Air Borne Lead Contamination of Trees in Metro Manila—A Study of Environmental Hazards Related to Leaded Fuels

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The environmental hazards originating from the use of leaded fuel in Metro Manila were analyzed indirectly by investigating the Pb content and contamination of leaves of trees exposed to heavy traffic along Taft Avenue in comparison with three sites with less or no traffic in the countryside. Both the internal Pb content and the external Pb load of plants were measured. Internal Pb content of samples along Taft Avenue and the countrysides yielded an average of 6.5 mg and 0.6 mg Pb per kg dry weight respectively while that of external Pb load were 18.9 mg and 2.74 mg Pb per kg dry weight respectively. These values from the two sampling areas were significantly different. The impact of Pb pollution was discussed in terms of phytotoxicity of Pb to plants and possible effects on the food chain, and consequently, on human health.

INTRODUCTION

Lead is a heavy metal that induces growth inhibition in most plants when exposed to high concentrations[16]. According to the WHO Report (1977), Pb in human blood should not exceed 100 μg Pb/L and 200 μg Pb/L in children and adults, respectively. Thus, cumulative Pb pollution may cause mental disturbances in children at concentrations lower than those which do not yield clinical symptoms of toxicity in adults.

There are two main sources of Pb pollution: soil and atmosphere; the contribution from water is generally less. The soil acts as a sink for Pb from the air. Lead in the soil becomes available to plants via absorption from the soil solution. In addition, Pb from the atmosphere can also deposit on leaf surfaces. Thus the internal Pb content of plants, obtained via absorption, and the external Pb load, via deposition, can both serve as bioindicators of the presence of Pb in the air. The total Pb content of plants (internal plus externally adsorbed Pb) enters the food chain, as plant material was eaten by animals and humans.

Pb has been used for a long time as an antiknock agent in fuels. Therefore the largest anthropogenic Pb source is the exhaust gas of motor vehicles, especially in big cities. Due to its health risk, the use of leaded fuel has been banned in Western industrialized countries since 1972. In Germany, for example, this policy reduced the airborne Pb pollution from 1980 to 1995 by about 70 %. However, there is a considerable lag in this respect in developing countries.

In the Philippines, a total emission of 407.3 tons of Pb per year from motor vehicles in Metro Manila was reported (The Philippine Environmental Quality Report 1990-1995[1]) as compared with the 240 t Pb per year for whole Germany. Of these emissions in Manila, 49 % originate from cars and 36 % from public utility vehicles (PUV), whereas bus traffic produces only 0.06 %[1]. Pb concentration in the air (mean monthly of 24 h monitoring) reaches values up to 1.2 μg Pb / m^3. These are high values, but still they are slightly under the Air Quality Standard of the Philippines of 1.5 μg Pb / m^3.

The German and US Air Quality Standards for Pb are lower. Pb concentration in big cities in Germany and the US scatter around 0.5 μg Pb/m^3, for Tokyo a value of 0.65 μg Pb/m^3 has been published[14]. Unpolluted continental air in the countryside has a Pb content in the range of about 0.01 μg Pb/m^3; similar values can be expected for rural sites in the Philippines. In the centre of big European cities the corresponding value was in the order of maximal 0.5 μg Pb/m^3 air at times when the Pb content of fuel was in the order of 400-500 μg Pb per litre fuel (before 1971). When the amount of the organic lead-containing antiknock agents in the fuel decreased stepwise to 150 μg Pb/l and finally to zero, a continuous concomitant decrease was observed.

Studies on Pb contamination of soils and plants from pollution of air started between 1960 and 1962 in the UK[3] and the USA[4,5,6]. Results can be summarised as follows[2]: Pb contamination of plants and soils along a strip between 15 and 150 m on both sides of roads is significantly higher than the corresponding values far away from roads. The gradient decreased exponentially. These early reports were later confirmed in other countries, mainly in Europe[2], Australia[9,10] and Asia[7,8].

The present study aims to describe the gradients of Pb contamination of plants obtained within the center of Metro Manila with high traffic density and from rural areas with low traffic density in the Philippines. Trees will be used as convenient model systems, although some may not necessarily be typical food plants.

MATERIALS AND METHODS

Sites of sampling. Collections were done from four different sites in Luzon Island, Philippines. The traffic situation in each sampling site is described as follows:

Site 1: Taft Avenue from De La Salle University Campus to Luneta Park, Ayala Avenue. The annual average daily traffic along the sampling points in this area are as follows[14]: (a) Between Vito Cruz and Quirino Avenue, 36,185; (b) Between Pres. Quirino and United Nations (U. N.) Avenue, 49,320; and (c) Between U. N. Avenue and Claro M. Recto, 64,960. The numbers were attributed to cars, jeepneys, buses, and trucks passing Taft Avenue per 24 h. Samples were collected from trees growing along Taft Avenue on April 4, 2000.

