

$[\text{RuL}_3]^{n+}$ and many $[\text{RuL}_2\text{L}']^{n+}$ complexes since the individual RuL units have near degenerate emitting energy levels. Third, the "spatially isolated optical orbital" implies that only noninteracting relaxation channels exist in the spin-triplet channel. However, *delocalized* MLCT excited states exist for the singlet manifold such that low P values, consistent with the presence of E symmetry states, are present (localized singlets also exist). Such a situation accounts for the observation that the *singlet* MLCT states have polarization properties largely determined by D_3 symmetry. Fourth, the localization occurring in the triplet manifold is fast and intrinsic, a vibronic effect. (The solvent, at

most, may play a role in the rate of intramolecular exciton hopping between chelate rings.) Finally, the emitting levels are triplet spin levels that behave as the spin states in a typical aromatic heterocycle, so the spacing of the three spin states is $\sim 0.1 \text{ cm}^{-1}$, indicating a very small spin-orbit interaction. Slow spin-lattice relaxation results in the failure of these spin states to equilibrate at low temperatures ($T < 20 \text{ K}$).

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Lead in Petrol. The Isotopic Lead Experiment

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Introduction

When cars were first introduced, fuel petrols were obtained by distillation of crude oil. These consisted of hydrocarbons formed over thousands and thousands of years and were composed of alkanes, mainly normal, some aromatics, and no olefins. The octane number (ON), which defines a scale to evaluate the antiknock quality of petrol, was low (around 60), as was the compression ratio of the engines, while the fuel consumption was high. Together with improvements in engines, new conversion processes have led to the production of relatively low cost fuels with ON values increased to 85-90. The discovery of the performance of lead alkyls, $\text{Pb}(\text{C}_2\text{H}_5)_2$ and/or $\text{Pb}(\text{CH}_3)_2$, allowed a further increase of ON (98-100) when lead was added to petrols at the level of 0.6-0.8 g/L. Therefore around 1970 the most favored refining processes were those leading to the isoparaffins, which had a high susceptibility to lead, and to aromatics to produce petrols with high ON at relatively low cost.

Starting from 1960, under pressure from ecologists, studies were undertaken all over the world to evaluate the impact of the gaseous emissions (CO , NO_x , HC, etc.) and of airborne lead particulates on the environment and on humans. However, the uncertainties regarding

the possible effects of hydrocarbon emissions were so great that a chemical characterization of the individual hydrocarbons was necessary. Vehicle exhaust emissions were found to consist of about 1000 different chemical compounds, most of which are or may be assumed to be injurious to human health. There is probably no harmless level of emission and exposure for these substances, lead included. This means that the risk can be reduced only by lowering the emission levels.

Many studies were dedicated to the evaluation of the impact of automotive lead on the environment and to the assessment of its absorption in the human population. They can be subdivided into two groups, those based on changes of air and blood lead concentrations¹⁻⁷ and those based on changes of air and blood lead isotopic compositions.⁸⁻¹²

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Sergio Facchetti was born on December 5, 1931, in Borgo Ticino, Novara, Italy. He received a Doctorate in chemistry in 1955 at the University of Pavia. During 1957-1959, he was engaged in research in the field of structural analysis of mineral oils at Milan Polytechnic. Since 1959 he has worked at the Ispra Establishment of the European Communities. Since 1967 he has taught mass spectrometry and instrumental techniques in many Italian universities, at present at Sassari University, and has been course coordinator of Ispra-Courses on analytical techniques. He has been active in the Italian Chemical Society. He was Chairman of the VII International Mass Spectrometry Conference (Florence, 1976). He is a member of the Editorial Board of the journal *Biomedical & Environmental Mass Spectrometry*. He has been deeply involved in the Safeguards activity and in the identification of environmental pollutants at trace levels. Of particular significance are the environmental studies carried out in connection with the Seveso disaster and the Isotopic Lead Experiment (ILE project, 1974-1985) to determine the automotive lead contribution to total lead in human blood and to airborne lead and to follow the pathway of lead through the environmental compartments.

