DOI: 10.1007/s001280000163



Lead Concentrations in *Eucalyptus* sp. in a Small Coastal Town

D. Halliwell, N. Turoczy, F. Stagnitti

School of Ecology and Environment, Deakin University, Warrnambool, Vic 3280, Australia

Received: 25 February 2000/Accepted: 25 July 2000

Large amounts of organic substances and heavy metals are emitted into the atmosphere daily as a consequence of human activity. The primary admissions of these chemicals as well as their atmospheric reactions are readily adsorbed into the biological matrix. Lead has no known essential role in the biological sphere. Serious environmental effects can result from high levels of atmospheric lead fallout, leading to detrimental changes in ecosystem and community function (Fatoki, 1996). Lead bio-accumulates in plant material and consequently its concentration can be magnified in the food chain. Serious toxicological effects can result from high levels of lead fall-out (Goldsmith, et al., 1977). High concentrations of lead have been linked to human health problems including nervous system dysfunction of fetus and infants, and in adults, hematotoxicity, reproductive dysfunction and alzheimer's disease (Grobler, et al., 1996; Rodamilans, et al., 1996; KRT, 1997; Ponka, 1998). Although the effects of chronic lead levels in human blood and tissue are well studied, the effects of low concentrations of atmospheric lead on the general population is less well understood. Some have suggested that even low concentrations may lead to increased problems associated with asthma, anemia and learning disabilities (Jensen, et al., 1994; Francek, 1997).

Lead pollution in the atmosphere results from two major sources: industrial processes and vehicle emissions. In the latter case, the major source of lead contamination is from combustion of lead-alkyl additives in petroleum products and the release of lead particulate in the exhaust. Evaporation of petrol, which chiefly occurs at filling stations, also contributes significantly to atmospheric lead levels (Grandjean, et al., 1979; Tumi, et al., 1990). Atmospheric lead may be deposited at great distances from the source. For example, anthropogenic sources of lead have been found in relatively pristine regions such as the Arctic, Antarctica and Greenland (Chiaradia, et al., 1997).

Organolead compounds have been used as antiknock additives in petrols since 1923, and few organic chemicals are now produced in greater quantities than these compounds (Clarke, et al., 1996). The major organolead constituents of "leaded"

petrol are tetraethyl-lead and tetramethyl-lead. These help the fuel to burn more efficiently in the combustion chamber. However, once the combustion process is complete, lead is emitted in a number of halogenated forms; PbCl.Br, alpha and beta forms of NH4Cl.Br and 2NH4 Cl.PbCl.Br). When phosphorus is present in fuel, about one-fifth of the exhaust lead is expelled as 3Pb3 (PO4) 2.PbCl.Br (Faith, et al., 1972).

Lead-free petrol was introduced into Australia in 1975. In 1987, the Australian government introduced legislation to restrict all new cars to use lead-free petrol. However, leaded petrol is still readily available in Australia and a significant number of cars manufactured prior to 1988 still use leaded petrol, although this number is diminishing every year. Most developed countries have noticed a very significant reduction in atmospheric lead concentrations since the introduction of lead-free petrols and improved industrial processes. For example in Spain, the lead in petrol additives decreased from 0.6 g/L in 1983 to 0.15 g/L in 1991. In Barcelona, the concentration of atmospheric lead has decreased from 1.03 - 1.55 mg/m3 in 1987 to 0.18 to 0.30 mg/m3 in 1996 (Rodamilans & Torra, 1996), In Helsinki, lead concentrations ranging from 333 ng/ m3 to 1150 ng/ m3 were found in 1980 and reduced to levels ranging from 7 ng/ m3 to 745 ng/ m3 in 1996. The estimated reduction in total lead emitted into the atmosphere was from 78 tons in 1980 to less than 1 ton in 1996. The significant reduction in atmospheric lead levels was correlated with reduced lead levels in the blood of children in a centrally located day-care centre ranging from 46 mg/L in 1983, to 30 mg/L in 1988 and 26 mg/L in 1996L (Ponka, 1998).

