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Certifying the contents of trimethyllead in an artificial rainwater reference material

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Certified Reference Materials are needed to verify the performance of analytical techniques used in lead speciation and for the quality control of trimethyllead determinations in environmental matrices. An interlaboratory programme has been conducted by the Standards, Measurements and Testing Programme to improve the state of the art of trimethyllead determination in urban dust and artificial rainwater. This paper describes the preparation of an artificial rainwater reference material containing trimethyllead, its homogeneity and stability studies and the analytical work performed for the tentative certification of the trimethyllead content at a level of ≈ 55 ng kg^{-1} (as trimethyllead). While the analytical results obtained by the certifying laboratories were considered to be suitable for certification, doubts on the long term stability of the material did not allow certification to be recommended.

Keywords: *Trimethyllead; rainwater; intercomparison; reference material; quality control*

Environmental contamination by lead is widespread, the major anthropogenic source of this element being the combustion of leaded gasoline. Although their use has been discontinued in some countries, the use of tetraalkyllead compounds as anti-knock agents remains the largest application of organolead compounds.[1] It is recognized that, owing to its ubiquity and concern over the toxicity of organolead compounds in the environment,[2] the monitoring of lead species will need to be continued over the next decade. Vehicular emissions of tetraalkyllead are subject to atmospheric breakdown to trialkyl- and dialkyllead and all three forms are scavenged from the atmosphere by rainfall.[3] Therefore, trimethyl- and triethyllead are found in road drainage and surface water.[4] As a consequence, a number of laboratories are performing analyses of *e.g.* rainwater and urban dust to monitor the levels of trialkyllead compounds in the environment. The techniques are generally based on a combination of different analytical steps including extraction, derivatization (*e.g.* ethylation or Grignard reactions), separation [*e.g.* gas chromatography (GC) or high performance liquid chromatography (HPLC)] and detection [*e.g.* atomic absorption spectrometry (AAS) or atomic emission spectrometry (AES)] which all pose risks of systematic errors (*e.g.* incomplete extraction, inhibition of derivatization reaction, incomplete separation *etc.*). In order to verify the performance of analytical techniques used in lead speciation and the quality control of trimethyllead determinations, certified reference materials are needed. As a response to this need, the Standards, Measurements and Testing Programme (formerly BCR) has recently produced an urban dust reference material certified for its trimethyllead content, CRM 605;[5] this paper describes the attempt to certify a reference material of artificial rainwater (RM 604) for its trimethyllead content.

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Preliminary investigations

In view of the urgent need to establish the state of the art of lead speciation analysis, a project was discussed and designed with a group of European laboratories in the framework of the Measurements and Testing Programme (formerly BCR) of the Commission of the European Communities. A feasibility study was carried out to investigate the stability of alkyllead compounds in solution[6] showing that dialkyllead compounds and triethyllead were not sufficiently stable in aqueous solution to be of use as possible reference materials. The only compound which could be stored without significant degradation was trimethyllead and, therefore, this compound was selected for the preparation of lead-containing solutions. Interlaboratory studies were designed[7,8] as the first step in a larger project whose aim was to produce reference materials of simulated rainwater and urban dust certified for their content of trimethyllead.

Organisation of the project

After a preparatory meeting in which all the requirements for certifying reference materials were discussed and restated, two bottles of the candidate CRM were shipped to the participating laboratories (see acknowledgements). Each laboratory that took part in the exercise was requested to perform six independent replicate determinations on at least two different bottles of the reference material on different days. The results were statistically evaluated, presented in the form of bar-graphs and discussed at a technical meeting to which all the participants were invited. The determinations were only performed when the method was under statistical control. Considering the difficulty inherent in calibration, the organiser of the certification provided the participants with trimethyllead calibrants which were checked for their stoichiometry and purity.