According to various authors,¹³⁻¹⁶ 50-66% of the lead added to petrol is mobilized in the atmosphere, while most of the remainder adheres to the walls of the exhaust system from which it is expelled by mechanical and thermal shocks in the forms of easily sedimented particles. The fraction directly emitted by engine exhaust fumes is found in the form of fine particles, which can be transferred a long way from the emitting sources.

However important the contribution of petrol lead to the total airborne lead may be, our knowledge does not permit a straightforward calculation of the percentage of petrol lead in total blood lead, which of course can also originate from other sources (e.g., industrial, natural). To evaluate this percentage in 1973, the idea of the Isotopic Lead Experiment (ILE project)¹⁰⁻¹² was conceived to label, on a regional scale, petrol with a nonradioactive lead of an isotopic composition sufficiently different from that of background lead and sufficiently stable in time. This Account summarizes the main results obtained by the ILE project.

Definition of the ILE Project

The objectives of the ILE project were (1) to determine the contribution of petrol lead to total airborne lead in urban and rural areas, (2) to determine the contribution of petrol lead to blood lead in various population groups, and (3) to follow the pathway of petrol lead through some environmental compartments.

The region of Piedmont was chosen as a test site. It lies in the Po valley (Valpadana) in the northwest of Italy. It is bordered to the north, west and south by the Alpine arc, resulting in generally low wind speeds or even stagnant air. Piedmont is a highly industrialized area with heavy traffic, and its main city, Turin, had in 1977 the highest cars/inhabitants ratio (0.4) of any city in Europe. The countryside under test is mostly located north of Turin, and it is mainly characterized by agricultural activities, by small textile and engineering industries, and, at least in the zone close to Turin, also by a high traffic density. Therefore for data evaluation the rural area was subdivided into two zones comprising localities of distances less than and more than 25 km from Turin, respectively.

It is worth remembering that the ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb isotopes are the final products of radioactive decays of ²³⁸U, ²³⁵U, and ²³²Th, respectively, and their isotopic abundances depend on the abundances of their parents, on their radioactive decay, and on the age of the deposits. A preliminary sampling of the environment showed that the ²⁰⁶Pb/²⁰⁷Pb isotope ratios in all

types of samples were within the range 1.16-1.20. Analysis of lead samples from various mines throughout the world that usually supply Europe showed that only Australian lead from Broken Hill mine had an isotopic composition (²⁰⁶Pb/²⁰⁷Pb = 1.04) distinctly different from that of the lead encountered in the environment of the test area. Furthermore, the amount of lead that might be imported from Australia was sufficiently large to cover the requirements for a long-term field experiment. Therefore from 1975 to 1979 the petrol lead distributed in the region of Piedmont, which formerly had a ²⁰⁶Pb/²⁰⁷Pb isotope ratio of around 1.19, was substituted by lead from the Australian Broken Hill mine (²⁰⁶Pb/²⁰⁷Pb = 1.04).

The project was subdivided into four periods according to the time schedule of the petrol lead substitution:

phase 0	background definition	July 1974-July 1985
phase 1	transition period from the initial 1.19 ratio lead to the Australian 1.04 ratio lead in petrol	August 1975-April 1977
phase 2	about 90% Australian lead as additive in petrol	May 1977-December 1979
phase 3	gradual, uncontrolled switch back to the initial lead in petrol	January 1980-September 1985

Throughout most of the period of the experiment, the lead concentration in petrol was constant at about 0.6 g/L; only from July 1981 on was it reduced to about 0.4 g/L.

To evaluate the contribution of petrol lead to total airborne lead and to blood lead level in various population groups, only the variations of isotope ratios ²⁰⁶Pb/²⁰⁷Pb are required, because the determination of lead concentration is necessary only if the exposure does not remain constant. However, for control purposes as well as for a better picture of the whole study, the total lead concentrations in both airborne particulate and blood samples were systematically determined.

