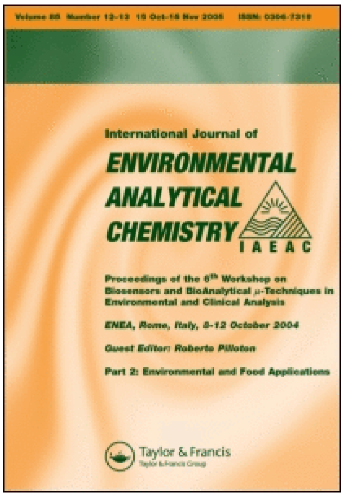

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B. C. Gälli[a]; P. R. Bürki[a]; U. P. Nyffeler[ab]; P. W. Schindler[a]
[a] Department of Inorganic Chemistry, University of Berne, Berne 9, Switzerland [b] Federal Office for Environmental Protection, Berne, Switzerland

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PARTICLE SIZE DISTRIBUTION OF AIRBORNE TRACE METALS*

B. C. GÄLLI, P. R. BÜRKI, U. P. NYFFELER† and P. W. SCHINDLER

Department of Inorganic Chemistry, University of Berne, Freiestr. 3, CH-3000 Berne 9, Switzerland

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Weekly samples of aerosol in seven size fractions were taken in the period from 13 October 1985 to 16 September 1986, and analyzed for cadmium, copper, manganese, sodium, lead, zinc and—partially—arsenic and vanadium.

Sampling locations were south of Berne, Switzerland, at elevations of 515, 750, 940 and 1550 metres above sea level, respectively.

First results indicate that the aerosol concentrations usually decrease with increasing elevation; the extent of this decrease is, however, largely controlled by the prevailing atmospheric stability.

During some weeks in February 1986 unusually high aerosol concentrations were observed. The meteorologic situation and the observed element spectrum indicate that transport from Eastern Europe had occurred.

Each component of the aerosol exhibits a typical pattern of particle size distribution which, for most elements, does not vary significantly with altitude.

KEY WORDS: Atomic absorption spectrometry, meteorology, aerosol, air pollution, impactor, size distribution, trace metals.

INTRODUCTION

The impact of atmospheric particulate matter on variable receptors is greatly dependent on particle size.[1] Among the receptors the human lung is of special interest. Particles larger than 10 μm are efficiently removed in the nasal cavity and are therefore not of great potential danger. Particles below about 1 μm are deposited in the tracheo-bronchial and alveolar regions where they can be harmful to the lung.[2] Thus the size distribution of potentially toxic elements on aerosols is an important parameter of speciation.[3]

The present paper reports parts of a larger study on trace metals in the atmosphere, their origin and deposition. Special emphasis is given to their size distribution and their concentration in the air as a function of both time and altitude.

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†Present address: Federal Office for Environmental Protection, CH-3003, Berne Switzerland.

Table 1 Impactor characteristics: 50% efficiency diameters and sampling interval as given by the manufacturer and sampling interval determined in a calibration experiment by Wang and John[4]

Impactor stage	*50% efficiency (μm)*	*Interval (μm)*	*Calibration (μm)*
1	0.086	0.06–0.125	0.082–0.13
2	0.18	0.125–0.25	0.13–0.21
3	0.35	0.25–0.5	0.21–0.43
4	0.7	0.5–1	0.43–0.96
5	1.4	1–2	0.96–2.1
6	2.8	2–4	2.1–4.2
7	5.7	4–8	4.2–8.6

EXPERIMENTAL

In the period from 13 October 1985 to 16 September 1986, weekly samples of aerosol were collected at four sites south of Berne using nine-stage Berner low pressure impactors (LPI 30/0.06/2). The characteristics of the impactors are given in Table 1. The aerosol was collected on Nucleopore polycarbonate foils which were digested in concentrated nitric acid (Merck, suprapur) in closed teflon vessels during 1 hour at 180°C. The resulting solution was diluted with doubly distilled water and analyzed for Na and Zn by flame atomic absorption using a Beckman AAS 1248, for Cd, Cu, Mn, Pb and partially V by graphite furnace AAS (Varian AA 1475 and GTA 95) and for As using the hydrogen generation technique.