Feasibility study

Preparation and verification of calibrants

One of the most critical points in organometallic chemistry analysis is the availability of calibrants of suitable purity and verified stoichiometry. This aspect was recognised at an early stage of the project and the purity of alkyllead compounds used in the feasibility study was carefully verified.[3] Additional experiments were performed on calibrants in the frame of the first interlaboratory exercise as described below.

Trimethyllead (TriML) and triethyllead (TriEL) compounds were obtained from Alfa products (Johnson Matthey) and their purity was verified as follows: carbon, hydrogen and chloride relative masses in the TriML and TriEL calibrants were

determined by elemental microanalysis; the chloride concentration was determined by ion chromatography. Total lead was determined in the calibrants by electrothermal atomic absorption spectrometry (ETAAS) using two different acid digestion procedures (concentrated nitric acid and nitric acid–hydrogen peroxide). Calibrants solutions of TriML and TriEL at the 25 mg l^{-1} level were prepared in distilled deionised water (DDW) and analysed; a 200 μl aliquot of each of these solutions was added to a solution containing NaCl (2 g), 0.5 mol l^{-1} NaDDTC (2 ml) and 0.1 mol l^{-1} EDTA (1 ml) in 30 ml DDW, and the mixtures were shaken manually in a separating funnel. Hexane (5 ml) was added to the funnel and the aqueous phase was removed after shaking for 4 min. The extracted alkyllead compounds were then re-extracted into dilute nitric acid–hydrogen peroxide, and determined by ETAAS.

An aqueous solution containing 500 ng l^{-1} of TriML and TriEL was prepared and extracted as above (except for the addition of nitric acid and hydrogen peroxide). The hexane extract was transferred to a 25 ml conical flask and 0.5 ml of propyl magnesium chloride (Grignard) reagent was added, followed by gentle shaking for 8 min. The extract was then washed with 0.5 mol l^{-1} H_2SO_4 (5 ml) to destroy any excess Grignard reagent present. The organic phase was separated then dried with a minimum of anhydrous Na_2SO_4 and transferred to a 4 ml vial. TriML and TriEL were determined by GC–AAS. Students *t*-tests were used to compare the experimental results obtained with the predicted values.

The results showed that the content of alkyllead in the calibrants was slightly less than 100% and that extraction–analytical losses were the likely causes for this, rather than inorganic lead (since this was not significantly different from the expected concentration). In addition, if there was contamination by inorganic $PbCl_2$ in the calibrants, the concentration of chloride obtained by microanalysis and ion chromatography, would have been higher than expected. In the two chloride analyses, it was found that the chloride concentration was slightly lower than expected, probably due in part to the tetraalkyllead detected in the calibrants (around 2% of the total lead content). In the light of these results, it was thought unlikely that significant amounts of either inorganic lead or other ionic alkyllead compounds were present. It was concluded, therefore, that the two trialkyllead calibrants were not less than 98% pure.

Interlaboratory studies

For the first intercomparison exercise, participants were asked to dilute the solution 1000-fold, *i.e.* to determine levels of TriML of ≈40 μg l^{-1}. Some laboratories also analysed the solutions after a 10 000-fold dilution.

The participating laboratories in the second interlaboratory study received two sets of solutions containing ≈50 and 5 μg l^{-1} of TriML respectively. They were requested to perform five replicate analyses of, respectively: 10-fold dilution of the 50 μg l^{-1} concentrated solution (solution A); 10-fold dilution of the 5 μg l^{-1} concentrated solution (solution B); 100-fold dilution of the 5 μg l^{-1} concentrated solution (solution C); and 1000-fold dilution of the 5 μ l^{-1} concentrated solution (solution D).

Sample preparation

The sample preparation was performed by the School of Chemistry, University of Birmingham (Birmingham, UK). Preliminary investigations have shown that ethyllead compounds are more sensitive towards degradation than the corresponding methylated compounds;[5] TriML was found to be stable in solutions kept at ambient temperature in the dark. Consequently, a batch of solutions containing 40 mg l^{-1} of trimethyllead chloride (as Pb) and 100 mg l^{-1} of lead nitrate (as Pb) added as interferent was prepared and the stability was verified over a period of six months.

