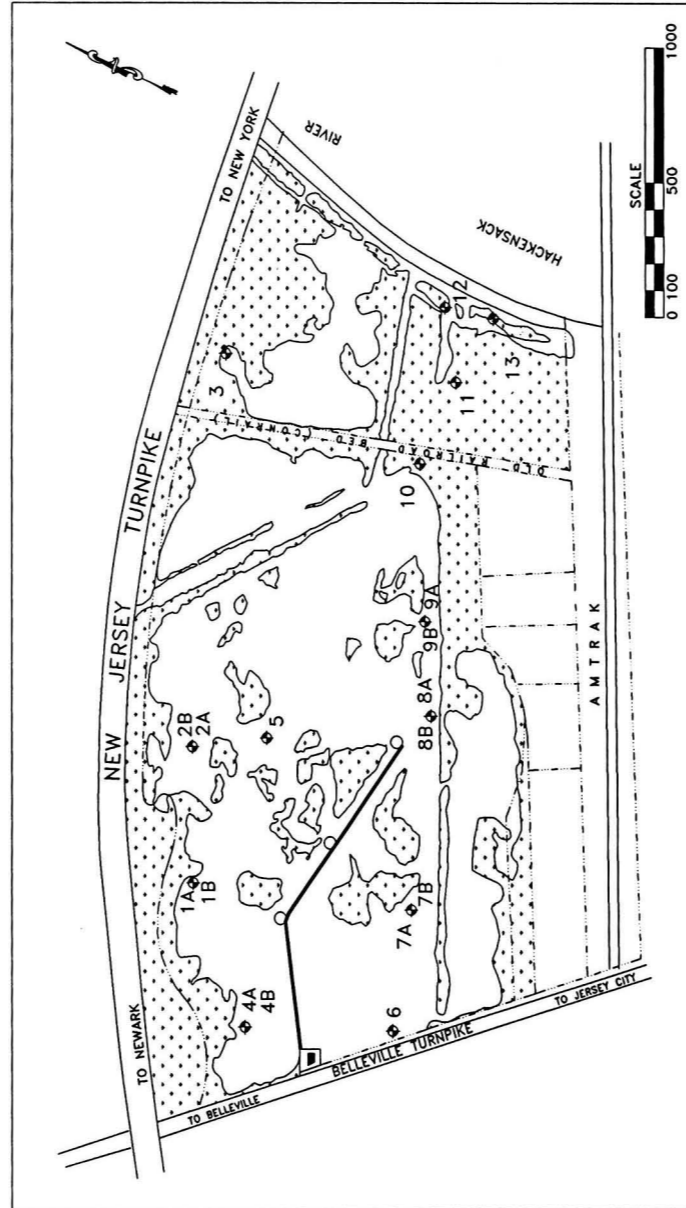


Fig. 2. Sampling locations in the Hackensack Meadowlands Area I.

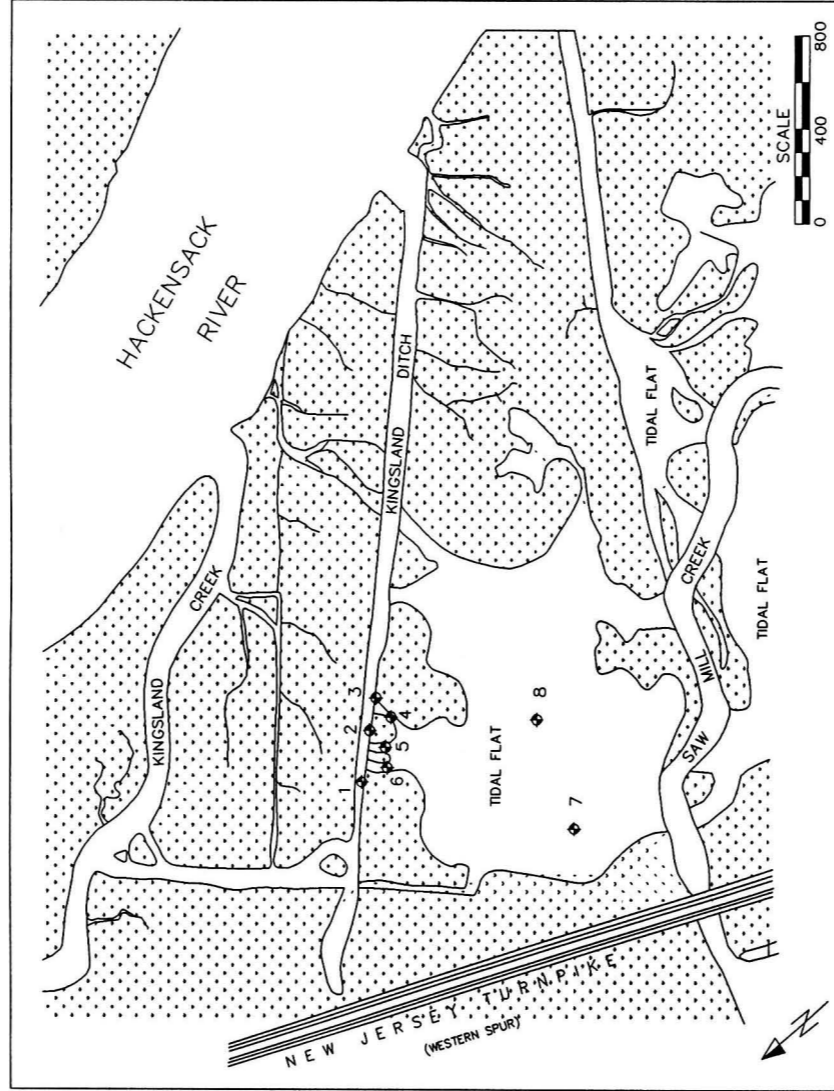


Fig. 3. Sample locations in the Hackensack Meadowlands Area II.

Table 1. Lead concentrations (mg/kg dry wt.) measured in surficial sediments from two wetland areas along the Hackensack River.

Wetland Area	Sample Id	Concentration
Hackensack		
Meadowlands Area I	1A <sup>a</sup>	130
	1B <sup>b</sup>	121
	2A <sup>c</sup>	165
	2B	240
	3A	410
	4A	280
	4B	170
	5B	134
	6B	704
	7A	430
	7B	300
	8A	310
	8B	270
	9A	190
9B	210	
10A	300	
11A	340	
12A	490	
13A	290	
Hackensack		
Meadowlands Area II	1A	122
	2A	25
	3A	30
	4A	52
	5A	31
	6A	33
	7A	102
	8A <sup>c</sup>	50

a. "A" indicates that the sample was collected in October, 1991.

b. "B" indicates that the sample was collected in November, 1991.

c. Concentrations represent the mean of two replicate analyses.

## HAZARD ASSESSMENT

### Comparison to historical data

A summary of earlier studies describing lead levels in surficial sediments from Newark Bay is presented in Table 2. Some of these studies are unpublished, and for many, it is not possible to fully verify the quality assurance. Therefore, the most important use of these data is included for comparative purposes.

In general, the results of these historical studies were similar to those reported in the current investigation. Studies conducted throughout the past two decades have reported lead concentrations ranging from non-detect (<6 mg/kg) in Lower New York Bay (Greig and McGrath 1977) to 3.21 g/kg (dry weight basis) in the Arthur Kill at the confluence with Moses Creek (Meyerson et al. 1981). Similar to the results from the sampling program described above, elevated lead concentrations have historically been

found throughout the lower portion of the Passaic River (Suzzkowski 1978; Meyerson et al. 1981; USEPA 1981), the Hackensack River (Goeller 1989; HMDC 1989), and the Arthur Kill (Meyerson et al. 1981; USEPA 1982). In addition, concentrations as high as 613 mg/kg have been reported in the North Reach Channel of Newark Bay near the confluence with the Passaic River, and in the South Reach Channel near Shooter's Island (Suzzkowski 1978; Meyerson et al. 1981). High lead concentrations have also been reported near the Port Newark Channel (Meyerson et al. 1981).

### Comparison to sediment quality criteria

To evaluate the potential for adverse effects to biota, lead sediment concentrations derived for each waterway were compared to sediment quality criteria and guidelines (Fig. 4). Concentrations in most sediment samples exceeded the Bivalve Larvae AET value of 71 mg/kg proposed in California (CSWRCB 1990), as well as the Effects Range-Median value of 110 mg/kg derived by NOAA (1991a). According to NOAA (1991a), low mortality rates and only minor effects have been observed in marine biota at concentrations below 110 mg/kg. However, behavioral impacts such as reduced burrowing rates and avoidance behavior have been observed in bivalves (*Macoma balthica*) exposed to sediment concentrations as low as 74 mg/kg lead (McGreer 1979).

Lead concentrations measured in Hackensack River sediments exceeded the threshold value of 132 mg/kg derived by the USEPA (1985). Adverse effects have been demonstrated in biota exposed to similar levels of lead in sediment; for example, ten percent mortality has been observed in shrimp (*Palaemonetes pugio*) when exposed to sediments containing 123 mg/kg lead (NOAA 1991a). Moderately toxic effects have been observed in Commencement Bay, Washington sediments containing 170 mg/kg lead (NOAA 1991a). Sediments in most locations in Newark Bay also exceeded the Benthic Apparent Effects Threshold (AET) level of 150 mg/kg value derived by CSWRCB (1990), which represents the concentration at which reductions in species abundance and diversity have reportedly been observed in marine ecosystems (CSWRCB 1990).

In the Arthur Kill, all sediments, except at one location near Prall's Island, approached or exceeded the Canadian Severe Effects Level of 250 mg/kg (Persaud et al. 1991). The Severe Effects Level is the threshold concentration at which significant adverse effects almost always occur in freshwater ecosystems (Persaud et al. 1991). The sensitivity of biota to lead in



Table 2. Summary of lead contamination (mg/kg dry wt.) in the Newark Bay estuary.

Waterway	Range	n	Median Values	Reference
Passaic River	604 - 1,000	3	696	Suszkowski, 1978
	55 - 560	3	72	EPA, 1981
	526	1	--	Meyerson et al., 1981
Hackensack River	97 - 321	5	221	Suszkowski, 1978
	316	1	--	Meyerson et al., 1981
	97 - 249	15	164.3	HMDC, 1989
	110 - 239	8	192	Goeller, 1989
Cromakill Creek	324	1	--	HMDC, 1989
Mill Creek	25	1	--	HMDC, 1989
Berry's Creek Canal	125	1	--	HMDC, 1989
	174	1	--	Goeller, 1989
Sawmill Creek	23.7 - 100.5	2	62	HMDC, 1989
Overpeck Creek	258	1	--	HMDC, 1989
Newark Bay (South Reach) <sup>a</sup>	68 - 499	8	327	Meyerson et al., 1981
	67 - 593	34	313	Suszkowski, 1978
Newark Bay (Middle Reach) <sup>b</sup>	104 - 566	4	206.5	Meyerson et al., 1981
	50 - 455	27	292	Suszkowski, 1978
Newark Bay (North Reach) <sup>c</sup>	70 - 441	6	131.5	Meyerson et al., 1981
	100 - 613	27	287	Suszkowski, 1978
Kill van Kull	205 - 451	3	447	Meyerson, et al., 1981
Arthur Kill	99 - 3,209	4	400	Meyerson et al., 1981
	58 - 613	5	492	Suszkowski, 1978
	220 - 1,100	13	355	USEPA, 1982
Raritan Bay (West End)	40 - 355	13	170	Greig and McGrath, 1977
Raritan Bay (East End)	ND <sup>d</sup> (15) - 210	26	130 <sup>e</sup>	Greig and McGrath, 1977
Sandy Hook Bay	95 - 200	5	170	Greig and McGrath, 1977
Lower New York Bay	ND <sup>d</sup> (6) - 185	23	35 <sup>e</sup>	Greig and McGrath, 1977
Upper New York Bay	140 - 830	3	202	Williams et al., 1978