Site 2: Highway in Botolan, province of Zambales, Central Luzon. A bus regularly passed thru the highway 14 times per day. Additional trips were made depending on the number of passengers. This number excludes cars, jeepneys, and mini-buses. The trees were found along the highway. Samples were collected along this site on September 3, 2000.

Site 3: Town centre of Sta. Cruz, province of Zambales, Central Luzon. In this sampling area, about 700 tricycles and 60 jeepneys plied the route. There were also 47 trips to various destinations made by minibuses. A leading bus company regularly makes 14 trips/day to and from this area. Trees were growing along the town's plaza. In this site, samples were collected on September 3, 2000.

Site 4: Mount Pinatubo crater, Tarlac, Central Luzon. The elevation was 989.94 m. (3,248 ft.). The area was inaccessible to any form of vehicles. Samples were collected in this site on May 2, 2000.

Table 1. Tree species investigated in this study, common names, and sites of collection. Gray underlay: Food plants.

Species	*Common Philippine Name*	*Sites*
Acacia auriculiformis	Acacia	1, 2, 3
Annona squamosa	Atis	1, 3
Azadirachta indica	Neem	1
Bambusa sp.	Bamboo	2
Bauhinia malabarica	Alibangbang	1, 2
Chrysophyllum caimito	Kaimito	1, 3
Diospyros philippensis	Mabolo	1, 2
Ficus benjamina	Balete	1, 3
Ficus elastica	Rubber tree	1
Ficus lyrata	Fiddle-leaved fig	1
Ficus religiosa	Bo tree	1
Lagerstroemia speciosa	Banaba	1, 2
Mangifera sp.	Mango	1, 2
Muntingia calabura	Datiles	1, 3
Nauclea orientalis	Bankal	1
Phyllostachis nigra	Bamboo	1
Pisonia alba	Lettuce tree	1,2
Plumeria obtusa	Kalachuchi tree	1
Polyalthia longifolia	Indian tree	1, 3
Polyscias sp.	Papua	1
Psidium guajava	Guava	2
Pterocarpus indica	Narra	1
Ptychosperma macarthurii	Palm	1
Samanea saman	Rain tree	1, 3
Sandoricum koetjapa	Santol	1
Swientenia macrophylla	Mahogany	1
Tamarindus indica	Tamarind	1, 3
Terminalia catappa	Talisay	1, 3
Thevetia peruviana	Campanilla	1
Trema orientalis	Anabiong	4
Zisiphus jujuba	Manzanitas	2

Sampling of Plants and Soils.

Mature leaves were collected from the different plant species listed in Table 1. Nine of these plants have edible fruits. Samples were gathered from a height of 1-3 m.

Soil samples were collected from two representative sites in the Philippines for comparison with representative samples obtained from other countries (Table 2). Soils were collected using a steel trowel. The depth of collection was 0.2 m.

Preparation of materials for analysis.

Two sets of leaf samples for each plant species were prepared for analysis: washed and unwashed. The surfaces of washed leaves were thoroughly cleaned with running water before drying. Both sets of leaves were dried (75° C, 24 h) and were homogenized to powder in a mortar. The dry powder was stored in Eppendorf cups in a dessicator until analysis. Soil samples were pulverized and oven dried.

Analytical methods. The powdered leaf samples and pulverized soil samples were subjected to pressurized extraction with 65 % HNO_3 (170° C, 10 h). Lead contents of both leaf (100 mg aliquots) and soil samples were determined by ICP-AES emission spectroscopy (ICP Jobin 70 plus, ISA GmbH, Grasbrunn, Germany). Data were calculated as mg Pb per kg dry weight of leaves or soil. The differences of the Pb content of unwashed and washed leaves were taken as "*Pb load*" or "*Pb contamination*" of leaves by air. The standard references used in this study were CRM 100 and CRM 277 of the Commission of the European Communities (Community Bureau of Reference). The detection limit of this instrument for lead was 10 ppb.

Statistics. The statistical analysis was carried out using the STATISTICA program (V. 5.5., Statsoft, Tulsa, OK, USA). Results were expressed as means with standard deviations (S.D.). ANOVA was performed with log10-transformed data for significance tests. Post hoc pair wise comparisons were done using Duncan's test (washed *versus* unwashed leaves).