Two sets of simulated rainwater solutions were prepared. Aliquots of solution (100 ml) were transferred into eighteen 100 ml Nalgene bottles for each of the samples. The bottles were wrapped with aluminium foil and then sealed in plastic bags.

Results

In most cases, the pretreatment techniques used were based on complexation, GC separation and employed various different detection techniques [*e.g.* microwave induced plasma (MIP), AES, AAS, ICP-MS]. No specific comments were made during the meeting to discuss the first interlaboratory study, except that the participants considered it impossible to correct the results for impurities in the calibrant matrix. The second interlaboratory study generated detailed discussed which are summarised below.

Solution A (10-fold dilution of 50 μg l^{-1} *solution)*

One ETAAS technique employed did not include a separation step but the participants stated that EDTA extraction would only extract organic lead compounds; this technique was considered to be suitable for the analysis of a simple solution containing only one lead compound but would not be suitable for mixtures of lead species, *e.g.* the technique would not allow the separation of TriML and TriEL in a natural rainwater sample. In cases where different organolead compounds are to be determined in natural samples or solutions containing different lead compounds, ETAAS should be coupled to a separation technique, *e.g.* GC or HPLC.

A systematic difference was observed by one laboratory between two different sets of calibrants (calibrant solution made with a newer calibrant from the same producer). This highlighted the need to thoroughly verify the calibrant, *i.e.* not to rely on calibrants from one producer of which the quality could vary from one batch to another. Most of the laboratories actually used their own calibrants which were not verified for purity and stoichiometry. Only one laboratory used the calibrant previously verified and distributed in the first interlaboratory study.[1] It was stressed that calibration was an important issue and that more efforts should be put on the verification of calibrants in future exercises. It was agreed that the coordinator of the project would purchase calibrant from a chemical company and establish its purity; sets of verified primary calibrants would then be made available to participants in a further exercise to characterise their own calibrants.

The verification of extraction recoveries was also discussed. Most of the laboratories performed standard addition procedures and hence did not need to correct for recovery. The relative standard deviation (RSD) between laboratories was originally 20.9%. After technical scrutiny and rejection of suspect data, the RSD decreased to ≈4% which was found to be an excellent agreement.

Solution B (10-fold dilution of 5 μg l^{-1} *solution)*

An ICP-MS technique was still under development and not optimised at the level of concentration considered (0.49 ± 0.16 μg kg^{-1}).

DPASV was below the detection limits in one case; the reliable results could be obtained by doubling the deposition time (240 s instead of 120 s); the participant mentioned, however, that this concentration corresponded to the limit of determination of his technique.

The RSD between laboratories was 14.8% before the evaluation and dropped to 10% after some sets of results were

removed on technical grounds. This degree of agreement was found acceptable at this level of TriML concentration.

Solution C (100-fold dilution of 5 μg l^{-1} solution)

A good agreement was obtained between laboratories at this level of concentration (RSD of ≈10% between laboratories).

Concluding remarks on the interlaboratory studies

The preparation of samples for a possible certification exercise was discussed. The participants agreed that a solution containing 500 ng l^{-1} of TriML in a 10-fold concentrated artificial rainwater solution would be a suitable candidate reference material. The bulk solution should be analysed as such and after a 10-fold dilution in order to obtain values matching the concentration levels found in a natural rainwater samples (≈50 ng l^{-1}).

Preparation fo the candidate CRM

The composition of the artificial rainwater matrix was chosen to reflect that of natural rainwater which falls over continental land masses.[9] A stock solution was prepared at 100-times the required concentration of the artificial rainwater by the addition of inorganic compounds to deionised water. One litre of stock solution was prepared by the addition of the following compounds (Analytical reagent grade) to 1000 ml of Milli-Q deionised water: $CaCl_2$ (11.3 mg); $MgCl_2$ (9.5 mg); $(NH_4)_2SO_4$ (39.6 mg); $NaNO_3$ (34.0 mg); NaCl (11.7 mg); KCl (3.7 mg); and HCl (17.4 μl).