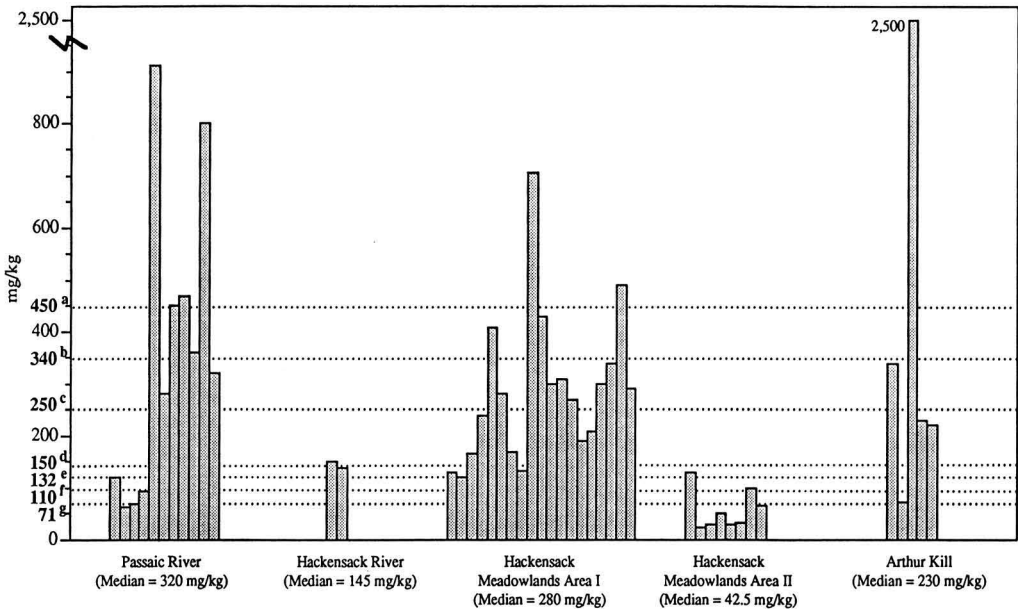
a. The South Reach was defined as the portion of Newark Bay from Staten Island north to the Bayonne City Park.

b. The Middle Reach of Newark Bay was defined as extending from the Port Newark Channel, south to the Bayonne City Park.

c. The North Reach is defined as the portion of Newark Bay extending north from the Port Newark channel to Kearny Point and the confluence with the Hackensack and Passaic Rivers.

d. ND = Non Detect. Detection limit presented in parenthesis.

e. One-half the detection limit was used to calculate the median concentration.



- a. Washington State Department of Ecology (1991) Marine Sediment Criteria.  
 b. California State Water Resources Control Board (1990) Amphipod bioassay AET value.  
 c. Ontario Ministry of the Environment Severe Effect Level (Persaud, 1991).  
 d. California State Water Resources Control Board (1990) Benthic Evaluation AET value.  
 e. USEPA (1985) Threshold Value.  
 f. NOAA (1990) Effects Range - Median.  
 g. California State Water Resource Control Board (1990) Bivalve Larvae bioassay AET value.

Fig. 4. Comparison of lead concentrations in individual surface sediment samples to benchmark sediment quality guidelines.

an estuarine system, such as Newark Bay, could differ from that of a freshwater ecosystem due to differences in sediment and water chemistry (NOAA 1991a). However, in bioassays reviewed by NOAA (1991a), adverse effects were consistently observed in species exposed to sediment lead concentrations above 300 mg/kg in both fresh and estuarine ecosystems. Decreased species abundance has been observed at locations on the Palos Verdes Shelf, California, where lead concentrations as high as 338 mg/kg were measured (Swartz et al. 1985). Sediment lead concentrations greater than 321 mg/kg were reported to result in depressed growth rates in some benthic species in the Hudson-Raritan Estuary (Tietjan and Lee 1984).

Sediments collected at the confluence of the Elizabeth River and the Arthur Kill and at three locations below the Dundee Dam on the Passaic

River exceeded the marine sediment quality criteria of 450 mg/kg developed by the Washington State Department of Ecology (WADOE) using the AET approach (WADOE 1991). Severe toxicity effects and greater than 50 percent mortality in several species including oyster larvae and *R. abronius* have been observed at concentrations above 450 mg/kg (NOAA 1991a).

Lead concentrations in sediments collected from the Hackensack Meadowlands also exceeded several of the benchmark sediment quality guidelines (Fig. 4). Within Area I, all but one sample contained concentrations exceeding the NOAA ER-M value of 110 mg/kg. Eleven of the nineteen samples exceeded the Canadian Severe Effects Level of 250 mg/kg (Persaud et al. 1991). Two samples (Sample 6 and 12) contained concentrations exceeding the marine sediment quality criteria of 450 mg/kg (WADOE 1991).

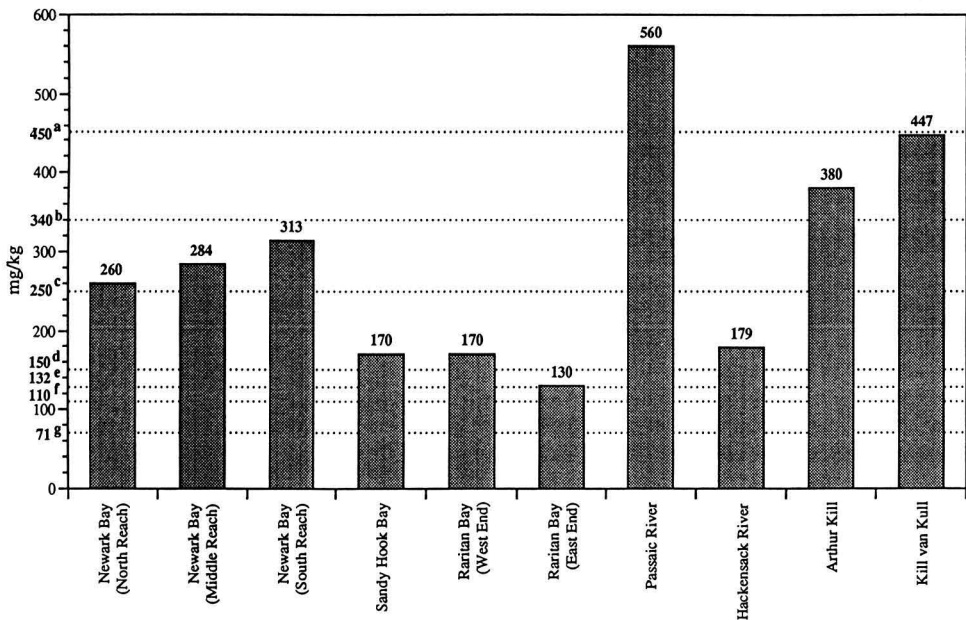
Lead concentrations measured in Area II were lower than those measured in Area I; however, two samples contained concentrations of lead exceeding the CSWRCB (1990) Bivalve Larvae AET value.

Median concentrations derived from historical studies were also compared to benchmark sediment quality guidelines (Fig. 5). These comparisons were similar to those reported in the current investigation. Lead concentrations in sediments were found to exceed NOAA's (1991a) median toxic effects level of 110 mg/kg and the CSWRCB (1990) Bivalve Larvae AET of 71 mg/kg in all parts of the estuary. With the exception of Raritan Bay, the CSWRCB (1990) Benthic AET value of 150 mg/kg was exceeded in all waterways. Median lead concentrations in the North, Middle, and South Reach Channels of Newark Bay, the Kill van Kull, and the Arthur Kill exceeded the Canadian Severe Effects Level of 250 mg/kg (Persaud et al. 1991). The highest median lead concentration (560 mg/kg) calculated from the historical data

was derived for the Passaic River. This value exceeds the WADOE (1991) marine sediment quality criteria.

## DISCUSSION AND CONCLUSIONS

Although little research has been conducted to correlate sediment contamination with observed impacts to biota in Newark Bay, several reviews have suggested that levels of lead and other trace metals found in sediments present in the estuary may be associated with declines in fish populations, lower species diversity, and increased incidences of various infectious and non-infectious diseases (NJMSC 1987; Pearce 1988; Moser 1990). A recent review of available data on the distribution of benthic species in the estuary suggests that the Arthur Kill, Newark Bay, and the Hackensack River are unable to support the expected populations of amphipods, possibly as a result of elevated water and sediment concentrations of metals and organics (Cristini 1991). Extensive fin



- Washington State Department of Ecology (1991) Marine Sediment Criteria.
- California State Water Resources Control Board (1990) Amphipod bioassay AET value.
- Ontario Ministry of the Environment Severe Effect Level (Persaud, 1991).
- California State Water Resources Control Board (1990) Benthic Evaluation AET value.
- USEPA (1985) Threshold Value.
- NOAA (1990) Effects Range - Median.
- California State Water Resources Control Board (1990) Bivalve Larvae bioassay AET value.

Fig. 5. Comparison of median lead concentrations in historical surface sediment samples to sediment criteria, guidelines, and toxic effect values.

erosion has also been observed in several fish species (Mahoney et al. 1973). Other diseases associated with environmental degradation have been reported in several species from the estuary, including lymphocystis, ulcers, and micro-sporidiosis in finfish, and shell disease among anthropods (Cali 1988). Reduced growth rates in several benthic species have been observed in bioassays using substrate from the Arthur Kill, Newark Bay, and the Kill van Kull (Tietjan and Lee 1984). Greig et al. (1982) reported a possible association between extensive gill pathology and the presence of lead and other metals in rock crabs collected near a sewage disposal site in the New York Bight Apex.

Newark Bay is surrounded by a highly urbanized environment and is subjected to substantial pollutant loadings from both point and non-point sources (Meyerson et al. 1981). Pollutants have been estimated to enter the New York/New Jersey Harbor Estuary at a rate of 2 500 million gallons per day ( $9 \times 10^9$  L/d) (HydroQual 1991). Wastewater and atmospheric inputs have been cited as important sources of lead to the estuary (NOAA 1982; Rod et al. 1989). It has been estimated that approximately 27.4% of the lead input to the New York/New Jersey Harbor estuary is from municipal discharges, while 14.2% is from combined sewer outfalls (CSO) (HydroQual 1991). Environmental lead contamination within the estuary can also be associated with the presence of several industries such as paint and pigment manufacturers, refining industries, metal recyclers, and power plants, as well as combustion engines, motor vehicle emissions, and runoff from asphalt roadways (USEPA 1982; Goeller 1989; Rod et al. 1989).

While it is difficult to accurately determine specific sources of lead based on the recent and historical data, several possible sources can be ascertained by examining areas with elevated levels of contamination. For example, the high lead concentrations measured in the upper Arthur Kill may possibly be associated with the presence of several petroleum refining facilities located in the Bayway, New Jersey industrial complex (USEPA 1982). The high concentrations observed along the Passaic River and the lower Hackensack River may be due to the historical presence of scrap metal yards, tanneries, and paint and pigment manufacturers, as well as a municipal water treatment plant at Kearny Point (HydroQual 1991). The distribution of contaminated sediments in the Passaic River may be due, in part, to turbulent conditions generated by the hydroelectric system at the Dundee Dam, as well as discharges from contaminated storm sewer overflows during periods of

heavy rain. Concentrations measured in the Hackensack Meadowlands, where relatively few direct industrial inputs are evident, most likely represent inputs from atmospheric deposition, nonpoint source runoff from urban areas, and automobile traffic from the nearby highways and rail lines. These sediments may reflect the emissions of lead from automobile traffic prior to the regulations limiting the level of lead in gasoline. Although sediment characteristics, tidal currents, dredging activities, and sediment depositional patterns are likely to influence the distribution of sediment bound materials (NOAA 1984; Munns and Rubinstein 1990), the relatively high concentrations found in these areas warrant further investigation.

