Model simulation on the origin of lead and cadmium deposition over Hungary
L. Bozó

Institute for Atmospheric Physics, H-1675 Budapest, P.O. Box 39, Hungary

Abstract

Lead and cadmium concentrations both in the air and precipitation have been monitored since the early 80’s under regional background conditions in Hungary. Lead and cadmium are attached to aerosol particles mostly in fine size range. Hence, they can be transported hundreds or more kilometers from their sources before gradually being removed from the atmosphere by dry or wet depositions. The geographical origin - country-to-country budgets - of lead and cadmium deposited in Hungary was estimated by means of a European-scale long-range transport model.

1 Introduction

In recent years there has been an increased interest in trace metals in the atmosphere and the environmental effects of their deposition. This is to large extent because heavy metals can accumulate in the biosphere and may be toxic to living systems. Lead and cadmium are among the most toxic elements in the atmosphere. The rate of contamination can vary from place to place as a function of source densities and intensities of heavy metals flux as well as meteorological conditions. Aerosols containing heavy metals can also be transported far away from their sources before being deposited. It means that a significant part of lead and cadmium emitted in a given country can be removed from the atmosphere beyond its boundaries causing environmental pollution problems over other countries.
2 Monitoring of Pb and Cd in Hungary

The monitoring of Pb and Cd in atmospheric aerosols and precipitation were begun at the regional background air pollution monitoring station, K-Puszta, in 1981. This station is located on the Hungarian Great Plain (19°33'E, 46°58'N) in central Hungary, about 75 km southeast of Budapest. The nearest city is 25 km southeast (100,000 inhabitants).

Aerosol samples are taken every second day by means of Whatman 41 filters. The sampling period is 24 hours and during this time around 20 m³ of ambient air are pumped over. Lead and cadmium contents in the samples are determined using graphite furnace atomic absorption spectrometry at the Department of Analytical Chemistry of Eötvös Loránd University, Budapest Bozo & Horváth [1].

Annual variation of Pb and Cd air concentrations are demonstrated in Fig.1. It can be seen that the annual average Pb concentrations were significantly higher in the early 80's than in the second part of the decade and early 90's. The decreasing trend could be caused by the fact that the amount of lead added to a unit volume of gasoline has continuously been decreased (since January, 1993 its value is 0.15 g L⁻¹ in Hungary) and the use of unleaded gasoline has become more widespread in recent years mostly in Western Europe.

Concerning the Cd air concentration, higher yearly average values can be seen in the first half (1982-1986) of the monitoring period than in the second one (1987-1993). The decrease may be caused by the development and installation of environmental friendly industrial technologies in the western part of Europe and the economical decline (mostly in the late-80's) in Eastern Europe.

![Figure 1. Multiyear variation of Pb and Cd air concentrations at K-puszta](image)
Analyses of Pb and Cd content of precipitation were started in 1983 (Pb) and 1984 (Cd). Wet only collectors are used for precipitation sampling. Monthly average lead and cadmium concentrations are determined by mixing the individual precipitation samples. For the chemical analyses graphite furnace atomic absorption spectrometer is used. Wet deposition are calculated by multiplying the trace metal concentration in precipitation and the precipitation amount.

The results of the measurements are shown in Fig.2. Highest annual lead depositions were calculated for 1984 and 1987. Since 1987 there has been a decreasing trend in wet deposition of lead over Hungary. The highest Cd wet depositions were detected in 1984 and 1985 and the lowest ones in 1989 and 1993. Generally it can be stated that the annual Cd wet depositions were higher in the period of 1984-1987 than those of between 1988-1993.

![Figure 2. Multiyear variation of Pb and Cd wet deposition at K-puszta](image)

3 Modeling of long-range transport of Pb and Cd

The TRACE model developed at IIASA, Laxenburg, Austria Alcamo et al. [2] focuses on European-scale emission, transport and deposition of As, Cd, Pb and Zn. An application of this model to the estimation of total deposition and budgets of heavy metals over Eastern Europe is given in Bozó et al. [3]. The model relies on long-term average meteorological data as input, and computes the long-term (annual) average levels of trace metals in the
atmosphere. The calculations are based on mid-80’s (Cd) and late-80’s (Pb) emission inventories with 150 X 150 km² grid resolution over Europe.