Several previous studies have reported the impacts of lead pollution in major cities (Migon, et al., 1993; Sithole, et al., 1993; Simpson, et al., 1994; Bono, et al., 1995; Devey, et al., 1995; Henshaw, et al., 1995; Liu, et al., 1995; Orlova, et al., 1995; Stromberg, et al., 1995; Liu, et al., 1996; Mao, et al., 1996; Rodamilans & Torra, 1996; Chiaradia, et al., 1997; Ponka, 1998; Vanwijnen, et al., 1995; Davydova, 1996). Other studies have reported atmospheric lead levels in highly industrialized regions (Tumi, et al., 1990; Defre, et al., 1994; Gromov, et al., 1994; Jensen & Fenger, 1994; Kunguru, et al., 1994; Lytle, et al., 1995). Yet other studies have reported the concentrations of lead found in agricultural soils resulting from the application of insecticides containing lead arsenate (Sheppard, et al., 1991; Francek, 1997). However, very few studies have examined the impact of lead pollution in small regional communities that are free from lead-producing industries. This paper therefore investigates the spatial variation of lead concentrations found on the leaves of native vegetation in a small rural coastal town. The selected town has no major lead-producing industry and therefore lead concentrations on plant leaves can be directly correlated with vehicle emissions. This paper is the first part of a long-term study to investigate the effects of legislative changes in vehicle emissions on atmospheric pollution and consequent impacts on vegetation and ecosystem function. We anticipate repeating this study in the same town in 5 and 10 years time to investigate long-term changes, if any.

MATERIALS AND METHODS

The study was conducted in Warrnambool, a small rural coastal town with a population of 28,000 people. Warrnambool is located approximately 260 km west of Melbourne, Australia. This town was chosen for study because it has very few industries. The small factories that operate in this town do not emit lead in any appreciable quantity. The influence of vehicle emissions could therefore be more easily discerned. The primary industry in Warrnambool is tourism and consequently the town has a significant influx of tourists who mainly travel by car from metropolitan Melbourne, particularly in the summer months. The selected survey area extends for approximately a ten kilometre radius from the centre of the town.

Thirty roadside sites were sampled. The sites were chosen at random, depending on tree availability, across the 10 km radius, with the intention of establishing lead concentration contours throughout the area. All trees were sampled within 20 m of a road.

Sampling was conducted on Eucalyptus sp. trees. This tree species was chosen because it meets two basic criteria. First, Eucalyptus sp, a native tree to Australia, is widely distributed throughout the whole sampling area. Eucalyptus sp have a relatively large leaf area, thus concentrations of lead and other particulate matter may be more easily detected compared with other plant species. Second, the movement of lead from the outer surface to the inner parts of the leaves is known to be relatively very small (Ewers, et al., 1991). The adsorption of lead into the biological material of plants is highly variable (Kim, et al., 1994). Some plants, for example mosses (sphagnum auriculatum and polytrichum formosum) have been found to readily incorporate lead (Markert, et al., 1994; Vasconcelos, et al., 1998).

The method described by Christian, (1971) for lead determination was adopted with some modifications outlined below. Based on the results of a prior pilot study, it was determined that between 15 to 30 leaves would be required to obtain significant lead concentrations over a six day accumulation period. This number of leaves provides a compromise between gaining detectable levels of lead and the increase in time required to calculate larger surface areas.

The leaves were collected in large polyethylene bags (approximately 30 x 50 cm), which had been pre-soaked in 0.1 M Nitric Acid. When taking samples, care was taken not to touch the leaves being sampled. The leaves were broken at the beginning of their stems to avoid problems of calculating surface area of the stems. The plastic bags were sealed in the field, by tying a knot in the bag. All samples were analysed within 24 hours of collection.

The leaf surfaces were washed with 40 mL of 0.1 M nitric acid (AnalaR, BDH) by punching a hole in polyethylene bag containing the leaf samples and pouring in

the acid. The hole was then clamped with thumb and forefinger, taking care to keep them clear of the hole, and the bag was shaken vigorously to ensure that all leaf surfaces were thoroughly washed. The washings were then poured into a 50.0 mL volumetric flask which was then made up to the mark. The washings were analysed for lead using a Hitachi 6000 Polarised Zeeman Atomic Adsorption Spectrophotometer using the standard conditions for lead for the instrument. Standards were made from a stock solution of lead nitrate which contained 0.8004 grams of Pb(NO3)2 diluted to 500 mL (source). All glassware (and the plastic funnel) were rinsed twice in 10% nitric acid, twice with distilled water and twice with 0.1 M nitric acid prior to use.

