

Trace Element Pollution of Soils Collected near a Municipal Solid Waste Incinerator: Human Health Risk

M. Schuhmacher, M. Meneses, S. Granero, J. M. Llobet, J. L. Domingo

Laboratory of Toxicology and Environmental Health, School of Medicine, "Rovira i Virgili" University, San Lorenzo 21, 43201 Reus, Spain

Received: 10 June 1997/Accepted: 26 September 1997

Incineration is often proposed as the treatment of choice for processing municipal solid wastes (MSW) and hazardous wastes. In spite of the known advantages of incineration, this process produces residues, mainly the bottom ash, hazardous filter ash, and additional products of flue gas cleaning operations (Jakob et al. 1995). In relation to those residues, the filter ash contains high concentrations of heavy metals and polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs), whereas the bottom ash contains high concentrations of heavy metals and PCDD/PCDFs (Stern et al. 1989; Jakob et al. 1995). Moreover, some metals can be also found as suspended particulates in air resulting from incineration of MSW (Lisk 1988).

Heavy metals exert a broad range of toxic effects. Elements such as cadmium or chromium are carcinogenic, other metals such as lead, mercury or thallium possess a wide spectrum of toxicity that includes neurotoxic, hepatotoxic, nephrotoxic, teratogenic or mutagenic effects.

MSW incinerators represent a significant source of heavy metals released into the environment. Soils can be contaminated by these metals, which can then bioaccumulate in plants and animals eventually making their way to humans by way of the food chain or contamination of drinking water (Schuhmacher et al. 1991,1993,1994; Frink 1996).

Soil ingestion is usually the most significant route of exposure to soil-bound environmental contaminants, especially in residential areas (Kimbrough et al. 1984). It is particularly important for pollutants that may be consumed by children in the form of dust and dirt originating from soil. Although metal emissions from MSW incinerators are strictly controlled, data on metal deposition in soil samples collected near those facilities are very limited. Particularly scarce is the information on the human health risks derived from ingestion of metal-polluted soils by individuals living in the vicinity of those plants. It should be taken into account that humans may be exposed to soil-borne metals by multiple routes including inhalation, oral and

Correspondence to: J. L. Domingo

dermal (Felter and Dourson 1997). For metal emissions from MSW incinerators, human exposure via intake can dominate exposure by inhalation. The purpose of the present study was to determine the current levels of metals in soils taken near an old (more than 20 years operating) MSW incinerator placed in Montcada (Catalonia, Spain), as well as to assess the health risks derived from metal ingestion through soils by subjects living in the neighborhood of the facility.

MATERIALS AND METHODS

Soil samples were collected in the vicinity of a MSW incinerator located in Montcada (Spain) at approximately 10 km from Barcelona. The plant is placed in a residential area with important industrial activity and began operating in 1975. There are some small mountains at the south and south-east of the facility. A motorway with a heavy traffic is near to the incinerator. A close meteorological station provides data on the wind. The impact area under the influence of the facility was defined by the US EPA dispersion model Industrial Source Complex Long-Term (ISC-LT). Twenty-four sites were selected for sampling, taking into account considerations on the prediction of the time averaged emission plume obtained from the gaussian model (ISC-LT). Duplicate soil samples were taken at 100, 250, 500, 750, 1000, 1500, 2000 and 3000 m from the stack in each of the three directions that the wind blows normally in that area (S, NW, NE). At each site sampling, soils were taken from the upper 3 cm of solum and scrapped into polyethylene sampling bags. To avoid possible contamination, special care was taken during the sampling and subsequent handling. Soil samples were dried at 30°C until constant weight and sieved to pass a 2 mm mesh. About 0.5 g of soil were treated with 6 ml of HNO₃ (65% Suprapur, E. Merck, Darmstadt, Germany) and 1 ml H₂O₂ in teflon bombs. Samples were subsequently heated at 100°C for 8 h. After cooling, solutions were filtered and made up to 25 ml with deionized water (Schuhmacher et al. 1996). Berillium (Be), cadmium (Cd), chromium (Cr), manganese (Mn), nickel (Ni), lead (Pb), tin (Sn), thallium (Tl) and vanadium (V) were determined by inductively coupled plasma spectrometry (ICP-MS, Perkin Elmer Elan 8000). Arsenic (As) and mercury (Hg) were determined by using hydride generation. Rhodium was used as internal standard. The accuracy of the instrumental methods and analytical procedures used was checked by duplication of the samples, as well as by using a reference solution (NIST 1643c), which was run after every 10 samples to check for drift in the sensitivity of the instrument. For each element, quantification was based on the most abundant isotope of that element free from analytical interferences.