The results of this investigation indicate that the presence of lead in surficial sediments throughout the Newark Bay estuary may pose an ecological hazard to aquatic biota. Lead concentrations were elevated above estimated background levels in sediment samples collected from nearly all sampling locations. Lead concentrations generally exceeded benchmark sediment quality criteria and guidelines, indicating a potential for significant adverse effects to aquatic biota. Consequently, the presence of lead in surface sediments must be considered a potentially significant factor in the overall environmental quality of Newark Bay. Spiked sediment toxicity and bioaccumulation bioassays are needed to more fully characterize the ecological hazard posed by lead contamination.

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## DRY DEPOSITION OF NITRATE AND SULPHATE ON SURROGATE SURFACES

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Airborne velocities and deposition fluxes (DF) of  $\text{NO}_3^{-1}$  and  $\text{SO}_4^{-2}$  on various surrogate surfaces, specifically petridishes of polypropylene and glass, marble slabs, ceramic tiles, and stainless steel plates, were measured between January and June, 1991. DFs were higher in the summer months, but varied with the surface, decreasing in the order above. In the summer,  $\text{NO}_3^{-1}$  DFs were higher than corresponding  $\text{SO}_4^{-2}$  values. In the winter, this trend was reversed. Deposition velocities of both species were of the order of  $10^{-3}\text{cm s}^{-1}$ .

### INTRODUCTION

The fate of airborne pollutants is controlled by wet and dry removal mechanisms. The immediate and long-term effects of dry deposition (i.e., N and S compounds) and associated acidity are of interest because of their potential influences on materials, structures, forest canopies, plant health, plant biomass production, nutrient balance, fuel accumulation rates, and the fire cycle. Wet and dry deposition processes have been studied extensively in the temperate mid-latitudes (Galloway et al. 1976; Harrison 1987; Davidson et al. 1985; Gorham et al. 1984). Recently, a few studies on surrogate surfaces have also been conducted in temperate regions to explain mechanisms of the dry deposition process of  $\text{SO}_4^{-2}$  and  $\text{NO}_3^{-1}$  on various surfaces. These studies have revealed that the deposition depends on the size, shape, roughness, and composition of surfaces (Davidson et al. 1985; Noll et al. 1988; Bytnerowicz

et al. 1987; Vandenberg and Knoerr 1985), including meteorology (Hicks 1986; Chamberlain 1980; Voldner et al. 1986), particle size (Voldner et al. 1986; Altshuller 1983; Milford and Davidson 1987; Seinfeld 1988), and seasonal variations (Voldner et al. 1986; Davidson et al. 1989).

Experiences gained from studies in the temperate regions cannot be applied to tropical regions where little is known about deposition processes. In the tropics, daytime temperatures are generally high; solar radiation is intense; and strong surface winds are common. This intense sunlight makes many photochemical processes in the atmosphere more efficient. The elevated temperatures and winds render the atmosphere unstable and turbulent during the hot summer months, favoring the dilution of pollutants by dispersion. Yet, high total suspended particulate matter (TSPM) values have been recorded in several tropical countries including India in the dry summer months as compared to the winter (Mahadevan et al. 1989; CUPS 1987; NEERI 1983; Sharma et al. 1983). Soil-oriented particles are driven into the atmosphere by strong winds.

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Unlike in the temperate regions, the tropics either have long, dry spells or a rainy season with persistent daily rain. The tropics are also known to have a different pattern of pollutant emission. In the tropics,  $\text{NO}_2$  is converted to  $\text{HNO}_3$  by reaction with the OH radical—most of which occurs in the troposphere and exists only in the daytime (Rodhe et al. 1988).  $\text{SO}_2$  in the gaseous phase may be converted to sulphate by the hydrolysis-oxidation or the oxidation-hydrolysis mechanism (Durham 1984). The dominance of the two processes is controlled by the relative humidity of the atmosphere. For countries like India, where dry conditions prevail for a major part of the year and precipitation is confined to a short, rainy season, dry deposition seems to be an important process. Although a few studies on wet deposition have been carried out (Khemani et al. 1989; Saxena et al. 1991; Mahadevan et al. 1989), dry deposition and, specifically, work on surrogate surfaces remains unexplored. Therefore, a short-term study has been conducted in Agra to quantify the seasonal dry deposition fluxes of atmospheric  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  particles on different surrogate surfaces; glass and polypropylene petridishes, ceramic tiles, stainless steel plates, and marble slabs. Surrogate surfaces have been extensively used to monitor dry deposition (Bytnerowicz et al. 1987; Davidson et al. 1985; Dasch 1983). They provide a common surface for application in a variety of environments, and they furnish the only means by which a chemical sample can be obtained with convenience (Davidson et al. 1985). These surfaces were of particular interest to us, since Agra is famous for the Taj Mahal and other monuments constructed of marble and redstone, in addition to structures of other building materials.

## EXPERIMENTAL

### Site description

Agra ( $27^{\circ}10'N$ ,  $78^{\circ}05'E$ ) lies in the semi-arid zone, adjacent to the Thar desert of Rajasthan. Dayalbagh which lies on its northwestern outskirts has no major industries except agriculture. Summers in Agra are characterized by high daytime temperatures (maximum temperatures ranging between 33 and  $44^{\circ}\text{C}$  during April, May, and June) and cool nights (minimum temperatures range between 17 and  $31^{\circ}\text{C}$ ). Typical days are sunny with strong dust-laden surface winds. The winter months (November to February) are cool (maximum temperatures range between 16 and  $27^{\circ}\text{C}$  and minimum temperatures are between 5 and  $17^{\circ}\text{C}$ ).

The sampling site was the roof of the Faculty of Science building at our institute. The height of the roof is about 8m and the surrounding area is completely free of constructions or tall trees that could obstruct wind flow.

### Sample collection

**Dry deposition.** After washing all surfaces with detergent and deionized water, they were clamped horizontally at the same level to laterally extending arms of an iron stand. Since there were six surfaces, the arms were arranged such that the angle they subtended to each other was  $30^{\circ}$ . This apparatus was set up in duplicate at a distance of about 5m from the first. Samples collected in the winter were exposed from 8-18 January and 23 January-2 February, 1991. Those collected in the summer were exposed from 11 March-1 April, 6-16 April, 25-27 May, 28-31 May, and 1-10 June, 1991.

The surfaces were exposed for longer periods during the winter in order to achieve concentrations that were within the detection limits of the method of analysis (Ion Chromatography on a Dionex 2000i/SP).

At the end of the exposure period, deposition on the surfaces was washed into polyethylene bottles using 50-75 mL of deionized water (in fractions) and the final volume of each was made up to 100 mL.

### Aerosol

Airborne  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were sampled using Whatman EPM 2000 glass fibre filter paper with a high volume sampler (HVS, APM 415 Envirotech Instruments, New Delhi). The HVS was operated at an average flow rate of  $1.35\text{ m}^3\text{ min}^{-1}$  for about 20 h. Aerosol samples were collected on three days (8 January, 11 January, and 12 February, 1991) during the winter period and on every alternate day of the summer sampling period. The filters were weighed before and after sampling and were desiccated for 24 h prior to each weighing. They were then folded twice and a steel die used to punch out four circles (diameter 2.0 cm). These discs were refluxed in 100 mL of deionized water for 2 h to extract the water soluble content and then filtered through Whatman 41 paper into polyethylene bottles which were refrigerated at  $4^{\circ}\text{C}$  until analysis was completed. Filter blanks were treated in the same fashion. Nitrate and sulfate in both the dry deposition and aerosol samples were estimated on a Dionex 2000i/SP Ion Chromatograph using conductivity detection (AS4A column, eluent: mixture of 1.8 mM  $\text{Na}_2\text{CO}_3$  and 1.7 mM  $\text{NaHCO}_3$ , regenerant: 25mN



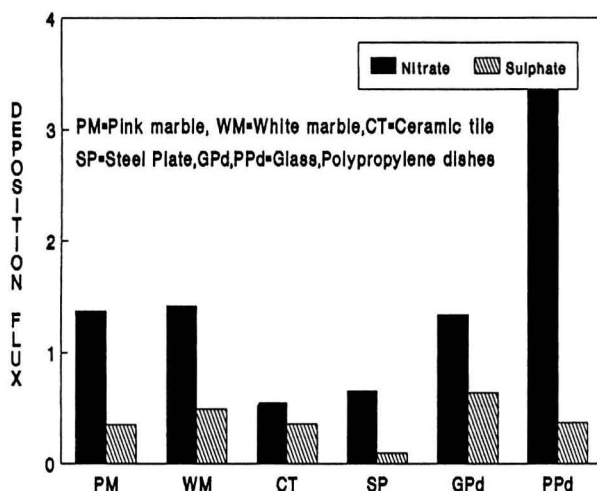


Fig. 1. Deposition flux on surrogate surfaces (in  $\mu\text{g cm}^{-2} \text{ s}^{-1} \times 10^4$ ).

$\text{H}_2\text{SO}_4$ ). Sample and standards were run at a sensitivity of  $30 \mu\text{S}$ .

Meteorological parameters such as maximum and minimum temperatures and relative humidity were also recorded.

## RESULTS AND DISCUSSION

Figure 1 is a histogram showing the variation of deposition flux of  $\text{SO}_4^{-2}$  and  $\text{NO}_3^{-1}$  with the different surfaces. The highest deposition was measured for the polypropylene petridishes and decreased successively for glass petridishes, marble, ceramic tiles, and stainless steel plates. This trend could be attributed to surface roughness and composition. The low deposition flux on the stainless steel plate suggested that its relatively smoother surface compared to the others probably offered less stickiness and adhesive properties; thus, particles bounced off it (Davidson et al. 1985; Noll et al. 1988). The relatively rougher surfaces represented by marble and the ceramic tile prevented the reentrainment of deposited particles and, therefore, had higher deposition fluxes. The greater deposition on glass and polypropylene petridishes could be attributed to the rim. The rim, besides adding to the surface area and preventing the reentrainment of particles, also increased the atmospheric turbulence close to the surface and resulted in greater wind-eddy transport to the petridishes. The observed trend with the rimmed petridishes is the same as reported by Davidson et al. (1985). The

deposition flux values for both  $\text{NO}_3^{-1}$  and  $\text{SO}_4^{-2}$  are found to be low compared to the values reported on surrogate surfaces in California (Bytnerowicz et al. 1987) and in Champaign, Illinois (Davidson et al. 1985).

### *Variation of $\text{SO}_4^{-2}$ and $\text{NO}_3^{-1}$ between and within two seasons*

*Between two seasons.* The sampling period January 1990-June 1991 covers broadly, two seasons; winter from December to February and summer from March to June. Figures 2 and 3 are histograms showing the variation in deposition flux of  $\text{SO}_4^{-2}$  and  $\text{NO}_3^{-1}$  for the winter and summer respectively. The deposition flux of both species on all surfaces is greater in the summer months than in the winter. Our observations may be explained on basis of typical summer conditions in this area where factors such as high temperature, low relative humidity, and strong surface winds result in an unstable and turbulent atmosphere (Voldner et al. 1986) resulting in a net dilution of pollutants. However, the enhanced levels of soil-derived particles in the atmosphere during this season explain the higher deposition fluxes (Mahadevan et al. 1989; CUPS 1987; NEERI 1983; Sharma et al. 1983).