To test for reproducibility of lead concentrations, duplicate samples were taken from a random subset of 8 sites (see Table 1). Seven of these sites showed an error of less than 10 % and one site recorded an error of 20 %. The chemical method therefore exhibited excellent reproducibility.

Table 1. Standard errors for the eight replicate samples.

Sample Site	Reading 1 (ng/cm ²)	Reading 2 (ng/cm ²)	Error
10	25.48	27.41	3.7
12	19.98	16.88	8.4
14	12.84	13.47	2.4
18	25.19	29.14	4.0
20	22.43	33.41	20.0
22	15.20	18.22	9.0
24	11.64	12.50	3.6
36	10.20	11.07	4.1

After washing, the leaves were photocopied onto A4 paper, The surface areas of the leaves were then determined by a cut and weigh method. The average area of sampled leaves was approximately 1200 cm².

RESULTS AND DISCUSSION

Lead accumulations on the leaves varied from as low as 2 ng/cm2 up to 82 ng/cm2. A contour map was generated from the lead concentration data collected over the 30 sites and is presented in Figure 1. The figure displays contour lines at an interval of 10 ng/cm2. Significantly higher levels of lead were recorded in central Warrnambool and elevated levels extended out along the highways. There

are two major roads into Warrnambool: the Princess highway, which runs from east to west through the town, and the Hopkins highway (Mortlake Rd) which runs to the north of the town (see Figure 1). These roads are interrupted by a series of traffic lights in the city centre. Regions neighbouring these roads exhibited the highest lead concentrations. Most of the industries in Warrnambool are located in an industrial estate to the west of the town. The lead concentration levels in western part of the town were low in comparison to central Warrnambool. Therefore, it is unlikely that the few industries present in Warrnambool emit significant amounts of lead into the atmosphere, verifying the assumption that local industries have little impact on lead concentrations in this town.

These results indicate a number of points. First, the primary lead source is from car emissions, confirming our original contention. Second, higher levels of lead are found on roads that have greater traffic use and/or have traffic lights. It is well known that when vehicles are accelerating and decelerating they are operating at lower efficiencies and consequently emit more particulate matter (Faith & Atkinson, 1972).

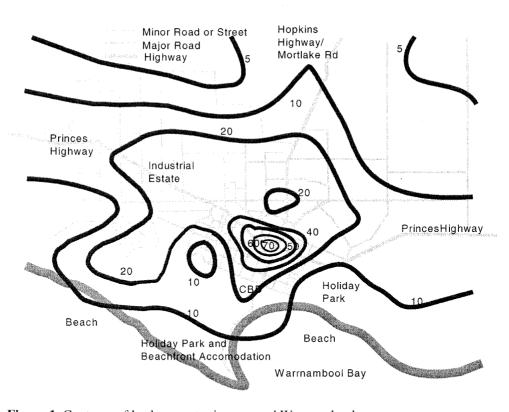


Figure 1. Contours of lead concentrations around Warrnambool.

The highest concentration of lead was 82 ng/cm2. There is no lead fallout standard for Australia. However some European countries, for example, West Germany have set maximum levels of 250 mg of Pb/m2/day (Ewers & Schlipkoter., 1991). If after heavy rain the leaves are washed clean, then the total accumulated lead after six days at a concentration level of 82 ng/cm2 would convert to an equivalent 137 mg of Pb/m2/day, which is well below the European safety limit. Assuming the European standards are based upon toxicity levels and are directly applicable in Australia, there should be no health risk associated with lead poisoning in Warrnambool. There are no other directly comparable studies in Australia.