The components were dissolved using an ultrasonic bath and the final solution was filtered through a 0.2 μm (47 mm diameter) cellulose acetate membrane in order to eliminate algal and bacterial particles. No problems were encountered with solubility of any of the components of the stock solution.

Trimethyllead chloride (purity 98%) was used for spiking the candidate CRM. A large batch of artificial rainwater (100 l) was prepared in a rigid high-density polythene bin (0.14 m^3) with a lid. The solution was protected from light by wrapping the bin in aluminium foil. Trimethyllead chloride was added to give a concentration of 500 ng l^{-1} (as lead). The solution was thoroughly stirred for an extended period of time to ensure proper mixing by means of a glass rod which was passed through a hole in the lid of the container. The solution was dispensed into Nalgene bottles (125 ml) by syphoning it through a Teflon tube (6.4 mm diameter) which was passed through the hole in the container lid. The Nalgene bottles (800 units) were rinsed out with two small aliquots of the solution and 100 ml of the artificial rainwater matrix was dispensed to each bottle. The bottles were capped, wrapped in aluminium foil, sealed in polythene bags and then stored in a cold room at +4 °C.

The concentrations of the components present in the stock solution and in the artificial rainwater are presented in Table 1.

Table 1 Concentration of artificial rainwater components

Component	Concentration in stock solution/μmol l^{-1}	Concentration in final solution/μmol l^{-1}
NH_4^+	600	60
K^+	50	5
Ca^{2+}	120	12
Mg^{2+}	100	10
Na^+	600	60
Cl^-	900	90
SO_4^{2-}	300	30
NO_3^-	400	40
$(CH_3)_3Pb^+$	500 ng kg^{-1}	50 ng kg^{-1}

Homogeneity study

The between-bottle homogeneity of the reference material (RM 604) was verified by a single determination of TriML in each of eight bottles randomly selected from the 800 bottles produced. The methodological uncertainty was determined by performing five replicate determinations of the trimethyllead content in one bottle. The artificial rainwater samples (100 ml) were diluted with 900 ml of Milli-Q deionised water prior to analysis. Sodium chloride (20 g), 5 ml of 0.25 mol l^{-1} NaDDTC and 5 ml of hexane were added to the diluted sample and the solution was shaken mechanically for 30 min. The organic fraction was transferred to a 25 ml conical flask, 0.5 ml of propylmagnesium chloride was added and the flask was gently shaken for 8 min. The extract was washed with 5 ml of 0.5 mol l^{-1} H_2SO_4 to destroy any excess Grignard reagent present. The organic phase was dried over anhydrous Na_2SO_4 (≈100 mg) and then transferred to a 4 ml vial. A 50 μl extract was injected into a GC instrument and trimethyllead was determined by AAS. The extraction efficiency for trimethyllead was determined by spiking the sample with 100 ng l^{-1} of the compound and performing the analysis as described above. The mean recovery for 4 replicates was 90.2% with an RSD of 2.9%.

The RSDs ($\pm RSD/\sqrt{2n}$) for TriML in the reference material were 6.8 ± 1.7% and 4.3 ± 1.4% for between-bottle homogeneity and method variances. An *F*-test at a significance level of 0.5 did not reveal significant differences between the between-bottle and method variances. On the basis of these results, no inhomogeneities of the material were suspected. It was concluded that the material is suitable for use as a CRM.

Stability study

The stability of the trimethyllead content was tested to determine the suitability of this material as a candidate CRM. Bottles were kept in the dark at 4 °C over a period of 12 months (starting in February 1994) and TriML was determined at regular intervals during the storage period. Analyses were repeated after 37 months (March 1997) to verify the long term stability. In addition, a short-term stability study was carried out at +37 °C to simulate worst-case transport conditions.