Experimental

Sampling and sample preparation were performed under stringent conditions in order to reduce contamination effects.

The total lead determinations in airborne particulate were carried out in three laboratories by X-ray fluorescence (XRF) and/or by atomic absorption (AA) techniques. However, values near or below the limit of sensitivity of XRF (<3 μg of Pb total) were always checked by AA. About 17% of the samples were double-checked, approximately 70% by XRF and 30% by AA. The results obtained showed that 80% of the samples with lead concentrations lower than 2 μg/m³ were within V = ±10% from the mean and 94% within ±20%; similarly the samples higher than 2 μg/m³ were 86% within ±10% and 99% within ±20%.

The total lead concentration in blood was measured in at least two laboratories by anodic stripping voltammetry, flame atomic absorption (extraction, Delves cup), and flameless atomic absorption. For the latter technique, two kinds of sampling were made, blood on paper disc (punched disc technique) and liquid blood, while for the other techniques only liquid blood was used. The blood samples were analyzed at monthly intervals,

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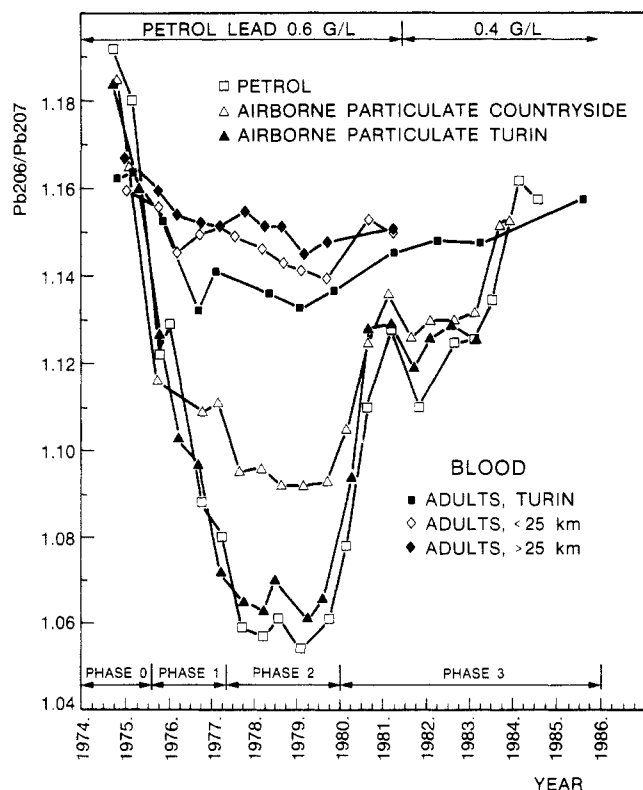


Figure 1. Change in $^{206}\text{Pb}/^{207}\text{Pb}$ ratios in petrol, airborne particulate, and blood from 1974 to 1985.

and the laboratories were requested to perform analysis not affected by bias within the range of 10 ± 3 and $60 \pm 5 \mu\text{g}/\text{dL}$. It is worth mentioning that no statistically significant difference was observed between blood samples analyzed immediately after sampling and blood samples stored in a refrigerator for 6 weeks.

The lead isotopic compositions were determined by single-focusing mass spectrometers using the surface (thermal) ionization technique. The high accuracy needed in the mass spectrometric measurements makes the problem of contamination of samples by extraneous material a very important one. Therefore, the samples were prepared in a clean laboratory (class 100) using exclusively containers made in Teflon FEP and quartz, and reagents purified by the subboiling distillation technique.

Continuous internal and external quality assurance programs for isotopic composition of lead in airborne particulate, blood, vegetation, and soil samples were also established. In particular for blood analyses, the most difficult ones, the randomly selected samples were coded and their aliquots distributed to the JRC (Ispra), the Free University of Brussels, and the NBS (Gaithersburg, MD) mass spectrometry laboratories. The entire set of values showed that the data was constantly good throughout the ILE project. The mean differences in the $^{206}\text{Pb}/^{207}\text{Pb}$ ratios (range of values 1.0490–1.1668) were always smaller than 0.0005 measured in the double blind samples by the same or another laboratory. The results refer to complete analytical procedures inclusive of chemical sample treatments and instrumental measurements.