*Within two seasons.* However, within a season, a comparison of the dry deposition fluxes of the two species on the various surfaces revealed that the deposition flux of  $\text{NO}_3^{-1}$  was higher than  $\text{SO}_4^{-2}$  in the

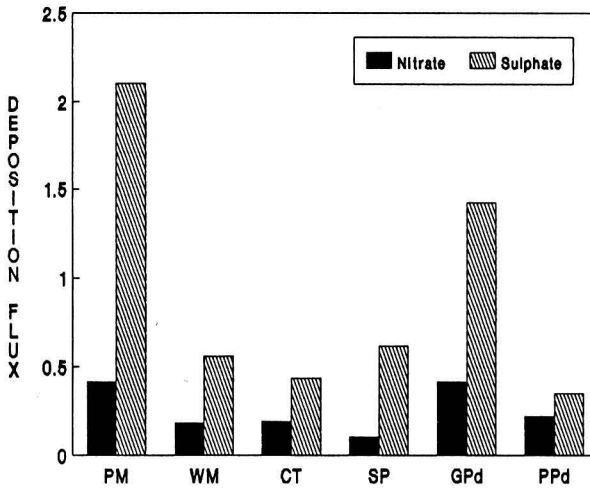


Fig. 2. Winter deposition flux (in  $\mu\text{g cm}^{-2} \text{s}^{-1} \times 10^4$ ).

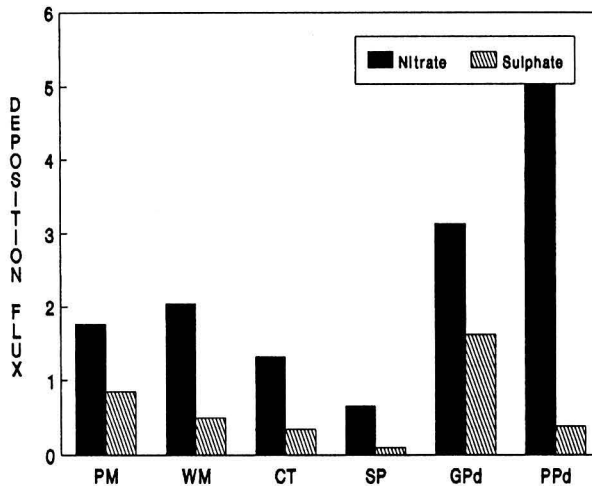


Fig. 3. Summer deposition flux (in  $\mu\text{g cm}^{-2} \text{s}^{-1} \times 10^4$ ).

the summer and that of  $\text{SO}_4^{2-}$  higher than  $\text{NO}_3^{-1}$  in the winter.

In our study, although summer deposition velocities of  $\text{NO}_3^{-1}$  were observed to be higher than corresponding  $\text{SO}_4^{2-}$  values ( $\text{NO}_3^{-1} = 1.13 \times 10^{-3} \text{ cm s}^{-1}$ ,  $\text{SO}_4^{2-} = 0.13 \times 10^{-3} \text{ cm s}^{-1}$ ), atmospheric concentrations of  $\text{NO}_3^{-1}$  were lower (mean winter aerosol concentration =  $8.65 \pm 2.03 \mu\text{g m}^{-3}$ , mean summer aerosol concentration =  $5.94 \pm 1.14 \mu\text{g m}^{-3}$ ) compared to winter values. Although conversion rates

of  $\text{NO}_2$  to  $\text{NO}_3^{-1}$  are highest in the hot, dry summer months (Rodhe et al. 1988), the atmospheric concentrations of the species are at a minimum probably due to greater deposition fluxes, a fact which could be related to the larger particle sizes associated with  $\text{NO}_3^{-1}$  (Voldner et al. 1986; Milford and Davidson 1987). A high  $\text{NO}_3^{-1}$  to  $\text{SO}_4^{2-}$  ratio in the total load of dry deposition has been reported earlier (Bytnerowicz et al. 1987).

With  $\text{SO}_4^{-2}$  particles, winter deposition fluxes were higher than  $\text{NO}_3^{-1}$  values. An explanation for the enhanced winter deposition flux could lie in the fact that the aqueous phase oxidation of  $\text{SO}_2$  (Durham 1984) dominates in this season, aided by the higher humidity (average R.H. recorded = 68% as compared to 36% in the summer). The deposition velocity of  $\text{SO}_4^{-2}$  particles in the winter is, however, found to be less than  $\text{NO}_3^{-1}$  particles ( $\text{SO}_4^{-2} = 0.21 \times 10^{-3} \text{ cm s}^{-1}$ ,  $\text{NO}_3^{-1} = 0.59 \times 10^{-3} \text{ cm s}^{-1}$ ) which could be because  $\text{SO}_4^{-2}$  particles, being in the sub-micron range (Seinfeld 1988; Altshuller 1983), are not so efficiently scavenged.

## CONCLUSION

The low deposition fluxes of  $\text{NO}_3^{-1}$  and  $\text{SO}_4^{-2}$  in this area despite high SPM values indicate low pollution levels in Agra. Compared to values reported for California, USA, our deposition fluxes are lower by three orders of magnitude. Moreover, since relative humidities are low for the major part of the year, the possibility of damage to monuments seems to be remote.

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## OZONE: CONCENTRATION VARIABILITIES IN A SEASONALLY DRY TROPICAL CLIMATE

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This paper reports seasonal and diurnal variations in the ambient air concentrations of ozone in Varanasi city, India for the years 1989 and 1990. Different monitoring zones were established in the city on the basis of pollution sources, traffic densities, and structure of built-up areas. At most of the monitoring stations, two-hourly ozone concentrations frequently exceeded  $60 \mu\text{g m}^{-3}$  (0.03 ppm) during summer months. However, the zone dominated by residential colonies, offices, and cultivated land showed maximum  $\text{O}_3$  concentrations less than  $60 \mu\text{g m}^{-3}$  (0.03 ppm). Higher concentrations of ozone were mainly recorded in warmer months between 11 am to 4 pm. The highest concentration (2-h mean) of  $160 \mu\text{g m}^{-3}$  (0.08 ppm)  $\text{O}_3$  was recorded in the month of April at Zone I which was mainly due to emissions from heavy-duty automobiles. Monthly average ozone concentrations were significantly correlated with temperature ( $r = 0.69$ ), relative humidity ( $r = -0.81$ ), wind speed ( $r = 0.76$ ), and atmospheric pressure ( $r = -0.46$ ). The frequency distribution of 2-h mean ozone concentrations was close to the logarithmic normal. Maximum 2-h mean  $\text{O}_3$  concentrations in some areas of Varanasi city were above the limit that has been shown to cause significant vegetation damage. The temporal variation in ozone concentrations was mainly due to the variations in meteorological conditions.

### INTRODUCTION

Many pollutants in urban air are phytotoxic in nature. For temperate countries, attention has been paid to acid-forming gases, but in countries with warmer climates, secondary pollutants like ozone have become the most important phytotoxic component of urban air (Mansfield and Freer-Smith 1981; Lefohn et al. 1989). Ozone, being a secondary pollutant, is not emitted as such by any specific source, rather it is formed during the atmospheric photochemical reactions involving oxides of nitrogen and reactive hydrocarbons emitted from automobiles (Krupa and Manning 1988). These reactions are principally controlled by sunlight. The increasing emission of reactive hydrocarbons and nitrogen oxides in urban areas has significantly increased the ground level ozone concentrations.

In urban areas like Varanasi city, where the climate is characterized by distinct wet and dry seasons and high levels of automobile emissions, elevated levels of ozone can be expected, specially during summer, when the maximum day time temperature reaches  $40^\circ\text{C}$ .

Ozone data from developed countries have shown diurnal variations. Importance of peak concentrations in significant vegetation damage has been emphasized (Lefohn and Mohnen 1986; Lefohn et al. 1989; Jaser et al. 1991). In India, however, there are no published data on diurnal patterns of  $\text{O}_3$  concentrations. There is no air quality standard for ozone. The basic objective of this study is to report the trends in ambient ozone concentrations. The times of peak occurrence of ozone concentrations were recorded and the relationships between ambient  $\text{O}_3$  concentrations and meteorological variables were drawn.

MATERIALS AND METHODS

The study area is situated in the eastern Gangetic plain of the Indian sub-continent. Varanasi city, population 1 026 500, is located at 25°18' N latitude and 83°01' E longitude and 76.19 m above sea level. The climate is tropical with a marked monsoonal

effect. The year is divisible into three distinct seasons: summer (March to June), rainy (July to October), and winter (November to February), with annual averages of 1000 mm precipitation, 24°C temperature, and 60% relative humidity. The first half of the summer season is associated with strong, hot dry winds and high temperature, while the second

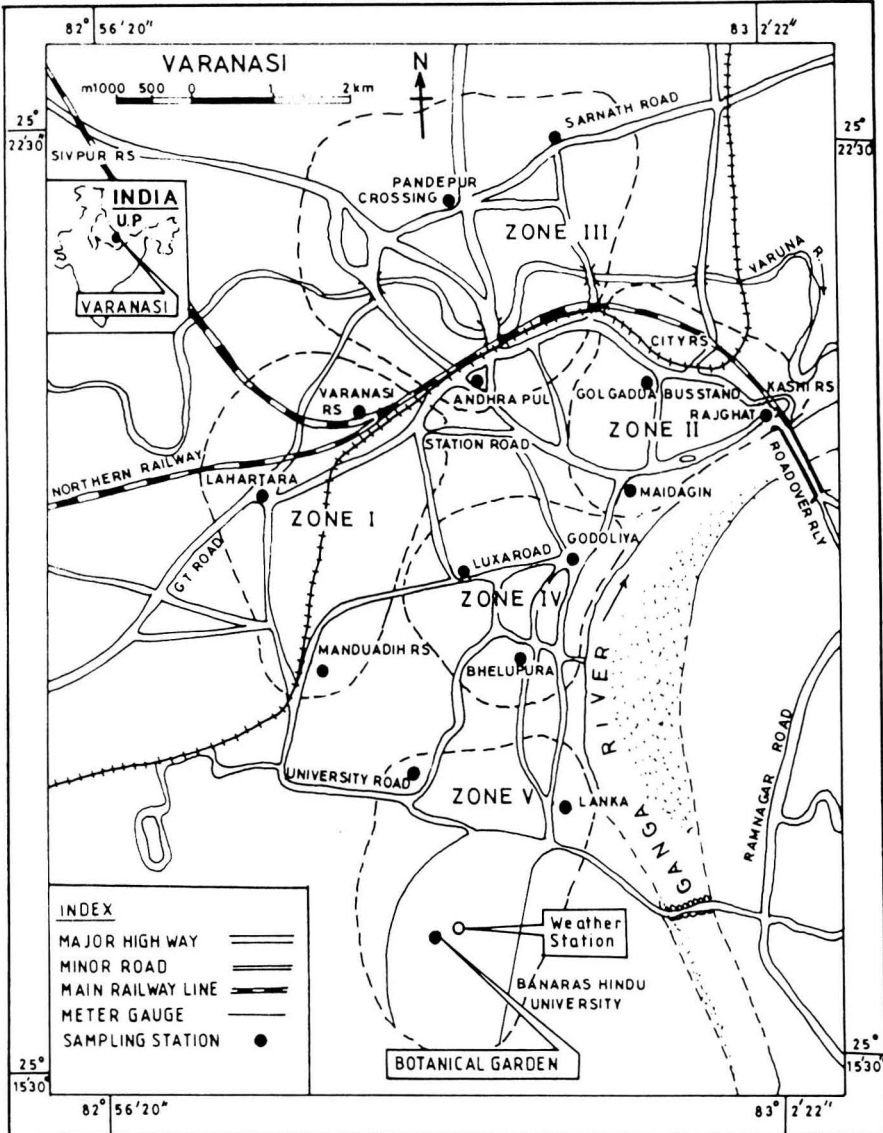


Fig. 1. Locations of monitoring sites in Varanasi city, India.

half is generally hot and humid. Temperature rises rapidly in March and reaches maxima of greater than 42°C in May and June. The rainy season which accounts for 90% of the total annual rainfall, starts at the end of June and continues until mid-October. During this period, the maximum temperature ranges from 24°C to 36°C, and the relative humidity from 70 to 95%. The mean monthly temperature falls after November. In winter, the day temperature varies between 10-25°C, and the night temperature sometimes drops below 6°C. Winter precipitation in the form of occasional light rains is received from the returning western monsoon. Wind direction shifts from predominantly westerly and northwesterly in October through April to easterly and northwesterly in the remaining months.