Tests were made at the beginning of the storage period and after 1, 3, 6, 12 and 37 months. Samples were analysed using the same procedures as for the homogeneity study. Trimethyllead was determined in triplicate (one replicate analysis in each of three bottles stored at 4 °C) at each occasion of analysis. For the short-term stability at +37 °C, 15 bottles of RM 604 were stored at +37 °C and five of these samples were analysed after 5, 10 and 15 d.

Any change with time in the content of an element or compound indicates an instability provided that good long-term analytical reproducibility is obtained. Instability may be detected by comparing the contents of different elements or compounds at various occasions of analysis, by comparison with the $t = 0$ values.

The results obtained on the samples at $t = 0$ were used as reference for the results obtained at each occasion of analysis ($t = 3$, 6, 12 and 37 months). Table 2 gives the ratios (R_t) of the mean values (X_t) of 3 measurements made after a period t and

Table 2 Normalised results of the stability study

Compound	Time/months	$R_t \pm U_t$
Trimethyllead	1	1.00 ± 0.02
	3	0.99 ± 0.02
	6	0.97 ± 0.02
	12	0.96 ± 0.03
	37	0.86 ± 0.06

the mean value (X_{t0}), from three determinations made at $t = 0$: $R_t = X_t/X_{t0}$.

The uncertainty U_t has been obtained from the RSD of 5 measurements obtained at each temperature: $U_t = (RSD_t^2 + RD_{t0}^2)^{1/2}. R_t/100$.

In the case of ideal stability, the ratios R_t should be 1. In practice, however, there are some random variations due to the error on the measurement. In almost all the cases, the value 1 is comprised between $R_t - U_t$ and $R_t + U_t$. On the basis of the results, it was concluded that no instability of the material could be demonstrated over a period of 12 months. However, the *t*-test result ($p = 0.01$) indicated that the difference between the initial concentration of TriML measured on the day of the rainwater preparation and the final concentration after 37 months storage was significant at the 95% confidence level. On average more than 10% of TriML in rainwater had decomposed at 4 °C after three years storage.

With respect to the data of the short-term stability study carried out at +37 °C (Table 3), an *F*-test (single-factor ANOVA) was used to determine whether or not a significant difference existed among the trimethyllead concentrations determined after 5, 10 and 15 d. According to this test, the stability of trimethyllead in the RM 604 may not be affected by the elevated temperature since the calculated *F*-value is less than the critical *F*-value is less than the critical *F*-value (*p*-value > 0.05); however, this statistical conclusion must be viewed with caution since the calculated *F*-value is so close to the critical *F*-value. The difference between the initial trimethyllead in the material (day 0) and the final concentration (day 15) was also determined using a *t*-test; the statistical result ($p = 0.0486$) at the 95% confidence level showed the TriML had decomposed significantly after 15 d storage at +37 °C.

Table 3 Stability study of trimethyllead in RM 640 at +37 °C after 15 d

Compound	Time/d	Mean ± *s* /ng l^{-1} as Pb
Trimethyllead	0	54.3 ± 4.0
	5	52.7 ± 3.0
	10	49.5 ± 3.2
	15	48.5 ± 3.0

Methods used by the participating laboratories

Techniques used by the participating laboratories were in most cases composed of different analytical steps (extraction, derivatisation, separation and detection), illustrating the high diversity of hyphenated techniques developed for Pb-separation analysis. The analytical techniques applied in this exercise are summarised in Table 4.