Previous studies have shown that the introduction of unleaded petrol has significantly reduced levels of atmospherically deposited lead in soils and plants and lowered blood lead concentrations in human populations. Lead in petrol has largely been replaced by an organic manganese compound as an antiknock fuel additive. A recent survey of roadside soil and plants conducted in Utah, U.S.A. has revealed that soil manganese concentrations in high traffic areas are as much as 100 fold higher than historical background levels (Lytle, et al., 1995). Future research, therefore should continue to monitor not only lead concentrations but also manganese.

In contrast to previous studies that have been concerned with the distribution of atmospheric lead concentrations in and around large cities or in regions with major industries, this study investigated concentrations of lead found on the leaves of a common native tree (Eucalyptus sp.) for a small coastal town, largely unaffected by major industrial development. Not surprisingly, the concentrations were found to be highest in the town centre, where two highways intersect, with elevated concentrations extending out along the highways. Comparatively low levels of lead on leaves were found in the major industrial area of the town, confirming unambiguously that the lead concentrations were directly correlated with vehicular use. Even though Australia introduced the use of lead-free petrol more than one decade ago, appreciable concentrations of atmospherically deposited lead resulting from vehicular use still exist, even in small rural communities, as this study demonstrates. Although the lead concentrations around the major intersection were much higher than background levels, when compared with European standards, they are not expected to pose any significant environmental toxicological or human health risk.

REFERENCES

Bono, R., Pignata, C., Scursatone, E., Rovere, R., Natale, P., and Gilli, G. (1995). Updating about reductions of air and blood lead concentrations in Turin, Italy, following reductions in the lead content of gasoline source. Environ Res 70:30-34.

Chiaradia, M., Chenhall, B. E., Depers, A. M., Gulson, B. L., and Jones, B. G. (1997). Identification of historical lead sources in roof dusts and recent lake

- sediments from an industrialized area: Indications from lead isotopes. Sci Tot Environ 205:107-128.
- Christian, G. D. (1971). Analytical Chemistry (Second ed.). New York, John Wiley and Sons.
- Clarke, A. G., Chen, J.-M., Pipitsangchand, s., and Azadi-Bougar, G. A. (1996). Vehicular particulate emissions and roadside air pollution. Sci Tot Environ 189/190:417-422.
- Davydova, S. L. (1996). Present situation and problems of lead pollution of the environment. Petrol Chem 36:283-287.
- Defre, R., Bruynseraede, P., and Kretzschmar, J. G. (1994). Air pollution measurements in traffic tunnels. Environ Health Persp 102:31-37.
- Devey, P., and Li, J. D. (1995). Soil lead levels in parks and playgrounds an environmental risk assessment in Newcastle. Australian J Pub Health 19:189-192.
- Ewers, U., and Schlipkoter. (1991). Lead. In E. Merian (Ed.), Metals and Their Compounds in the Environment: Occurrence, Analysis and Biological Relevance. New York. VCH Publishers.
- Faith, W. L., and Atkinson, J. A. A. (1972). Air pollution (Second Edition). John Wiley and Sons, Inc.
- Fatoki, O. S. (1996). Trace zinc and copper concentration in roadside soils and vegetation Measurement of local atmospheric pollution in Alice, South Africa. Environ Int 22:759-762.
- Francek, M. A. (1997). Soil lead levels in orchards and roadsides of Mission Peninsula, Michigan. Water Air Soil Pollut 94:373-384.
- Goldsmith, J. R., and Friberg, L. T. (1977). Effects of air pollution on human health. In A. C. Stern (Ed.), Air Pollution. New York. Academic Press.
- Grandjean, P., and Nielson, T. (1979). Organolead compounds: Environmental health aspects. In Residue Reviews: Residues or Pesticides and other Contaminants in the Total Environment. New York. Springer-Verlag Inc.
- Grobler, S. R., Theunissen, F. S., and Maresky, L. S. (1996). Evidence of undue lead exposure in Cape Town before the advent of leaded petrol. South African Med J 86:169-171.
- Gromov, S., and Emelina, E. (1994). Lead emission evaluation over the European part of the former Soviet Union. Sci Tot Environ 158:135-137.
- Henshaw, D. L., Keitch, P. A., and James, P. R. (1995). Lead-210, Polonium 210, and vehicle exhaust pollution. Lancet, 345:324-325.
- Jensen, F. P., and Fenger, J. (1994). The air quality in Danish urban areas. Environ Health Perspect 102:55-60.
- Kim, N. D., and Fergusson, J. E. (1994). Seasonal variations in the concentrations of cadmium, copper, lead andzinc in leaves of the horse chestnut (Aesculushippocastanum L). Environ Pollut 86: 89-97.
- KRT (1997), Air pollution effect on trees points to cause of Alzheimer's. The Weekend Australian, p. 48.
- Kunguru, K., and Tole, P. M. (1994). Contamination of soils, maize, wheat and milk with lead from motor vehicle emisssions in Uasin Gishu district, Kenya. Discovery Innovation 6: 261-264.