Statistical significance of the results was computed by the Kruskal-Wallis test or by Mann-Whitney U test. A probability of 0.05 or less was considered as significant. Linear regression analysis (Pearson's correlation coefficient) was also applied.

RESULTS AND DISCUSSION

Table 1 summarizes the levels of metals in soil samples collected at increasing

Table 1. Concentrations (mg/kg) of metals in soils at increasing distances (m) from the MSW incinerator

Metal	100-200	500-700	1000-1500	2000-3000	p
As	6.30 ± 2.37	7.38 ± 2.70	9.17 ± 1.86	10.05 ± 3.38	NS
Be	0.38 ± 0.05	0.45 ± 0.20	0.53 ± 0.17	0.52 ± 0.50	NS
Cd	0.44 ± 0.44	0.32 ± 0.08	0.33 ± 0.14	0.41 ± 0.21	NS
Cr	18.4 ± 16.50	16.94 ± 9.70	17.45 ± 4.27	17.47 ± 9.30	NS
Hg	0.07 ± 0.04	0.02 ± 0.01	0.11 ± 0.22	0.11 ± 0.16	NS
Mn	307.6 ± 116.9 ^a	357.30 ± 105.88 ^{ab}	484.50 ± 90.27 ^b	407.10 ± 61.20 ^{ab}	< 0.05
Ni	12.26 ± 7.31	11.98 ± 3.90	13.50 ± 6.88	13.36 ± 11.90	NS
Pb	37.36 ± 46.84	59.31 ± 15.36	35.25 ± 17.02	152.17 ± 249.42	NS
Sn	1.00 ± 0.75	0.61 ± 0.64	0.42 ± 0.262	0.30 ± 0.18	NS
Tl	0.10 ± 0.03	0.12 ± 0.04	0.12 ± 0.19	0.13 ± 0.04	NS
V	20.83 ± 7.39	28.13 ± 13.84	25.95 ± 5.79	25.97 ± 6.83	NS

NS: not statistically significant ($p > 0.05$). Values in the same row not showing a common superscript indicate significant differences at $p < 0.05$

distances from the MSW incinerator. Concentrations of most metals increased with distance from the stack. This is a logical finding taking into account that for longer distances particles need more time to settle out from the air. In relation to the specific case of cadmium and lead, our results differ from those reported by Hutton et al. (1988), who found decreased concentrations of both metals in soils with the distance from a refuse incinerator. However, it should be taken into account that a series of factors can affect the metal emission and the subsequent deposition in soils. Those factors include the composition and amount of MSW burned, as well as characteristics of the incinerator such as design, operating conditions during combustion, efficiency of emission control devices, and wind.

In general, the mean levels of metals in the soil samples were similar or even lower than those found in some recent surveys on trace element pollution of soils from different countries. Recently, Frink (1996) reported the most likely concentrations of EPA target analytes in uncontaminated soils of the northeast. These concentrations were of the same order of magnitude than the mean levels found in the present study. Moreover, for some of the most toxic elements, the current levels were similar to the concentrations previously reported for natural uncontaminated soils (Cd: 0.1-0.2 mg/kg; Hg: 0.05-0.08 mg/kg) (Denison and Silbergeld 1988).