Results and Discussion

The behavior of the isotopic ratio in air, on a monthly basis as the average of all the sampling stations both

Table I
Fractional Contribution of Local Petrol Lead to Atmospheric Lead in Piedmont

source of results	Turin	countryside	
	downtown	<25 km	>25 km
means of phase 0 and 2	0.873	0.595	0.579
least-squares regression semiannual means	0.834	0.594	0.605
nonparametric regression semiannual means	0.875	0.536	0.597

in Turin and in the countryside, shows that the isotopic ratio follows that in petrol more closely in the city than in the countryside (Figure 1). In fact, the isotopic ratio in samples from rural localities during phase 2 is significantly different from that in petrol. As the concentration levels over 24 h in rural areas (yearly average of $0.28\text{--}0.56 \mu\text{g}/\text{m}^3$ during the period 1974–1981) are much lower than in the city (yearly average value of $2.0 \mu\text{g}/\text{m}^3$ during the period 1974–1981), sources other than lead from local motor traffic may make a relatively high contribution. In fact, in the rural areas, there may be a significant contribution from suspension in the atmosphere of soil dust and chemical products used in agriculture and from transport from other zones of air masses with lead of different isotopic ratios than that of local petrol origin.

The fractional contribution of local petrol lead to total airborne lead during phase 2 was evaluated by using two approaches. The first approach is based on a system of equations referring to different times at equilibrium conditions, e.g., the equations $R_1X + f(I - X) = R'$ and $R_2X + f(I - X) = R''$ relate the mean airborne lead (R' and R'') and the mean petrol lead (R_1 and R_2) isotopic ratios of phases 0 and 2, respectively. X is the fraction of local petrol lead in airborne particulate due to petrols affected by the change in the lead isotopic ratio. The term $(I - X)$ represents the fraction of the sum of all other external sources of lead in the airborne particulate, factor f being the unknown isotopic ratio of the mixture of these sources. It is assumed that X and f remained constant over the period of the experiment, which implies a reasonable constancy of the lead-contributing sources in the test areas.

The second approach makes use of the least-squares and the nonparametric linear regressions of the semiannual airborne lead isotopic ratio on the semiannual local petrol lead isotopic ratio up to the first half of 1981. The results are given in Table I. They mean that during phase 2 the airborne lead in downtown Turin, congested by traffic, contained about 87% of Australian lead, i.e., about $1.7 \mu\text{g}/\text{m}^3$, and in the rural areas about 59%, i.e., about $0.33 \mu\text{g}/\text{m}^3$ in the countryside zone within 25 km from Turin and about $0.18 \mu\text{g}/\text{m}^3$ in the further than 25 km zone.

The estimate of the contribution of petrol lead to blood lead was based on measurements either on subjects sampled during the main phases 0 and 2 (follow-up subjects) or on subjects sampled just once or at times different than phase 0 or phase 2 or both. The totality of the results of the two categories is referred to as the general sample. For the estimation of petrol lead contribution to blood lead, three approaches, quite similar to those followed for airborne lead, were used. The first approach takes into account the initial (phase 0) and final (phase 2) blood lead isotopic ratios of followed-up

Table II
Fractional Contribution of Local Petrol Lead to Blood Lead
in Adult Subjects in Piedmont^a

source of results	Turin	countryside	
		<25 km	>25 km
follow-up mean values	0.236 (27)	0.121 (32)	0.095 (11)
follow-up median values	0.239 (27)	0.119 (32)	0.080 (11)
means of phases 0 and 2 of general sample	0.233 (128) ⁺	0.125 (121) ⁺⁺	0.089 (62) ⁺⁺⁺
nonparametric regression semiannual means	0.219 (179)	0.111 (185)	0.081 (87)
nonparametric regression semiannual means	0.226 (179)	0.106 (185)	0.076 (87)

^a Figures in parentheses are the number of individuals monitored; +, 44 subjects in phase 0 and 84 in phase 2; ++, 30 subjects in phase 0 and 91 in phase 2; +++, 3 subjects in phase 0 and 59 in phase 2.

subjects and the petrol lead isotopic ratios measured concurrently. Conditions of equilibrium may be considered to hold for total blood lead concentration because the individual variations were not statistically significant.