For air quality assessment with particular reference to O<sub>3</sub>, the city was divided into five zones. Three monitoring stations were set up in each zone (Fig. 1). Two of the zones (I and II) represent the area along major highways which receive emissions from motor vehicles, small scale industries, and railway tracks. The third zone (Zone III) is characterized by moderate traffic, but frequent traffic congestion. The fourth zone represents an area of highly congested shopping and residential complexes, receiving emissions from light vehicles and houses. The fifth zone represents a residential area with official buildings, gardens, and cultivated lands.

Air monitoring was started in January 1989 and continued until December 1990. The monitoring was carried out at 15 different sites by using High Volume Samplers located at 1.5 to 3.0 m above ground level. Ozone was scrubbed in buffered KI (0.1 N)

solution. Samples were collected at 2-h intervals and were analyzed colorimetrically (Byers and Saltzman 1958).

Meteorological data were collected from the Department of Geophysics, Banaras Hindu University, Varanasi.

## RESULTS AND DISCUSSION

Maximum ozone concentrations, monitored as daily and 2-h means at different zones of Varanasi during 1989 and 1990, are shown in Table 1. The majority of the monitoring sites experienced 2-h mean ozone concentrations above 40  $\mu\text{g m}^{-3}$  (0.02 ppm). The exception was Zone V which remained less influenced by local urban sources. In Zone V, maximum daily mean O<sub>3</sub> concentrations ranged from 12 to 20  $\mu\text{g m}^{-3}$  and maximum 2-h mean concentrations from 21 to 36  $\mu\text{g m}^{-3}$ . High concentrations of ozone ( $\geq 100 \mu\text{g m}^{-3}$ ) were more frequent in Zones I and II. Besides the high traffic densities, these zones receive emissions from railway and small scale industries. The third and fourth zones have somewhat intermediate levels of O<sub>3</sub>, ranging from 55 to 116  $\mu\text{g m}^{-3}$ . The highest 2-h mean concentration recorded in this study was 160  $\mu\text{g m}^{-3}$  (0.08 ppm) in Zone I. The concentrations found in our study agree with those reported at Trombay, India during 1982 (Netravalkar and Mohan Rao 1984). Ozone levels in Varanasi are, however, significantly lower than hourly ozone concentrations of 1180  $\mu\text{g m}^{-3}$  during October 1978, at Claremont, U.S.A. (Tuazon et al. 1981) and 800  $\mu\text{g m}^{-3}$  at upland, Los Angeles, U.S.A. during October 1980 (Grosjean 1983).

Table 1. Seasonal variations in maximum ozone concentrations ( $\mu\text{g m}^{-3}$ ) monitored as 24-h average and 2-h average (in Parenthesis) at different zones of Varanasi city during 1989 and 1990. W = winter season (November-February); S = summer season (March-June); R = rainy season (July-October).

Sites	1989			1990		
	W	S	R	W	S	R
I	27 (65)	68(160)	41 (76)	26 (71)	74(149)	49 (83)
II	26 (60)	64(149)	42(102)	30 (60)	81(159)	38 (92)
III	20 (55)	43(101)	36 (80)	24 (61)	59(105)	41 (73)
IV	24 (58)	48(116)	32 (61)	24 (69)	60(110)	30 (64)
V	14 (25)	17 (36)	12 (21)	16 (26)	20 (30)	14 (26)

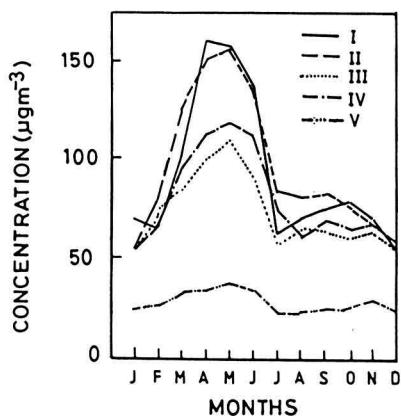


Fig. 2. Monthly variations in maximum 2-h mean ozone concentrations in five different zones of Varanasi city.

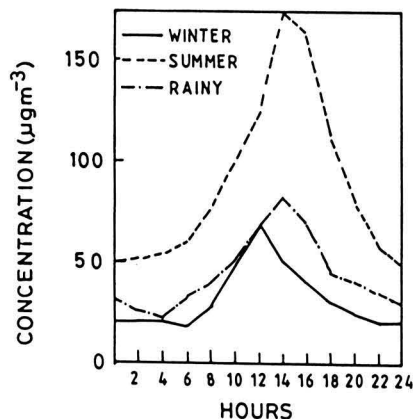


Fig. 3. Maximum seasonal 2-h mean ozone diurnal concentration patterns in Zone I.

The monthly variations in maximum ozone concentrations (2-h means) for different zones are shown in Fig. 2. Ozone concentrations varied from 52 to 160  $\mu\text{g m}^{-3}$  in Zone I with the highest in April and lowest in December. During April/May, maximum ozone values exceeded 75  $\mu\text{g m}^{-3}$  at most of the monitoring stations. On rare occasions, ozone peaks exceeded 150  $\mu\text{g m}^{-3}$  at Zones I and II. Lefohn and

Jones (1986) observed a consistent pattern of ozone maxima in the area influenced by local urban sources. Ozone formation depends much upon the intensity of solar radiation and air temperature. Significant positive correlation was observed between mean monthly and maximum temperature and monthly average concentrations of ozone ( $r = 0.69$ ,  $p < 0.01$ ). This indicates temperature-de-

Table 2. Correlation matrix of monthly mean ozone concentrations and meteorological variables.

$T_{\max}$  : Maximum temperature;  $T_{\min}$  : Minimum temperature;  $R_H$  : Relative humidity \*;  $p < 0.05$ ; \*\*;  $p < 0.01$ ; NS: Not significant.

	$O_3$	$T_{\max}$	$T_{\min}$	Rain-fall	$R_H$	Wind speed	Pressure
$O_3$	1.0	0.80**	0.43*	-0.27 <sup>NS</sup>	-0.81**	0.76**	-0.46*
$T_{\max}$		1.0	0.82**	0.27 <sup>NS</sup>	-0.49*	0.75**	-0.78**
$T_{\min}$			1.0	0.52**	0.049 <sup>NS</sup>	0.63**	-0.92**
Rainfall				1.0	0.52**	0.19 <sup>NS</sup>	-0.57**
$R_H$					1.0	-0.11 <sup>NS</sup>	0.09 <sup>NS</sup>
Wind speed						1.0	-0.75**
Pressure							1.0



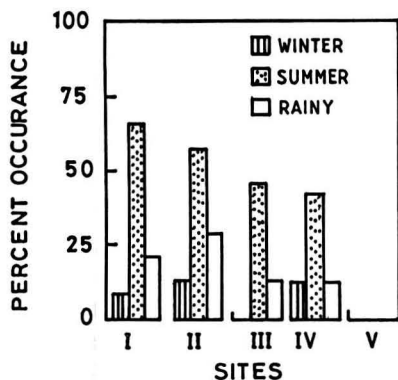


Fig. 4. Frequency distribution of 2-h average ozone concentrations exceeding  $60 \mu\text{g m}^{-3}$  (0.03 ppm). A comparison between different zones.

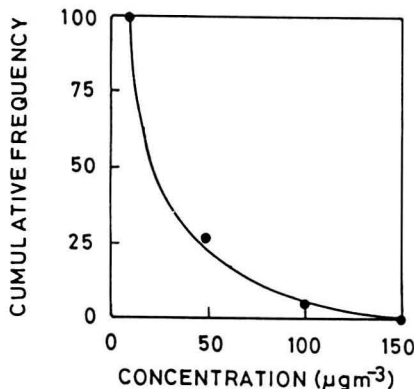


Fig. 5. Cumulative frequency distribution of 2-h mean ozone concentrations in Varanasi city (all sites combined).

pendent ozone formation resulting in ozone maxima during the afternoon. This agrees with the results of Bruckmann and Langensiepen (1981), who showed a significant positive correlation between  $\text{O}_3$  concentration, temperature, and solar radiation. Low concentrations of ozone in winter months were due to lack of strong solar radiation and low pressure. Ozone concentrations were negatively correlated with relative humidity and atmospheric pressure ( $p < 0.01$ ). However, a significant positive correlation was obtained between  $\text{O}_3$  concentration and wind speed. This was expected, as the wind speed was negatively correlated with temperature and pressure ( $p < 0.01$ ) (Table 2).

Figure 3 shows the diurnal variations in maximum 2-h mean ozone concentrations in Zone I during winter, summer, and rainy seasons. Such patterns occurred quite frequently at all monitoring stations. The peak concentrations of  $\text{O}_3$  at all monitoring sites occurred at mid-day and late afternoon in winter and summer seasons, respectively. Ozone concentrations were low in the late evening and early morning hours, and were extremely low at night due to lack of solar radiation and the frequent presence of  $\text{NO}$  as  $\text{O}_3$  scavenging species. A similar diurnal pattern was observed by Lefohn and Tingey (1984) from three air quality data bases (SAROAD, SURE and TVA), and by Lefohn and Jones (1986) at Big Meadows Shenandoah National Park and Sawmill Run Shenandoah National Park, Virginia, U.S.A.

Figure 4 shows the percent occurrence of 2-h mean  $\text{O}_3$  concentrations exceeding  $60 \mu\text{g m}^{-3}$  (0.03 ppm)

at five different zones. Daily occurrence of  $\text{O}_3$  with 2-h concentrations  $60 \mu\text{g m}^{-3}$  (0.03 ppm) was frequent in summer months. In Zone V, however, ozone did not equal or exceed this limit. Figure 5 shows the cumulative frequency distribution of 2-h mean  $\text{O}_3$  concentrations. About 75% of the 2-h mean  $\text{O}_3$  concentrations were less than  $50 \mu\text{g m}^{-3}$  (0.025 ppm). Most of the air samples were in the low concentration range; only few of the samples were in the high concentration range. The cumulative frequency distribution is found to be close to a logarithmic normal distribution pattern. Similar distribution patterns of ozone concentration were also observed by Lefohn and Jones (1986) in the areas influenced by local urban sources.

## CONCLUSION

Maximum 2-h mean  $\text{O}_3$  concentrations in Varanasi equalled or exceeded the limit that has been shown to cause significant vegetation damage. Ozone concentrations varied significantly among different seasons; the maximum being in summer (March-June), and the minimum in winter (November-February). All the monitoring sites influenced by urban emission experienced peak  $\text{O}_3$  concentrations from late morning to afternoon of the summer months. Spatial variation in  $\text{O}_3$  concentrations were mainly due to the variation in emission sources. Temporal variations, however, seemed to be due to the variations in meteorological conditions as evidenced by correlation coefficients.

