Atmospheric Heavy Metal Input to Forest Soils in Rural Areas of Denmark

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Atmospheric bulk deposition of heavy metals (HM) was measured from 1972/73 to the present time at five to ten forest sites in rural areas of Denmark. From 1979, HM in aerosols were measured at one to four forest sites. On the basis of these long-term continuous measurements, the atmospheric inputs to the forest floor have been calculated. Yearly HM emission estimates to the European atmosphere seems to correlate well with yearly average values of HM deposition, as well as with HM concentrations in the ambient atmosphere. HM emissions have been estimated since the 1950s. Using the correlation between emission and deposition, HM deposition values maybe extrapolated in reverse chronological order. The accumulated atmospheric HM deposition has been estimated in this way over a period of 50 years.

KEYWORDS: deposition, aerosols, metal accumulation

INTRODUCTION

Heavy metals (HM) are a natural constituency of igneous rocks, sediments, and soils. In soils, most HM occur in relatively small concentrations, less than 100 mg/kg DM; an exception is iron (Fe), normally found in concentrations over 5000 mg/kg.

The industrial revolution in Europe and North America in the mid-19th century enhanced the emissions of oxidized sulfur and nitrogen compounds, but also the emissions of particulate matter. Particles emitted from energy production and industrial processes are enriched in HM compared to the abundance in the earth’s crust and in pristine soils[1].

It has been documented by several authors that topsoils close to industrial emitters, as well as topsoils in major cities, are affected by local emissions[2]. The intention of the present project is to determine the accumulated atmospheric input to forests in typical European background areas, here represented by rurally located forests in Denmark. The empirical basis is long-term sampling in forests of atmospheric deposition of HM (Fig. 1).
FIGURE 1. Yearly bulk deposition of lead (Pb) and cadmium (Cd) measured at rural forest sites in Denmark since 1973. The fitted lines indicate the trend.

MEASUREMENTS

The atmospheric deposition of HM was assessed from 28 years of measurements of bulk precipitation at five to ten forest sites in rural areas of Denmark. Dry deposition of HM was estimated from the sampling of aerosols at one to four forest sites at some of the same stations; aerosol measurements started in 1979 (Fig. 2). Sampling and the analytical methods are described by Hovmand and Kemp[3,4].
The total atmospheric input to the forest ecosystem was calculated from bulk precipitation measurements and dry deposition estimates. Measurements of HM deposition made between 1973 and 1979 showed no systematic changes or trends in the yearly average values. After 1979, a pronounced downward trend is seen for the industrially emitted HM in bulk deposition as well as in HM concentrations of the aerosols. To our knowledge, no systematic measurements of HM were carried out at rural sites in Northern Europe before 1973. Therefore, deposition rates might have been even higher than the maximum levels measured during the early 1970s. To get a more complete picture of the accumulated deposition over time, other information should be taken into consideration. Peat bog profile analyses seem to be a useful tool to estimate HM deposited to the bog surfaces over time[5,6]. We took, however, another approach by comparing bulk deposition from background stations with regional emission estimates over the same span of years as our measurements covered.

EMISSION

European estimates on HM emissions were compiled by Pacyna[7]; estimates for three HM are summarized in Fig. 3. The trends in emissions are comparable to measured trends in bulk deposition and in aerosol HM concentrations. Emissions have been estimated back to 1955. Most HM seem to peak around 1960–1965; Pb is an exception with highest emissions around 1975 due to the wide use of leaded gasoline at that time.

Measured HM deposition and calculated HM emissions were compared over six 5-year periods. The correlation between paired data sets was remarkably high for Pb, Zn, and Cd. Results are shown in Table 1.

The slope of the correlation line describes the ratio between yearly emissions and depositions. These ratios are used to calculate depositions of HM from 1955–1970. The calculated yearly bulk depositions shown as 5-year average values are indicated in Fig. 4. Filled columns are measured values and unfilled columns are calculated.
In Table 2, the accumulated bulk deposition and the accumulated total atmospheric deposition (sum of dry and wet deposition) to the forest surface over a period of 50 years are summarized. Bulk deposition measurements do not fully take into account the dry deposition flux of aerosols to the forest canopy. Pb deposition was measured in a beech and spruce forest in Denmark over a period of 1 year[8]. The results concluded that the total atmospheric input of Pb on average was a factor 1.6 higher than average bulk...
deposition measured at a nearby open field site in the forest. This dry deposition flux corresponds to an average aerosol deposition velocity of $2 \times 10^{-3}$ m·s$^{-1}$.

**TABLE 2**

Bulk and Total Deposition of Pb, Zn, and Cd to the Forest, Accumulated over a 50-Year Period (1955–2005)

| Element | Bulk Deposition (mg·m$^{-2}$) | Total Deposition (mg·m$^{-2}$) |
|---------|-------------------------------|-------------------------------|
| Pb      | 357                           | 571                           |
| Zn      | 1086                          | 1738                          |
| Cd      | 8                             | 12                            |

**SOIL**

The total atmospheric deposition of HM to the forest is expected to accumulate in the upper layer of the forest soil, due to the fixation potential of the humus layer found in most Northern European forests. A typical vertical soil profile from a beech forest shows the highest concentrations of HM in the uppermost layers of the forest soil. Cu and Zn are taken up by the trees and deposited with the litter, but atmospheric deposition also adds to the buildup of these HM in the top layer of soil. Atmospheric deposits of Pb are responsible for the steep vertical gradient seen in Fig. 5.

![Soil profile from a typical beech forest. Zn, Cu, and Pb concentrations (in ppm DM) are decreasing with depth.](image)

**FIGURE 5.** Soil profile from a typical beech forest. Zn, Cu, and Pb concentrations (in ppm DM) are decreasing with depth.
Ongoing research, based on HM analyses of forest soils, tries to quantify the fraction of soil HM content attributed to atmospheric deposition. (Project: “Trace Metal Dynamics in Forest Ecosystems 2005–2007”. MFH@BI.KU.dk; University of Copenhagen).

CONCLUSION

Long-term measurements of HM deposition at five to ten rural forest sites document an accumulated input over time. The deposition input has the potential to change the relative element composition of the forest topsoils in the Northern European background area.

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