The second approach considers the mean blood lead isotopic ratios in phase 0 and 2 of the adult general sample and the mean petrol lead isotopic ratios.

The third approach takes into account the least-squares and the nonparametric linear regressions of the adult semiannual blood lead isotopic ratio on the semiannual local petrol lead isotopic ratio. The regressions were limited to data collected up to the first half of 1981, i.e., until the time when the lead concentration in petrol was constant at ca. 0.6 g/L. The results are shown in Table II.

Consequently, the contribution of petrol lead to the blood lead of adults can be estimated at around 24% (i.e., about 5.15 $\mu\text{g}/\text{dL}$) in Turin, about 12% (i.e., 2.75 $\mu\text{g}/\text{dL}$) in the rural area of less than 25 km, and about 8% (i.e., 2.25 $\mu\text{g}/\text{dL}$) for the area beyond. Their approximate 99% confidence limits are 4.09–6.19, 1.55–3.78, and 0.08–7.09 $\mu\text{g}/\text{dL}$, respectively.

These estimations take into consideration both the ingested and the inhaled local petrol lead. In fact, other factors, in particular diet, might overshadow the positive association usually found between blood lead and air lead. For instance, the determination of lead concentration in Piedmont wines produced from 1970 to 1983 gave a mean value of 155 $\mu\text{g}/\text{L}$ with a standard deviation of ± 67 $\mu\text{g}/\text{L}$. The $^{206}\text{Pb}/^{207}\text{Pb}$ ratio was 1.1642 ± 0.0031 ($n = 13$) in phase 0, 1.1558 ± 0.0041 ($n = 15$) in phase 2, and 1.1608 ± 0.0057 ($n = 26$) in phase 3. The data of phases 0 and 2 provide a contribution of local petrol lead to wine lead of ca. 7% or, considering the wine mean lead content of 155 $\mu\text{g}/\text{L}$, of about 10 $\mu\text{g}/\text{L}$. As a daily wine consumption of about 0.5–1 L/day might be reasonable for rural drinkers, the importance of wine as a lead source becomes evident.

The values obtained through the ILE could be considered high or low according to the different aspects that are considered. We list, therefore, the conditions that may influence our findings.

Conditions that contribute to high values: (1) The lead concentration in petrol was equal to ca. 0.6 g/L. (2) The test site was characterized by a low-intensity

atmospheric circulation for about 60% of the time. Calm periods are characterized by strong inversions and multiple stratification, which impede vertical mixing. (3) The city of Turin had the highest cars/inhabitants ratio in Europe (0.4 in 1977), and its traffic volume corresponded to 5.6% of that of the whole of Italy.

Conditions that contribute to low values: (1) The continuous exchange between the blood lead and the accessible bone lead due to the remodeling of bones. Presumably at the start of the ILE project the bones released lead of the same isotopic composition as that of the blood lead. Progressively the blood isotopic ratio continuously decreased, paralleled by a decrease of the lead ratio in the accessible parts of the bones. With time the isotopic ratio of blood lead and of the accessible bone lead were likely to converge continuously in an asymptotic manner. Since the time scale over which this occurs may be of the order of years, only very long term experiments could give definitive results. Therefore, the blood lead isotopic values measured at the end of phase 2 could not represent the maximum decrease in isotopic ratio. In fact, there was no full evidence that equilibrium was reached.

The lead isotopic determinations can also give information on the uptake, transport and accumulation of airborne lead in plants under natural conditions.