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# USE OF POULTRY MANURE FOR AMENDMENT OF OIL-POLLUTED SOILS IN RELATION TO GROWTH OF MAIZE (*Zea mays* L.)

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The use of poultry manure for amelioration of oil-polluted soil was investigated by growing maize (*Zea mays* L.) under two experimental conditions: increasing the poultry manure rate from 0-20 kg ha<sup>-1</sup> at 0.03 L/kg oil treatment level; and increasing the rate of oil treatment from 0 - 0.2 L/kg, while the poultry manure amendment remained at 20 kg ha<sup>-1</sup>. A linear correlation existed between the rate of poultry manure added and the enhancement of maize growth ( $r = 0.99$ ). But only a 16-kg ha<sup>-1</sup> poultry manure rate and above exerted some beneficial effects on the maize growth relative to the unpolluted, unamended soil. Conversely, increasing oil concentration, regardless of the poultry manure level added (20 kg ha<sup>-1</sup>), depressed maize growth, but only at oil levels of 0.03 L/kg. A positive correlation was recorded between maize height and leaf area growing in oil-treated soil (0.03 L/kg) amended with different poultry manure rates ( $r = 0.99$ ) and growing in oil-treated soils (0 - 0.2 L/kg) amended with 20 kg ha<sup>-1</sup> poultry manure ( $r = 0.90$ ). Amending oil-contaminated soils with poultry manure, should possibly improve soil fertility and maize production.

## INTRODUCTION

Agronomic problems associated with oil-contaminated soil ecosystems have been identified by Odu (1972), Udo and Fayemi (1975), Cook and Westlake (1976), and Bossert and Bartha (1984). Nitrogen and phosphorus become limited in soil following oil pollution (Jobson et al. 1974), thus impeding biodegradation of oil. Any available nutrient element, e.g. N or P in soil, is rapidly assimilated by soil microbes, because of their capacity to rob plants of scarce nutrient reserves. Therefore, aside from the

environmental problems of oil pollution, the microbiological, agronomic, and economic aspects are of major significance.

The objective is to augment the native fertility status of such soils to enhance the rate of oil biodegradation, minimise contamination of scarce ground water sources, and improve soil and crop production.

Several reports have addressed the fate and/or effect of oil pollution on soils (Ellis and Adams 1961; Schweindinger 1968; Odu 1972), microbes (Atlas 1981) and crop/plants (Baker 1970; Udo and Fayemi

1975; Amadi 1990). However, the adoption of remediation measures to rapidly rehabilitate the soils has received comparatively little attention (Toogood and McGill 1977; Song et al. 1990; USEPA 1990). These reports have addressed temperate zones only. Similar studies are needed for the tropics.

The use of organic nutrient supplements for remediation of stressed environments is one potential option that has yielded good results (Jobson et al. 1974; Amadi 1992). The prospects for organic manure usage appear greater.

This study involving two experiments was designed to test the use of varying levels of poultry manure on oil-treated soil; and to optimize the poultry manure rate, while increasing oil concentration.

## MATERIALS AND METHODS

This study covered a period of about 14 weeks from date of soil treatment with oil. The crude-oil type used was the Bonny (Nigeria) light crude with specific gravity of 0.8343, containing sulphur at less than 30 mL/L. Soil was collected from a fallow patch of land within the university complex. The soil of the area was characterized as a sandy loam Typic Paleustult (ultisol). Surface soil (0-20 cm) was collected using a shovel and bulked to form a composite sample which was transferred to the laboratory, air

dried, sieved through a 2-mm mesh, and analysed. Treated soils were analysed again prior to planting (Table 1). All soils were transferred into 3-kg portions in a 5-kg size plastic planting pot, watered to their field capacity for two days, and then treated with crude oil to their respective levels. Soils were subsequently amended with poultry manure after one week of oil treatment, and allowed to decompose for another four weeks before planting the maize seeds.

### Experiment I

The prepared soils in 5-kg planting pots were treated to give a 0.3-L/kg oil concentration, except for the untreated pots that served as experimental control. All pots were amended with 0-20 kg ha<sup>-1</sup> poultry manure, one week after oil treatment. Then, four seeds were planted per pot five weeks after initial treatment of the soils with oil, and thinned to two at two weeks after planting (WAP).

The design was a randomized complete block, replicated four times. Plant height and leaf area were measured. For leaf area measurement, length and width at the broadest portions were taken. All measurement were made at harvest.

Table 1. Soil properties before experimentation, prior to planting, and after harvest.

Parameter	Before Amendment	After Amendment (at harvest)	
		(a)	(b)
Texture	Sandy loam	Sandy loam	Sandy loam
pH	5.90	6.20	6.60
Organic Carbon (g/kg)	23.9	31.4	41.4
Total Nitrogen "	1.9	2.2	2.1
Available PO <sub>4</sub> - P "	0.041	0.056	0.078
Exchangeable Cations			
- Ca <sup>2+</sup> (meq/100g soil)	3.40	6.78	6.18
- Mg <sup>2+</sup> "	0.20	2.48	2.10
- K <sup>+</sup> "	0.82	1.48	1.48
- Na <sup>+</sup> "	0.19	0.43	0.41
Manganese(mg/kg)	1.17	0.38	0.52
Ferrous (g/kg)	0.054	0.047	0.056
Petroleum hydrocarbon content (Total) (g/kg)	0.004	570.0	685.0

(a) represents mean values of four replicates at 0.03 L/kg oil level.

(b) represents mean values for soil polluted with 0 - 0.20 L/kg oil and amended with 20kg/ha<sup>-1</sup> poultry manure. Petroleum hydrocarbon content (total) on columns (a) and (b) represents decreases from 100% initial treatment.

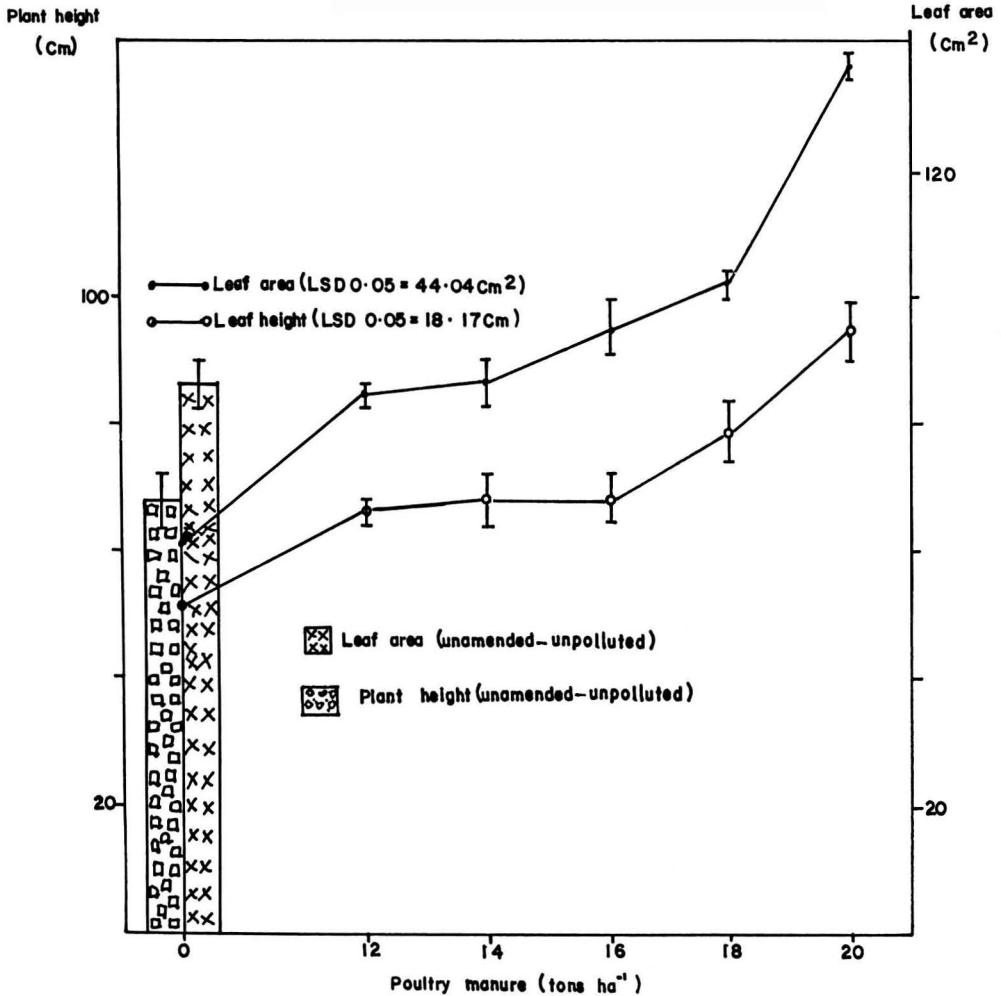


Fig. 1. Changes in plant height and leaf area of maize (*Zea Mays* L.) after application of varying rates of poultry manure in oil-polluted soil. Vertical bars indicate the standard deviation from the mean.

**Experiment II**

Soil samples were prepared and treated with crude oil and poultry manure as above, but at different application rates. For example, the oil level was increased beyond the critical level of 0.03 L/kg (except in the untreated control), to include 0.05, 0.10, and 0.20 L/kg. The poultry manure rate however, remained at optimum 20 kg ha<sup>-1</sup>. Two additional treatments were incorporated to act as a double control or check (Amadi 1992). First, soil was not treated with oil and not amended with poultry manure. Second, soil was treated with 0.03-L/kg oil but un-

amended with poultry manure. The objective was to compare the significance of oil treatment and nutrient supplementation on the recovery of soil and growth of maize.

Four maize seeds were planted per pot at the 5th week after the initial oil treatment, and thinned to two per pot after two weeks. The experimental design and measurement of physical parameters were the same as in Experiment I.