Calibration

A trimethyllead calibrant was prepared by the University of Plymouth (Plymouth, UK) for the purpose of the certification campaign in order to enable participating laboratories to verify their calibrants. A portion of 39 g of tetramethyllead and toluene (80% m/m) was placed in a round-bottomed flask and hexane (250 ml) was added. Dried hydrogen chloride gas was bubbled through the mixture for 10 min at a flow rate of 150 ml min^{-1}. A heavy white precipitate was formed and was removed by filtration; it was first washed with hexane (300 ml) and finally rinsed with pentane before being dried under reduced pressure. The original reaction mixture was discarded, the apparatus was

Table 5 Carbon, hydrogen and chlorine analysis of the trimethyllead calibrant

	% Composition sample	% Composition theoretical
Carbon	12.62	12.51
Hydrogen	3.24	3.14
Chlorine	12.04	12.30

Table 4 Summary of techniques and sample intakes

Sample pre-treatment, derivatisation and separation	Final detection (Lab. code)
EDTA complexation and hydride generation ($NaBH_4$*)	Zeeman ETAAS (Lab.01)
Addition of NaOH–HCl, hexane extraction, complexation with diethyldithiocarbamate (DDTC) and addition of HCl; derivatization with propylmagnesium chloride; separation by capillary GC	Quartz furnace AAS (Lab.02)
Addition of NaOH, hexane extraction; clean-up using silica gel; derivatization with $NaBEt_4$; separation by capillary GC	AAS (Lab.03)
Addition of NaCl and NaDDTC followed by hexane extraction; derivatization with propylmagnesium chloride; separation by packed GC	Quartz furnace AAS (Lab.04)
Addition of citric acid, ammonia and EDTA; derivatization with $NaBEt_4$ followed by hexane extraction; separation by capillary GC	Microwave induced plasma AES (Lab.05)
Addition of ammonium citrate–EDTA; derivatization with $NaBEt_4$; separation by capillary GC	Microwave induced plasma AES (Lab.06)
Addition of ammonium citrate–EDTA; derivatization with propylmagnesium bromide; separation by capillary GC	ICP-MS (Lab.06)
Addition of EDTA, citric acid and DDTC followed by hexane extraction; derivatization with butylmagnesium bromide; separation by capillary GC	ICP—MS (Lab.07)
EDTA complexation; derivatization with $NaBEt_4$; separation by capillary GC	Isotope dilution ICP-MS (Lab.07)
Addition of citric acid and ammonia followed by hexane extraction; derivatization with pentylmagnesium bromide; separation by capillary GC	MS (Lab.08)
Addition of ammonium citrate and NaDDTC followed by pentane extraction; derivatization with butylmagnesium bromide; separation by capillary GC	MS (Lab.09)
Addition of sodium sulfate, hydrochloric acid and mercury(II)nitrate; removal of inorganic lead by co-precipitation with barium sulfate after addition of barium chloride; use of glassy carbon counter electrode, Ag–AgCl–KCl reference electrode and glassy carbon–mercury film working electrodes	DPSAV† (Lab.10)

* This method is only appropriate for differentiation between organic and inorganic lead and does not provide any identification between different lead species. † Differential pulse anodic stripping voltammetry.