RESULTS

Pb content of soils

Arable lands contain in general Pb concentrations between 0.1 and 20 mg Pb kg^{-1} dry matter. Table 2 shows that garden soil from Metro Manila has a much higher

Table 2. Pb soil content at two sites in the Philippines. Reference samples: Unpolluted sites in India and Germany and a highly polluted site (mono fill) in Germany.

Site	Pb content of soil (mg/kg D.W.)
Close to Mount Pinatubo	2-3
Average for surface of earth[2]	13
Sand, Germany	10-14
Tropical soils close to Varanasi, India	16-21
Arable soils close to Würzburg, Germany	18
Garden soil, Metro Manila, Philippines	28
Soil Quality Standard, Germany	100
Slag of a mono fill, Hopferstadt, Germany (Highly polluted site)	530

Pb content than soil from Mount Pinatubo. It is also higher than comparable soils in other regions of the world, but still below when compared with the acceptable standard in Germany. Typical off-mine sites, mono fills and other industrial sites have much higher Pb contents. The German soil quality standard for Pb for land used in agriculture is 100mg/kg D. W. (Table 2).

Pb content of leaves

In Figure 1, the average Pb content of carefully washed leaves of trees from the 4 different sites is shown. These values were indicative of the uptake of Pb from the soil. Leaves of trees grown in Metro Manila exhibited the highest Pb content as compared with the 3 other sites: Botolan, Sta. Cruz, and Mount Pinatubo. The last three sites were comparable with each other.

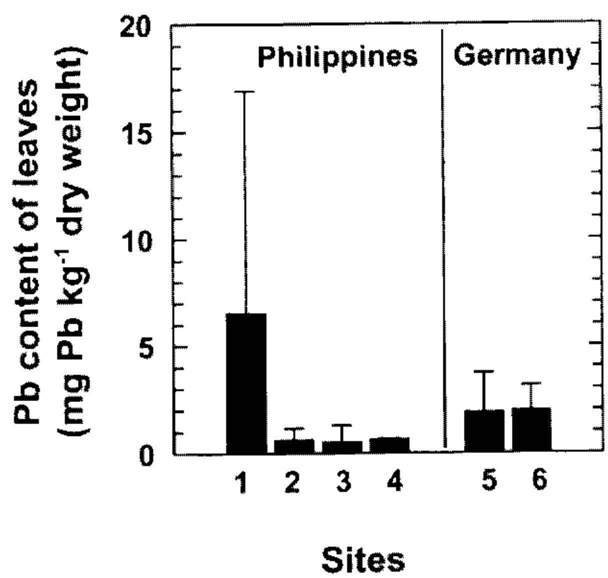

Figure 1. Pb content of washed leaves of shrubs and trees from 4 sites in the Philippines(1-4) and two sites in Germany (5,6). Site 1: Manila; site 2: Botolan, Zambales; site 3: Sta. Cruz, Zambales; site 4: Mount Pinatubo, Tarlac; site 5: Hopferstadt; site 6: Slag mono fill Hoferstadt (highly polluted site).

The difference in the lead content obtained from the unwashed and washed leaves gives the value of airborne Pb contamination on leaf surfaces, also referred to in the paper as external lead load. This amount originated preferentially from the burning of the organic Pb containing antiknock compounds in the engines of motor vehicles (Fig. 2).

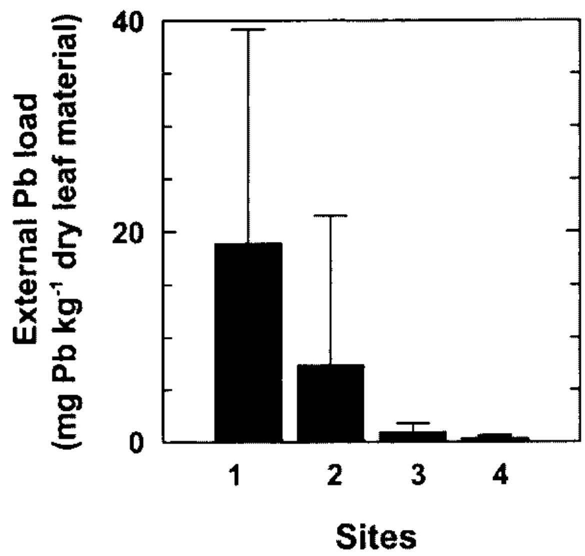

Figure 2. External Pb load on the surface of leaves of trees grown at four different sites in the Philippines (see fig. 1).