Results obtained were subjected to statistical analysis, i.e. significance test to distinguish the rate effect for oil and manure; and regression/correlation

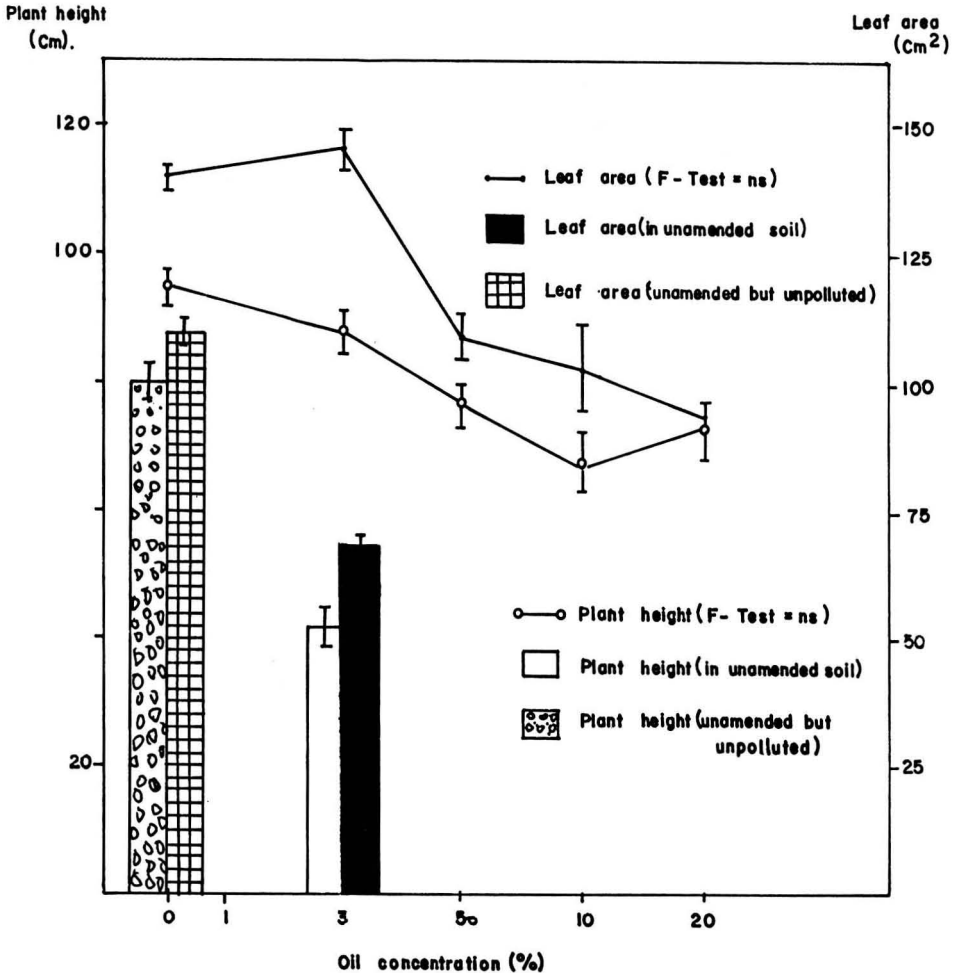


Fig. 2. Changes in plant height and leaf area of maize (*Zea mays* L.) in soil contaminated with varying concentrations of oil but remediated with a uniform rate of poultry manure (20 tons ha). Vertical bars indicate the standard deviation from the mean.

analysis to show how plant height and leaf area interrelate or co-vary under different treatment combinations.

#### Application of oil/manure

Appropriate quantities of oil reflecting their respective treatment rates were measured and applied evenly on soil surfaces, and allowed to sink gradually prior to further treatment with manure.

Poultry manure was applied by incorporating appropriate quantities into the soil and stirring to ensure even distribution within the soil volume. All treatments were watered with tap water at four days

interval to 50% of their field capacity. Poultry manure was applied on dry-weight basis to a maximum of 20 kg ha<sup>-1</sup>.

#### Analytical methods

Soil properties before and after experimentation were estimated. About 30 sampling stations were marked in a 1-ha plot in the field. At each station, four random points were located and 0-20 cm portions (top soil) collected using a shovel. All four points were bulked together and then all 30 stations were bulked to form a composite that was representative of the entire area under study.

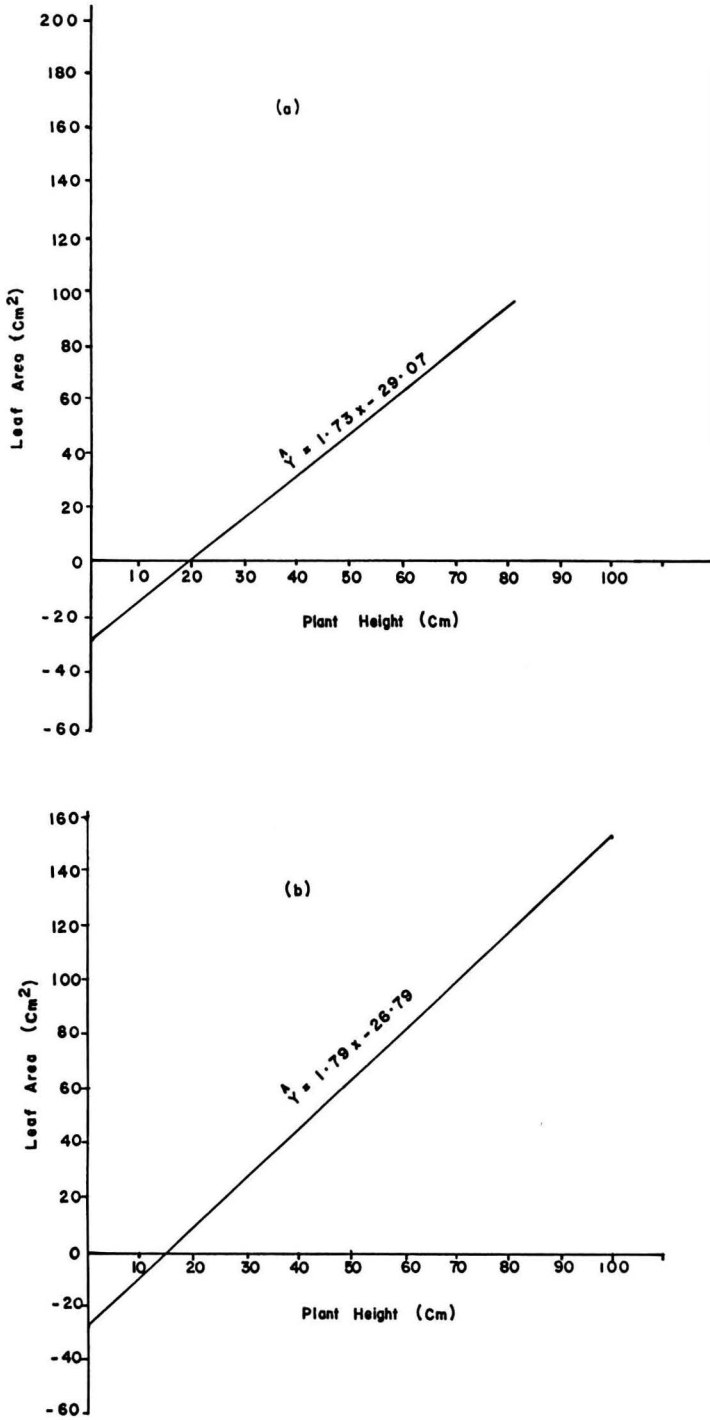


Fig. 3. Regression curve for maize height (x) and leaf area (y) in oil-polluted (3%) soil, amended with varying rates of poultry manure (a); and soils polluted to varying levels of oil but amended with 20-tons ha<sup>-1</sup> poultry manure in each case (b).

Four samples were analysed per parameter per treatment and the mean values reported. Standards originated from the Microbiology/Environmental Studies Laboratory, Department of Agronomy, University of Ibadan, Nigeria, which also served as a reference laboratory throughout this study. Blanks and standards were re-run at random to enhance quality assurance.

Textural classification was conducted using the hydrometer method (Day 1953); the pH of the soil was determined in a mixture of a 1:2.5 soil/water suspension using an electrode probe on a Cole-Palmer pH meter.

Other chemical parameters were analysed as follows: organic carbon by the wet combustion method of Walkley and Black (1934); total nitrogen by the Macro-Kjeldahl method (Bremner 1965); and available phosphorus by the Bray and Kurtz P1 method (1945).

Exchangeable cations were determined after extraction of the soil with 1-N ammonium acetate. Calcium, phosphorus, and sodium were determined using a flame photometer, while magnesium and other trace elements, i.e. ferrous and manganese, were determined using atomic absorption spectrophotometer.

## RESULTS AND DISCUSSION

The morphological status (height and leaf area) of maize in oil-treated soil amended with varying rates of poultry manure are shown on Fig. 1. Figure 2 illustrates the growth characteristics of maize in soil treated to different concentrations of crude oil and amended with optimum levels of poultry manure.

Addition of poultry manure enhanced plant height and leaf area with a linear correlation existing between poultry manure rates and maize growth pattern. The beneficial effect of poultry amendment became evident at or above a 16-kg ha<sup>-1</sup> level as shown in Fig. 1. On the other hand, increasing oil concentration generally depressed maize growth regardless of the amendment rate applied, as evidenced in the declining curve of Fig. 2.

A better inference of the contribution of poultry manure on maize growth is illustrated in the bar of Fig. 2. At a 0.03-L/kg oil level in the unamended soil, both leaf area and plant height decreased significantly ( $p = 0.05$ ) relative to their respective amended and oil treated soils.

Our observations show a pattern. The linear relationship between increasing rates of poultry manure and enhanced maize growth is expected, since a certain amount of degradation of crude oil would have oc-

curred in the soil over a long time. Secondly, the C, N, and P ratio may have narrowed as a consequence of the amendment with poultry manure and/or through decomposition and mineralization. With a narrowed C/N ratio and availability of essential nutrients to microbes, degradation was enhanced as shown by rate of soil recovery (Table 1). This observation agrees with the finding of Bossert and Bartha (1984) and Cook and Westlake (1976).

Figure 3 shows the correlation and regression analyses of growth characteristics of maize in soil with a 0.03-L/kg oil level and in amended soil ( $r = 0.99$ ); with oil treatment in varying rates, i.e. of 0-0.2 L/kg and in amended soil of 20 kg ha<sup>-1</sup> ( $r = 0.90$ ). Amendment with organic manure was responsible for the increased maize height and leaf area. This is corroborated by the soil analysis result presented in Table 1. The soil increased in fertility after the oil treatment/poultry manure amendment.

Given, that the pattern of decomposition varies with soil type and biota, the presence of any toxic degradation products may alter the soil's physico-chemical status. However, the nutritional status of the manure suggests that increasing the poultry manure rate up to 20 kg ha<sup>-1</sup> exerted a corresponding positive effect on the maize growth. For instance, unamended soil showed negative impact at an oil concentration of 0.05 L/kg; but at <0.3 L/kg oil, poultry manure amendment of 20 kg ha<sup>-1</sup> enhanced maize growth even more than unoiled, unamended soil.

Again, the decrease in maize height and leaf area shown in Fig. 2 was remarkable. However, the enhanced growth of maize represented in Fig. 3 is an indication of the positive effect of soil amendment with poultry manure. As with bioremediation treatment under suitable conditions (Song et al 1990), it should be possible to ameliorate soil conditions in oil-polluted tropical soils for maize cultivation.

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## BOOKS

*Environmental Quality; 22nd Annual Report; The Council on Environmental Quality*, U.S. Government Printing Office, Washington, D.C., 1992. 382 pp. (ISBN 0-16-036168-0) softcover.

*U.S. Waste Management Policies: Impact on Economic Growth and Investment Strategies*, American Council for Capital Formation; Center for Policy Research, Washington, D.C., 1992, 104 pp., \$25.00 softcover.

*U.S. Environmental Policy and Economic Growth: How Do We Fare?* American Council for Capital Formation; Center for Policy Research, Washington, D.C., 1992, 131 pp. \$25.00 softcover.

The report of the Council of Environmental Quality (CEQ) is mandated by U.S. law and is a compilation of information prepared by various U.S. agencies, notably the U.S. Environmental Protection Agency. It covers topics ranging from agriculture to wetlands. Considering that it is prepared by a governmental agency and deals with a highly contentious issue, the report is surprisingly factual and unbiased.