Plants take up lead from soil and directly from the atmosphere. Furthermore, lead can be taken up from moist dead plant materials at the air–soil–plant interface.¹⁷ This is supported by the fact that lead accumulates near the soil surface. Referring to absorption by roots, the binding and exchange capacity of soils is important in determining lead availability to plants as the plant uptake is probably related to the soluble fraction rather than to the total lead in soils. As far as aerosol deposition is concerned, this may be dependent on the characteristics of the leaf surface, with a possible difference in deposition and absorption between rough pubescent leaves and old smooth waxy leaves of the same tree. In a preliminary survey, parts of trees grown in Turin and Viù (countryside zone further than 25 km from Turin) gathered in spring and the following autumn were examined, in particular, trees with broad leaves (horse chestnut) and with medium leaves (poplar and prunus) and, finally, shrubs with small leaves (privet).

The data show a rather close correspondence between the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio in leaves and in air and consequently the importance of the direct air–plant deposition process. For horse chestnut growing at Viù, the isotopic lead ratio in leaves differs more in spring than in autumn from that in airborne lead. This may be attributed to the contribution of the rising sap being greater than the fallout, while in autumn the opposite is true. It is also interesting to observe that both in Turin and Viù in spring the petioles have a higher isotopic ratio than the leaves. This is probably due to a lower contribution of fallout because of their lower surface/volume ratio.

Parts of plants exposed to the atmosphere (bark, husk of fruit), have a lower isotopic ratio than the nonexposed parts (cortex, wood, chestnut), sometimes with

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a gradient toward the center of the stem. Therefore the following tentative hypothesis can be formulated: (1) a lead translocation from roots to the new parts of the plants; (2) a mobility of lead, absorbed through the leaves in the preceding years, from the old parts toward the new ones. However, before drawing definite conclusions, it is necessary to consider that soil types and/or plant species may have a determining effect on the quantity and origin of lead taken up by the plant. Among the most important parameters are the soil pH, cationic exchange capacity, and organic material content, as well as the age and species of the plant.

In conclusion, it might be observed that in the solution of problems related to exposure assessment mass spectrometry plays an important role through precise isotope ratio determinations. As human populations can be significantly exposed to lead in air, food, water, and dust, current blood levels of populations in industrialized societies reflect this impact of human activities. Thus the isotopic tracing of lead from petrol, a source of major significance, to the atmosphere to human blood of exposed populations has allowed estimation of the size of the atmospheric lead contribution to human lead levels. To this purpose two field investigations have been carried out, the first in Turin, Italy (the ILE experiment here described), and the second in Dallas, TX.⁸⁻⁹ The latter study has used existing natural shifts in isotopic proportions, and the blood lead isotopic composition of a group of subjects was monitored for comparison with the isotopic composition of the air they breathed. If the differences between the regions—much higher lead levels in air and blood in Turin—are taken into account, the estimates of the two studies are consistent for the direct inhalation contribution.

Another significant experiment, always based on stable lead isotopes, was carried out by Rabinowitz and co-workers¹⁸⁻²¹ on five adult male volunteers housed in

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the metabolic research wards of the Sepulveda and Wadsworth VA hospitals in Los Angeles. They were given low-lead diets with controlled lead content, supplemented by tracer lead salts at different times. As in other studies based on lead concentration determinations, the fraction of ingested stable isotope lead tracers absorbed into the blood was much lower when lead was consumed with meals ($10 \pm 2.2\%$) than between meals ($35 \pm 13\%$). This implies greater significance of lead ingestion from leaded paint and from dust and soil when consumed between meals. Inhalation slopes were also estimated, and even if they resulted as the lowest that can be reasonably derived from the experiment, this was probably due to a smaller time-averaged air lead exposure.

It can therefore be claimed that, because of the capability of mass spectrometry to trace a lead source, a more important role of this technique can be expected not only in environmental studies but also in biomedical studies, regulatory activities, and criteria assessment, not limited to lead but extended to the majority of the elements.

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