In Fig. 2, the average level of Pb load is presented. It shows a similar trend of lead content as that of the washed samples, that is, site 1 reflecting the highest value as compared with the 3 other sites. However, site 2 is slightly higher as compared with sites 3 and 4. The Pb load is very high in Manila, exceeding the Pb content of washed leaves by a factor of about 3. The Pb load in Botolan was lower than that in Manila, but the load exceeded the internal content by a factor of almost 12. The sites in Sta. Cruz and in the region of Mount Pinatubo exhibited a low external Pb load, amounting to factors of 0.4 - 1.2 in comparison to the internal Pb content.

Statistical analysis (ANOVA) revealed (Table 3) a highly significant difference in mean Pb content among sites. A significant difference was also observed in the lead load.

Table 3. ANOVA of site effect on Pb content and Pb contamination. $p<0.001$ highly significant . df = degrees of freedom; F: F-statistics.

Parameter	Error	df	F	p
Pb content	26.0	2	48.8	0.0000
Pb load	18.8	2	34.8	0.0000

Table 4. Post hoc comparisons (Duncan Test) of the Pb leaf content and Pb leaf contamination at the surface. $p<0.05$ significant, $P<0.001$ highly significant. Statistics compare table 3.

Pairs of comparison: level of significance (p)	Pb content of washed leaves	Pb contamination at the leaf surface
Manila *vs*. Botolan	0.0001	0.0001
Manila *vs*. Sta. Cruz	0.0001	0.0001
Botolan *vs*. Sta. Cruz	0.4349	0.0923

Post-hoc pair-wise comparisons of lead content and lead load is shown in Table 4. For the lead content, the pairs Manila *vs*. Botolan and Manila *vs*. Sta. Cruz exhibited highly significant difference, whereas pair Botolan vs. Sta. Cruz showed no significant difference. Data indicated that the soil in Metro Manila was polluted by lead. Consequently, the average Pb content of leaves of trees was higher in Manila than at the other sites. In a previous study the Pb content of leaves of 29 shrubs and trees growing on non-polluted sites close to Würzburg (Germany) was analyzed. An average of 1.8 mg Pb kg^{-1} dry leaf material was observed (Fig. 1). These values matched very well the values from the countryside in Luzon.

For the Pb load, pairs Manila *vs*. Botolan and Manila *vs*. Sta. Cruz (Table 4) also showed significant differences similar to Pb content. In contrast to the Pb content, for the Pb load the pair Botolan *vs*. Sta. Cruz exhibited marginal significance.

DISCUSSION

Based from the results in Fig. 1 and Fig. 2, the very high internal Pb content and external Pb load of trees in Metro Manila are indicative of high Pb concentration in the soil and air, respectively. In a work done by the authors in Wurzburg, Germany, the lead content of leaves of 29 shrubs and trees growing on nonpolluted sites were analyzed to serve as reference point as to the level of lead content. An average of 1.8 mg Pb kg^{-1} dry leaf material (Fig. 1) was observed. These reference values are comparable with the values obtained from sites 2, 3, and 4, which are located in less polluted sites outside Metro Manila. However, the absolute Pb content of leaves of the trees grown in Manila was high in comparison to the corresponding values of a heavily polluted site in Germany (Pb content of soil: 530 mg kg^{-1} dry soil), where for 15 species, an average leaf content of about 2 mg kg^{-1} dry leaf material was measured.

Results also indicate that there was almost no contamination of the soil in Botolan as compared to the situation in Manila. However, results in Fig. 2 may also indicate possible higher air-borne Pb level. This could be attributed to the continued use of leaded fuel. Although unleaded fuel had been introduced in the local market in February, 1994, 90% of its total sales was accounted for in Metro Manila[15]. The number of tricycles, jeepneys, mini-buses, and buses that regularly plied the route in Botolan and Sta. Cruz represented only about 0.5% that of Manila. At the different sampling points in Manila, the average total number of vehicles that passed along these routes per day was 150, 465[14]. In addition, it is also reasonable to assume that the high level of lead both in air and soil is due to accumulation of lead over time as traffic condition also worsens with time. With time, Pb air pollution will also strongly contaminate soil which acts as sinks for aerosolic Pb (source). Massive traffic existed in Manila much longer times before it spread, by the highways, to the countryside.

Phytotoxicity of Pb

As yet, the lead content of leaves obtained in this study (6-19 mg Pb per kg^{-1} dry leaf) is not within the lead toxicity limit of 30 and 300 mg Pb per kg^{-1} dry leaf material as reported[12]. Supportive of this, in a previous study[13], 12 species of trees sampled within the immediate vicinity of Taft Avenue (i.e. Site 1a) was measured for their photosynthetic capacity expressed as quantum yield of photosystem II (Y) as a function of Pb content (Fig. 3). The average value obtained for Y is 0.717 + 0.025 whereas for a total of 70 various Philippine plants, an average of 0.723 was measured (unpublished data).