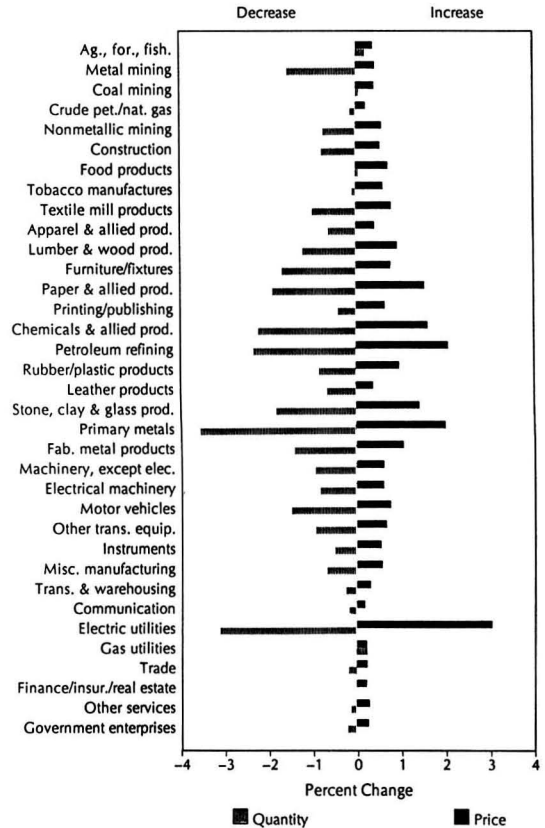
The first 15 pages of this report are in fact the report of the CEQ to the U.S. Congress. A major segment of the report deals with the economics of pollution control. According to the CEQ, the total cost of pollution control in the U.S. in 1990 was \$115 billion, of which about \$30 billion was devoted to solid waste. Note that the costs of the Clean Air Act Amendments of 1990 were not included because that law took effect after 1990.

The report of the American Council for Capital Formation (ACCF) on waste policy discusses the same issue in some detail. In that report, Paul Portney representing Resources for the Future argues that these costs are significantly lower because the ordinary pickup of garbage costs \$18 billion. This cost would have to be incurred regardless of environmental regulations. In effect, Portney concedes that there are actions that need to be taken and actions that are merely desirable to take, a concept that is hotly disputed by environmental advocacy groups.

A most interesting part of the ACCF is an estimate of the total cost of clean up of uncontrolled disposal

sites. The estimate for private sector costs is over \$1 trillion, with those of governmental agencies approximating \$300 billion. Considering that the gross national product of the U.S. in 1990 was approximately \$5.5 trillion, these are very large expenditures even for a U.S.-size economy.

The report of ACCF on environmental policy contains cost estimates for compliance with the Clean Air Act of 1990. The following figure shows increases in price and decreases in quantity of products associated with compliance with that act extrapolated for the year 2005. According to this figure, electric utilities and primary metals will be most adversely impacted by the requirements of this law.



Regulation*	Year issued	Health or safety	Cost per premature death averted million \$
Unvented Space Heater Ban	1980	S	0.1
Aircraft Cabin Fire Protection Std.	1985	S	0.1
Auto Passive Restraint/Seat Belt Std.	1984	S	0.1
Steering Column Protection Std.**	1967	S	0.1
Underground Construction Stds.***	1989	S	0.1
Trihalomethane Drinking Water Stds.	1979	H	0.2
Aircraft Seat Cushion Flammability Std.	1984	S	0.4
Alcohol and Drug Control Stds.***	1985	H	0.4
Auto Fuel-System Integrity Std.	1975	S	0.4
Stds. for Servicing Auto Wheel Rims***	1984	S	0.4
Aircraft Floor Emergency Lighting Std.	1984	S	0.6
Concrete & Masonry Construct. Std.***	1988	S	0.6
Crane Suspended Pers. Plat' m Std.***	1988	S	0.7
Passive Restraints - Truck & Bus (P)	1989	S	0.7
Side-Impact Stds. for Autos (Dynamic)	1990	S	0.8
Children's Sleepwear Flam'ility Ban****	1973	S	0.8
Auto Side Door Support Stds.	1970	S	0.8
Low-Alt. Windsh'r Equip. & Train. Std.	1988	S	1.3
Electrical Equip't Stds. (Metal Mines)	1970	S	1.4
Trenching & Excavation Stds.***	1989	S	1.5
Traffic Alert & Collision Avoidance Sys.	1988	S	1.5
Hazard Communication Std.***	1983	S	1.6
Side-Impact - Truck, Buss & MPV (P)	1989	S	2.2
Grain Dust Explosion Prev'n Stds***	1987	S	2.8
Rear Lap/Shoulder Belts for Autos	1989	S	3.2
Radionuclides - Uran. Mines Stds.***	1984	H	3.4
Benzene NESHAP (Fugitive Emis'n)	1984	H	3.4
Ethylene Dibromide Drinking Water Std.	1991	H	5.7
Benzene NESHAP (Coke By-Prod.)***	1988	H	6.1
Asbestos Occup'l Exposure Limit***	1972	H	8.3
Benzene Occup'l Exposure Limit***	1987	H	8.9
Electrical Equip. Stds. (Coal Mines)***	1970	S	9.2
Arsenic Emis'n Stds. - Glass Plants	1986	H	13.5
Ethylene Oxide Occup'l Expos. Limit***	1984	H	20.5
Arsenic/Copper NESHAP	1986	H	23.0
Haz Waste List - Petrol. Refin. Sludge	1990	H	27.6
Cover/Move Uran. Tail'gs (Inact. Sites)	1983	H	32
Benezene NESHAP (Trans. Oper'ns)	1990	H	33
Cover/Move Uran. Tailings (Act. Sites)	1983	H	45
Acrylonitrile Occup'l Expos. Limit***	1978	H	52
Coke Ovens Occup'l Expos. Limit***	1976	H	64
Lockout/Tagout***	1989	S	71
Asbestos Occup'l Expos. Limit***	1986	H	74
Arsenic Occup'l Expos. Limit***	1978	H	107
Asbestos Ban	1989	H	111
Diethylstilbestrol (DES) Cattlefeed Ban	1979	H	125
Benezene NESHAP (Waste Oper'ns)	1990	H	168
1,2-Dichloropropane Drink. Water Std.	1991	H	653
Haz Waste Land Disposal Ban (1st 3rd)	1988	H	4,190
Municipal Solid Waste Landfill Std. (P)	1988	H	19,107
Formaldehyde Occup'l Expos. Limit***	1987	H	86,202
Atrazine/Alachlor Drinking Water Std.	1991	H	92,070
Haz Waste List - Wood Preser. Chems.	1990	H	5,700,000

The CEQ report estimates the cost of lives saved as a consequence of environmental controls. In other words, governmental actions result in a reduction of risk. Note that health risks are upper level estimates and true risks (median) are often one to two orders of magnitude lower. The preceding table is taken from the CEQ report.

Clearly, environmental costs are disproportionately high and require significantly more attention than they have been receiving. The CEQ report and the reports of ACCF on environmental policy and waste management are mandatory reading for environmental policymakers and those concerned with protection of human health and the environment.

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*Clean Air Act Amendments Conference; An Economic Opportunity for Illinois.* Conference Proceedings, 10-11 October 1991, Chicago, Il. The University of Illinois at Chicago, Chicago, Il., 1991. 345 pp. (Library of Congress No. 91-067711) softcover.

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*The Science of Christian Economy; and other prison writings,* by Lyndon H. LaRouche, Jr. Schiller Institute, Washington, D.C., 1991. 506 pp. (Library of Congress No. 91-062722) softcover.

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## NEW PATENTS

This section contains abstracts and, where appropriate, illustrations of recently issued United States patents and published patent application filed from over 30 countries under the Patent Cooperation Treaty. This information was obtained from recent additions to the PATSEARCH<sup>®</sup> online database in accordance with interest profiles developed by the Editors.

Further information about online patent searching can be obtained from Research on Demand, Inc., 2421 Fourth Street, Ste. C., Berkeley, CA 94710, U.S.A. [Tel. 510-841 1145; Fax 510-841 6311].

**5090972**

### **PARTICULATE ABATEMENT AND ENVIRONMENTAL CONTROL SYSTEM**

Joe C Eller, James E Leavens, Charles H Wyatt assigned to Enviro-Air Control Corporation

Improved techniques are provided for improving the environment of hazardous material abatement personnel operating within an enclosed working area while conducting, for example, asbestos removal operations. A negative pressure is established within the working area by a portable air moving unit, which exhausts air from the working area to prevent hazardous material leakage. The exhausted air is filtered, a significant portion of the exhausted air is conditioned by a portable refrigeration, heating and dehumidification system, and the temperature controlled air is returned to the working area. An air diverter is provided for regulating the ratio of the discharged air to the returned and conditioned air in response to the sensed pressure level within the working area. The temperature of the working area is regulated, thereby substantially increasing worker productivity and reducing safety risks. The humidity level within the working area may either be lowered to reduce the curing time for hazardous material final encapsulation operations, or increased to reduce the airborne contaminant level during wet abatement operations. Leakage from the working area is preferably minimized, while the efficiency of the portable conditioning unit is maximized.

**5091317**

### **ANALYSIS OF ACIDIC METAL CARBONYL HYDRIDE CONTAINING STREAMS**

James L Cooper, Jack Bogle assigned to Eastman Kodak Company

A method for determining the concentration of an acidic metal carbonyl hydride in an organic liquid by measuring the conductivity or pH of the liquid. The air addition to a continuous air demetallating unit may be controlled by feedback from the conductivity or pH measurement of the effluent.

**5092254**

### **ACID GAS CONTROL PROCESS AND APPARATUS FOR WASTE FIRED INCINERATORS**

Peter Kubin, Jiri Stepan assigned to Ogden-Martin Systems Inc

A method and apparatus for incinerating waste material and for reducing noxious byproducts of the incineration process. The apparatus consists of a furnace having a turbulent reaction zone whereby an overfire air header and additive distributor and lime injection nozzles connected to the incinerator through a plurality of nozzles located at even distances about the arrangement is the turbulent reaction zone of the furnace such that an additive mixture is injected into the turbulent zone through and distributed evenly during combustion across the entire width of the turbulent area. As a result, a reaction between the combustion products and the additive is optimized resulting in an overall reduction in acid gas content, acid dewpoint temperature and the corrosion levels inside the incinerator and auxiliary equipment.

**5094752**

### **AEROBIC WASTEWATER TREATMENT WITH ALKALINITY CONTROL**

H Forbe Davis, James P Harshman assigned to Davis Water & Waste Industries Inc

Aerobic wastewater treatment processes are controlled utilizing alkalinity measurements. A base line of alkalinity is determined by measuring the alkalinity profile of the influent to the aerobic wastewater treatment process. The alkalinity of the wastewater is sensed at a number of different points in the aerobic wastewater treatment process, such as at a clarifier effluent and sludge exit, aeration zone, and aerobic digester effluent. When the sensed alkalinity at any given point is above a predetermined amount over base line alkalinity, the air supply is increased and/or the concentration of microbes and food is reduced (as by increasing the sludge

withdrawal rate from a clarifier) until the sensed alkaline level is stabilized. When the sensed alkalinity is below a predetermined amount under base line, the air supply is decreased and/or the concentration of feed and microbes increased (as by reducing or terminating sludge withdrawal from a clarifier) until the sensed alkalinity level is stabilized. Anaerobic odors are eliminated, plant operation and efficiency is improved, settling is improved, there is enhanced removal from the secondary clarifier, and solids carryover is reduced increasing the quality of effluent from the clarifier.

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**Book:** Henderson, P.M. Inorganic geochemistry. New York, NY: Pergamon Press; 1982.

**Regulation:** USEPA (U.S. Environmental Protection Agency). National primary drinking water regulations: fluoride. 40 CFR Parts 141, 142 and 143. Fed. Reg. 50:47142-48933; 1985.

**Proceedings:** Swedjemark, G.A.; Mj6nes, L. Exposure of the Swedish population to radon daughters. Berglund, B.; Lindvall, T.; Sundell, J., eds. Proc. 3rd international conference on indoor air quality and climate. Vol. 2. Stockholm: Swedish Council for Building Research; 1984:37-43.

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