The calculation procedure of this climatological-type model is divided into two steps: first, the loss of pollutant from a parcel of air as it travels from a source to a receptor is represented by a simple loss term. This equation gives the air concentration of a pollutant at a receptor located any distance downwind from a source:

\[
c(x_r,y_r;x_s,y_s) = B \ E(x_s,y_s) \ R^{-1} \ (1-\alpha) \ e^{- kd + kw} t
\]

where \( c \) is the air concentration due to a single emission source; \((x_r,y_r)\) is the receptor position; \((x_s,y_s)\) is the source position; \( E \) is the emission at the source; \( R \) is the distance between source and receptor; \( \alpha \) is the local deposition coefficient (part of Pb and Cd emitted and deposited over the same grid); \( kd \) and \( kw \) are the first-order loss coefficients, which reflect the loss of Pb and Cd from the air parcel by dry and wet deposition processes, respectively. The factor \( B \) is derived by assuming mass conservation i.e. setting the upward flux of metals equal to the downward flux of deposition (for details see Alcamo et al. [2]). The total concentration at the receptor is computed from the sum of contributions coming from all emission sources, weighted according to the frequency of backward trajectories coming from any of 8 sectors.

In the second step of the calculation, wet and dry deposition of the pollutant at the receptor is computed from the air concentration:

\[
D_w = c(x_r,y_r) \ W \ P
\]

where \( P \) is the annual precipitation amount and \( W \) is the scavenging ratio. Dry deposition is calculated by:

\[
D_d = c(x_r,y_r) \ v_d
\]

where \( v_d \) is the dry deposition velocity computed on the basis of the semi-empirical model of Sehmel [4] as a function of particle size, surface roughness and friction velocity.

The model outputs (both air concentrations and depositions) were compared to background measurements carried out at the European monitoring sites. With regards the Pb, the average of computations and observations referring to air concentration are very close and all computations are within a factor of two of the observations. Calculations of wet deposition are also correlated to the available observations. Cd calculations clearly underestimate observations but are generally within a factor of two of observations. Model calculations of Cd air concentration are correlated with observations. The
reason for the underestimation can be the inaccuracy of Cd emission inventory and/or contamination of samples (Alcamo et al. [2])

4 The origin of Pb and Cd deposition over Hungary

The results of model calculations for the estimation of the country-origin of lead and cadmium deposition in Hungary are shown in Fig. 3. The highest relative Pb contribution was calculated for Hungarian sources (19%). However, the contribution of the former Soviet Union and Poland to the total (dry+wet) lead deposition over Hungary is also significant (18% and 12%, respectively).

Concerning the model calculations for Cd - based on a gridded European emission inventory valid for mid-80’s - the following conclusions can be drawn. Approximately the half of the total cadmium deposition over Hungary originates from industrial sources of Poland. On the basis of our model computations even the former Yugoslavian and Czechoslovakian sources provide more cadmium deposited over Hungary (11% and 9%, respectively) than Hungarian emitters do (7%).

Figure 3. Origin of Pb and Cd deposition over Hungary
As it was mentioned above, lead emission decreased significantly in the western European countries during the last decade due to the introduction of unleaded gasoline and using less lead added to the conventional fuel. Breaking down the contributions to (1) Hungary itself (2) western European countries and (3) other eastern European countries the following conclusions can be drawn (Fig.4.). The relative contribution of Hungarian sources increased from 16% to 19% during the period of 1982-1989 while that of western European sources decreased from 33% to 20%. At the same time the percentage contribution of eastern European sources increased from 51% to 61%. Unfortunately, the most recent gridded European emission inventory for lead is valid for 1989. It should also be noted, however, that during the recent years the consumption of unleaded gasoline has become more widespread also in Hungary, and the lead content of gasoline has been decreased as well. Lead emission from road traffic in Hungary between 1980 and 1991 is shown in Fig.5.

On the basis of TRACE-model computations the atmospheric Pb and Cd budgets over the eastern European countries for the mid-80’s have also been estimated (Bozó et al. [3]). Concerning lead, an up-dated estimation of the atmospheric budgets valid for 1989 is presented in Fig.6. In Bulgaria and Poland the emissions exceed the total deposition. For the other countries the total depositions are higher than the countries' own emissions.
Figure 5. Pb emission from the consumption of leaded gasoline in Hungary

Figure 6. Atmospheric budget of Pb over the eastern European countries
5 Conclusions

It can be concluded from the measurements and model calculations that the atmospheric concentration and deposition of Pb and Cd in rural areas of Hungary are greatly affected by sources hundreds of kilometers far away from the receptor area. In the case of Hungary, it is particularly true for Cd. Emission data for the model calculations were available only for the mid-80’s (Cd) and late-80’s (Pb). However, up-dated emission inventories are badly needed for the evaluation of the present rates of deposition and atmospheric budgets.

Acknowledgements: This paper was sponsored by the U.S.-Hungarian Science and Technology Joint Fund in cooperation with National Oceanic and Atmospheric Administration and Hungarian Meteorological Service under Project J.F.No. 293.

REFERENCES