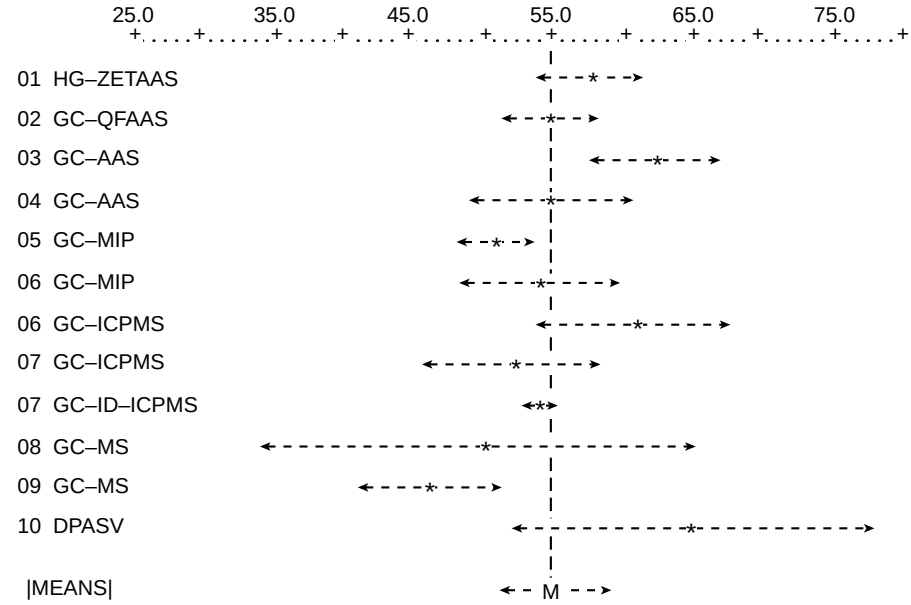

Fig. 1 Bar-graph for laboratory mean and 95% confidence interval. The results correspond to the trimethyllead content as mass fractions of $(CH_3)_3Pb^+$ (ng kg^{-1} as TriML); the mean of laboratory means obtained was (55.2 ± 3.5) ng kg^{-1} as TriML.

cleaned with hexane and a fresh preparation was undertaken in order to produce sufficient trimethyllead chloride for the purpose of the project. The purity of the product was assessed using NMR spectroscopy and was found to be > 99%. The product was also sent to an accredited external laboratory for carbon, hydrogen and chlorine analysis and the results are shown in Table 5. Good agreement was obtained between the observed percentage composition for these components and their theoretical values, providing confirmatory evidence of the purity of the product.

Technical discussion

The sets of results found acceptable after both the technical and statistical evaluation are presented in the Fig. 1. Each set of results is identified by the code number of the laboratory. The details of the analytical methods used by each laboratory are given in Table 4.

The technical discussion focused firstly on the calibration methods used by the participants. All laboratories used the TriML calibrant provided by the University of Plymouth, either for calibration or verification of their own calibrants. It was noted that some deterioration had been observed in a commercial calibrant over a 2 y period. No significant difference was observed by the laboratories between external calibration and standard addition. Some laboratories used tetraethyllead or tributyllead as internal standard.

With the exception of Lab. 01 which used a hydride generation method, Lab. 09 which applied DPASV and Lab. 10 which used HPLC, all the participants had used a GC separation following a Grignard derivatization of the analyte. Some differences were observed in terms of precision by two laboratories using the same separation and detection but different Grignard reagents; Lab. 05 had used a pentylated Grignard reagent and attributed some losses to this stage whereas Lab. 06 had used a butylation method which resulted in a cleaner reaction, thereby accounting for the discrepancy in precision. In the subsequent discussion, it was agreed that the conditions of the Grignard reaction in terms of temperature, concentration and length of the alkyl chain were key factors which require careful control. The longer the alkyl chain of the Grignard reagent, the greater the risk of degradation product formation and peak broadening in the chromatographic stage.

Conclusions

While the results obtained by the participating laboratories were in good agreement (Fig. 1) and illustrated the high quality of the measurements performed, the doubts expressed on the stability of the reference material did not encourage the SM&T programme to recommend this material for certification. It is thought that such material could be certified, providing that storage at low temperature (+4 °C or below) in the dark would be constantly maintained; in the case of the RM 604, the good results obtained over 12 months at +20 °C were obviously not sufficient since the material was shown not to be stable after 37 months.

The project has been carried out under EC contract MAT1-CT94-0071 coordinated by the University of Plymouth. The sample preparation, homogeneity and stability studies were performed by the University of Birmingham, School of Chemistry. The following laboratories participated in the certification: GALAB (Geesthacht, Germany); Institut für Chemo- und Biosensorik (Muñster, Germany); Technische Universität Wien (Austria); University of Antwerp (Wilrijk, Belgium); University of Birmingham (Birmingham, UK); University of Mainz (Mainz, Germany); University of Plymouth (Plymouth, UK); University of Umea (Umea, Sweden); Universidad de Zaragoza, Centro. Pol. Sup. de Ingenieros (Zaragoza, Spain); Vrije Universiteit Amsterdam, Institute Milieuvraagstukken (Amsterdam, The Netherlands).

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