The risk to human health posed by contaminated soil in a residential area depends on the potential extent of exposure to soil, as well as the toxic properties of the pollutants. Based on the Hawley model of exposure (Hawley 1985) the assessment of the health risks for the individuals living near the facility here examined shows that metal intake from soils would not exceed (with the exception of As and V) more than 1.1% of their estimated average daily intakes (Table 3). A heavy traffic in the area could increase the predicted exposure to vanadium. With regard to As, in a recent wide review on the total elemental composition of soils, the most likely mean concentration for As in uncontaminated soils was estimated to be 7.6 mg/kg (dry weight) (Frink 1996), value that is similar to the mean concentration of As in the soils examined here. It means that the MSW incinerator under evaluation should not contribute in a significant way to increase the arsenic risk for that population.

The presence and distribution of metals in soils observed here are very similar to those recently found in an assessment of the magnitude and distribution of a number of metals around a newer MSW incinerator located in an industrial area of Tarragona (Catalonia, Spain), which began to operate in 1991 (Schuhmacher et al. 1996). In both studies, the relatively elevated lead levels found in the soil samples collected at the greatest distances from the facilities was considered as an additional fallout of lead from motor vehicles emissions. However, most metal concentrations in soils found in the current survey were within the ranges reported in previous studies. The metal concentrations in soil samples given according to each of the three main wind directions are shown in Table 2. Although for some elements (Be, Hg, Pb, Tl and V) the highest levels in soils corresponded to the north-east direction, the observed differences were not statistically significant for any of the analyzed metals.

Table 2. Concentrations of metals in soil samples at the predominant wind directions

Metal	north-east	north-west	south	p
As	8.13 ± 2.31	7.42 ± 1.69	9.12 ± 4.16	NS
Be	0.54 ± 0.13	0.47 ± 0.12	0.39 ± 0.15	NS
Cd	0.32 ± 0.18	0.33 ± 0.17	0.47 ± 0.34	NS
Cr	13.88 ± 5.14	20.29 ± 13.22	18.52 ± 10.53	NS
Hg	0.16 ± 0.23	0.05 ± 0.04	0.03 ± 0.03	NS
Mn	84.87 ± 80.77	406.22 ± 102.81	376.27 ± 152.57	NS
Ni	9.52 ± 3.16	9.75 ± 4.34	20.92 ± 9.48	NS
Pb	117.73 ± 218.72	47.02 ± 37.62	48.32 ± 35.41	NS
Sn	0.31 ± 0.21	0.62 ± 0.57	0.72 ± 0.66	NS
Tl	0.12 ± 0.02	0.11 ± 0.02	0.11 ± 0.05	NS
V	26.76 ± 10.26	23.34 ± 6.21	25.57 ± 10.35	NS

Results are presented as mean values in mg/kg (dry weight ± SD. NS: not statistically significant (p > 0.05)).

Table 3. Predicted daily exposure to metals from soil on the study site and comparison with the average daily intake by the population living in the area under the influence of the MSW incinerator

Metal	Predicted exposure ¹ (mg/kg/d) x 10 ⁻⁶	Average dietary intake (mg/kg/d) x 10 ⁻³	Exposure as percent of dietary intake	RfD ² (mg/kg/d) x 10 ⁻³	Risk Index x 10 ⁻³
As	7.7	0.16 ³	4.8	0.3	26
Be	0.4	NA	---	5.0	0.08
Cd	0.3	0.80 ⁴	0.04	0.5	0.6
Cr	16.8	1.84 ⁵	0.91	700	0.024
Hg	0.07	0.23 ⁶	0.03	0.1 ⁸	0.7
Mn	366.9	35.7-71.4 ⁷	0.69	5.0	73
Ni	12.0	6.4 ³	0.19	20	0.6
Pb	67.3	1.64 ⁴	1.04	6.0	11
Sn	0.56	0.24 ³	0.23	NA	---
Tl	0.11	NA	---	NA	---
V	23.8	0.14-0.29 ³	11.1	7.0	3.4

¹According to the Hawley model of exposure (body weight = 70 kg). ²Data from Smith (1996). ³Data from Coyer (1991). ^{4,5,6}Data from Schuhmacher et al. (1991,1993,1994).