In a similar study conducted in Germany, a Y of 0.790 + 0.029 for 27 woody control plants was measured, whereas 14 shrubs and trees grown on a site polluted with Pb exhibited a quantum yield Y of 0.740 + 55. Statistical analyses demonstrated that these photosynthetic capacities were not significantly different.

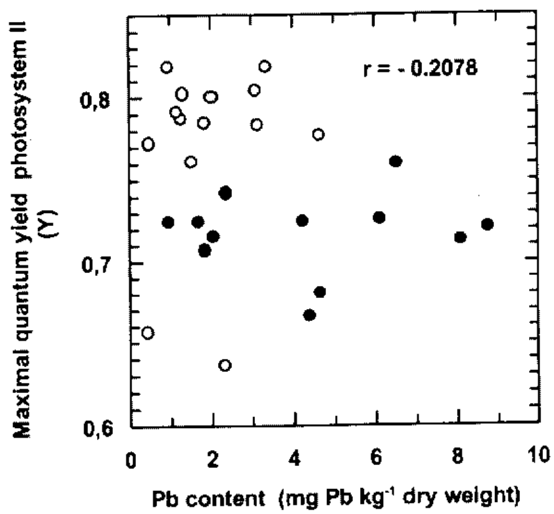

Figure 3. Maximal quantum yield of photosystem II (dark adapted plants) of trees growing in the immediate vicinity of Taft Avenue (site 1a, filled symbols) and of shrubs and trees grown on a slag mono fill in Hopferstadt (Germany) with high Pb in the soil (open symbols) as function of the Pb content of leaves. The correlation coefficient r indicates only a slight negative correlation between both parameters. The level of significance was p = 0.195 for the total set of data (= non significant). For the German site p was 0.304, for the Manila site 0.889 (N.S.).

Leaves are a twofold sink for the Pb emission. Part of the airborne lead of the atmosphere enters directly the soil and plants take up the lead from the soil solution through the roots. Since the soil from Metro Manila contains considerable amounts of Pb (Table 2), the uptake by the plants is high. However, not all of the Pb taken up by the plant is also inhibitory. Part can be retained in cell walls (apoplast) and another part can be translocated into vacuoles, where the damaging effect is less than in the cytoplasm[12].

The other part of Pb from the ambient air is loosely but directly attached to the surface of leaves. The surface of leaves is covered by layers of waxes and cutins. The cuticle is a hydrophobic barrier. It is extremely unlikely that heavy metals can enter a leaf through the cuticles. But cuticles can adsorb dust and Pb externally. The amount of such adsorption will depend on the chemical composition of the cuticles and its physical surface structure[17].

Pb toxicity to a plant is one thing, harm to the food chain and toxicity to humans is another concern. Pb contents of food plants higher than 30 mg Pb kg^{-1} dry weight are considered to be toxic although the WHO suggests an upper limit of 500 μg Pb per day (70 kg body weight).

While it is true that leaves of trees sampled in this study are not for human consumption, the results obtained may still imply that any plant, grown in Metro Manila, consumed as vegetables pose a potential health hazard for people living in these areas. This finding can also hold true for plants or vegetables cultivated along busy highways.

This health risk will not end immediately after stopping the use of leaded fuel. The Pb content of the air will decrease rapidly, but the Pb burden for the vegetation and the food chain will continue for a longer time, as the soil acts as a long lasting sink for Pb.

Assuming that a person would consume 500 g of fresh weight of vegetables, leaves or fruits, which have the average total Pb content of leaves of Manila trees (internal plus external Pb), this would yield a daily intake of about 1.3 mg Pb, which would exceed the maximal recommended value by the WHO already by a factor of 6.5. Fortunately, the preferred diet of people in Metro Manila is rice, wheat, and maize, originating from the rural areas. Thus the dietary exposure level e.g. for woman in Manila in reality is only 11 μg Pb per day[13]. This is because seeds of polluted plants contain much less of the pollutants than fruits or the vegetative parts of the plants. We found, for example, that the Pb content of grains of corn (*Zea mays*) growing on soil polluted with Pb is only 14 % of the Pb level in the leaves. For peas (*Pisum sativum*) the corresponding value was 30 %.

Although the above calculations of the potential Pb intake of Manila-grown food plants reflect a worst case scenario, it should be kept in mind too that even plants sampled at the Botolan site, 5-6 h away from Metro Manila (Fig. 2), showed that the total Pb are not totally out of the range assumed to pose health risk.

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