- Liu, X. D., Gao, N., Hopke, P. K., Cohen, D., Bailey, G., and Crisp, P. (1996). Evaluation of spatial patterns of fine particle sulphur and lead concentrations in New South Wales, Australia. Atmos Environ 30: 9-24.
- Liu, X. D., Hopke, P. K., Cohen, D., and Bailey, G. (1995). Sources of fine particle lead, bromine and elemental carbon in South Australia. Sci Tot Environ 175:65-79.
- Lytle, C. M., Smith, B. N., and Mckinnon, C. Z. (1995). Manganese accumulation along Utah roadways a possible indication of motor vehicle exhaust pollution. Sci Tot Environ 162:105-109.
- Mao, I. F., and Chen, M. L. (1996). Airborne lead pollution in metropolitan Taipei (Republic of China). Water Air Soil Pollut 91:375-382.
- Markert, B., and Weckert, V. (1994). Higher lead concentrations in the environment of former West Germany after the fall of the Berlin wall. Sci Tot Environ 158: 93-96.
- Migon, C., Alleman, L., Leblond, N., and Nicolas, E. (1993). Evolution of Atmospheric lead over the northwestern Mediterranean between 1986 and 1992. Atmos Environ 27:2161-2167.
- Orlova, A. O., Bannon, D. I., Farfel, M. R., Thomas, V. M., Aleschukin, V., Kudashov, V. V., Shine, J. P., and Kruchkov, G. I. (1995). Pilot study of sources of lead exposure in Moscow, Russia. Environ Geochem Health, 17:200-210.
- Ponka, A. (1998). Lead in the ambient air and blood of children in Helsinki. Sci Tot Environ 219:1-5.
- Rodamilans, M., and Torra, J. (1996). Effect of the reduction of petrol lead on blood lead levels of the population of Barcelona, Spain. Bull Environ Cont Toxicol 56:717-721.
- Sheppard, S., and Sheppard, M. (1991). Lead in boreal soils and food plants. Water Air Soil Pollut 57-58:79-91.
- Simpson, R. W., and Xu, H. C. (1994). Atmospheric lead pollution in an urban area Brisbane, Australia. Atmos Environ 28: 3073-3082.
- Sithole, S. D., Moyo, N., and Macheka, M. (1993). An assessment of lead pollution from vehicle emissions along selected roadways in Harare (Zimbabwe). Int J Environ Anal Chem 53:1-12.
- Stromberg, U., Schutz, A., and Skerfving, S. (1995). Substantial decrease of blood lead in Swedish children, 1978-1994, associated with petrol lead. Occ Environ Med 52:764-769.
- Tumi, S. O., Kumar, N. S., and Hinshery, A. K. (1990). Lead contamination levels in roadside vegetation of Tripoli area, Libya. Bull Environ Cont Toxicol 45:718-721.
- Vanwijnen, J. H., Verhoeff, A. P., Jans, H. W. A., and Vanbruggen, M. (1995). The exposure of cyclist, car drivers and pedestrians to traffic related air pollutants. Int Arch Occ Environ Health 67:187-193.
- Vasconcelos, M. T. S. D., and Tavares, H. M. F. (1998). Atmospheric metal pollution (Cr, Cu, Fe, Mn, Ni, Pb and Zn) in Oporto city derived from results for low-volume aerosol samplers and for the moss Sphagnum auriculatum bioindicator. Sci Tot Environ 212:11-20.