⁷Data from Keen and Zidenberg-Cerr (1994). ⁸As methyl-Hg. NA: not available.

With regard to the relationship between metal levels and the physicochemical characteristics of the soil, only As ($r = 0.7093$, $p < 0.001$), Ni ($r = 0.5246$, $p < 0.01$) and Tl ($r = 0.5378$, $p < 0.001$) showed a significant correlation with the organic matter content (mean value: 5.88%, range: 2.25-12.08%) of the soil samples. No significant correlations between metal concentrations and the pH values (mean: 7.51, range: 6.85-7.96) of the soils could be established. The low variability in the pH values would suggest a low mobility of the metals in soils. According to the levels of most metals in soil samples found in the vicinity of the MSW incinerator, and taking into account the contribution to human daily intake derived from soil ingestion, the health impact of metal emission on the population living in the area under the influence of the plant seems to be very small.

ACKNOWLEDGEMENTS. This study has been funded by the Entitat Metropolitana del Medi Ambient, Barcelona, Spain.

REFERENCES

- Denison RA, Silbergeld EK (1988) Risks of municipal solid waste incineration: an environmental perspective. *Risk Anal* 8:343-355
- Felter SP, Dourson ML (1997) Hexavalent chromium-contaminated soils: options for risk assessment and risk management. *Regul Toxicol Pharmacol* 25:43-59
- Frink CR (1996) A perspective on metals in soils. *J Soil Contam* 5:329-359
- Goyer RA (1991) Toxic effects of metals. In: Amdur MO, Doull J, Klaassen CD (eds), Casarett and Doull's toxicology. The basic science of poisons, Pergamon Press, New York, 638-655
- Hawley JK (1985) Assessment of health risk from exposure to contaminated soil. *Risk Anal* 5:289-302
- Hutton M, Wadge A, Milligan PJ (1988) Environmental levels of cadmium and lead in the vicinity of a major refuse incinerator. *Atmos Environ* 22:411-416
- Jakob A, Stucki S, Kuhn P (1995) Evaporation of heavy metals during the heat treatment of municipal solid waste incinerator fly ash. *Environ Sci Technol* 29:2429-2436
- Keen CL, Zidenberg-Cherr S (1994) Manganese toxicity in humans and experimental animals. In: Klimas-Tavantzis DJ (ed), Manganese in health and disease, CRC Press, Boca Raton, FL, pp 193-205
- Kimbrough RD, Falk H, Stehr P, Fries G (1984) Health implications of 2,3,7,8-tetrachlorodibenzodioxin (TCDD) contamination of residential soil. *J Toxicol Environ Health* 14:47-93
- Lisk DJ (1988) Environmental implications of incineration of municipal solid waste and ash disposal. *Sci Total Environ* 74:39-66
- Schuhmacher M, Basque MA, Domingo JL, Corbella J (1991) Dietary intake of lead and cadmium from foods in the Tarragona Province, Spain. *Bull Environ Contam Toxicol* 46:320-328
- Schuhmacher M, Domingo JL, Llobet JM, Corbella J (1993) Dietary intake of copper, chromium and zinc in Tarragona Province, Spain. *Sci Total Environ* 132:3-10

- Schuhmacher M, Batista J, Bosque MA, Domingo JL, Corbella J (1994) Mercury concentrations in marine species from the coastal area of Tarragona Province, Spain. Dietary intake of mercury through fish and seafood consumption. *Sci Total Environ* 156:269-273
- Schuhmacher M, Granero S, Bellés M, Llobet JM, Domingo JL (1996) Levels of metals in soils and vegetation in the vicinity of a municipal solid waste incinerator. *Toxicol Environ Chem* 56: 119-132
- Smith RL (1996) Risk-based concentrations: prioritizing environmental problems using limited data. *Toxicology* 106:243-266
- Stern AH, Munshi AA, Goodman AK (1989) Potential exposure levels and health effects of neighborhood exposure to a municipal incinerator bottom ash landfill. *Arch Environ Health* 44:40-48