The sampling sites are shown in Figure 1. Site A is lying near the city of Berne and near the major traffic route, but the surroundings of stations A to C are generally rural. Site D is quite far away from major human emission sources, although there is a road leading up into the mountains at about 2 km away from the site. At sites C and D the major wind directions are given.

In Figure 1b the height profile of the sampling locations is shown. The sites are at altitudes of 515, 750, 940 and 1550 m above sea level, respectively, thus separated by more than 1000 m in vertical direction. They were chosen to approximate the monitoring of a vertical air profile and to study the transport of pollutants.

RESULTS

The elements measured are known in the literature as tracer elements for important types of sources.[5–7] Their origin as well as the annual average concentration and the concentration ranges of the particle mass and of the elements are given in Table 2. As a comparison the regulatory standards defined by the Swiss Federal Air Quality Control Regulation are also included. The concentrations measured were well below these limits.

Figure 2 shows the temporal variation of the total particle concentration at the

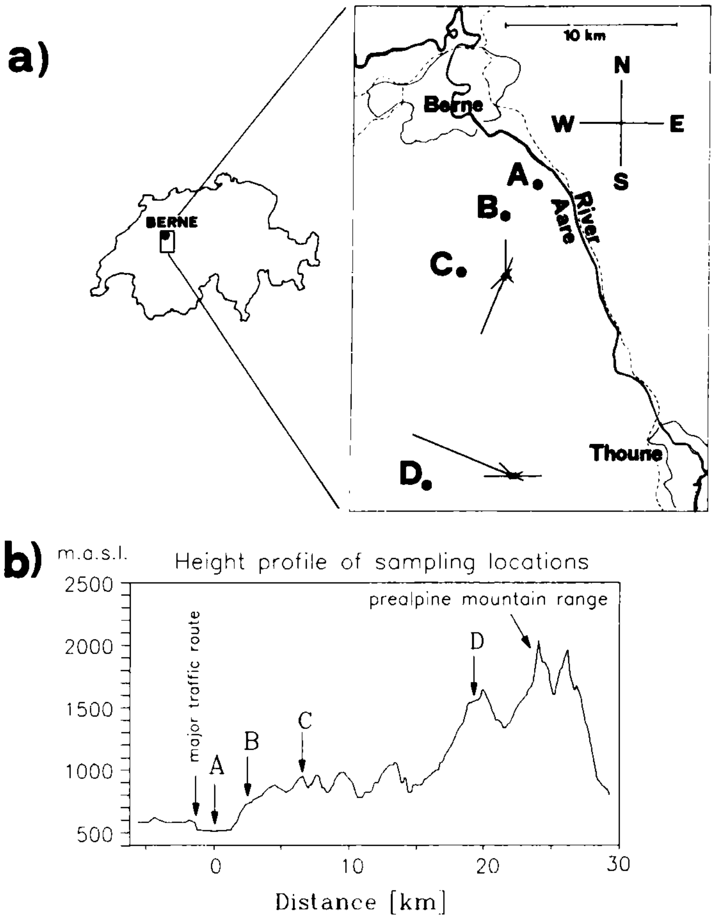

Figure 1 (*a*) Map of the region south of Berne, Switzerland with the sampling sites A, B, C and D and the cities of Berne and Thoune. The dashed lines indicate the major traffic routes. At sites C and D the major wind directions are given. The bars represent the windspeed multiplied by the time during which the wind was blowing from that direction, thus giving a measure for the displacement of air masses. (*b*) Height profile of the sampling locations.

four sites. The important effect of the meteorological factors, essentially the atmospheric stability, is obvious. During fall and winter the emitted aerosol is concentrated in the lower air layers. The frequently occurring temperature inversion prevents an efficient mixing of the air masses. As a result, the air at the uppermost site, situated above the inversion layer (800–1200 m), is very clean. This large reduction of aerosol concentration is not observed during spring and summer when the atmosphere is well mixed. The concentration evolves parallel at all the sites. As the contaminants are injected into a larger volume of air during spring and summer, the concentrations at site A are generally lower than during fall and winter.

One special event occurred in winter 1986, from 4 February to 25 February, when air masses from Eastern Europe were transported to Switzerland. This

Table 2 Annual average concentrations (and concentration ranges) of the elements in the air at the four sites together with the standards imposed by the Swiss Federal Air Quality Control Regulation (LRV) and the major sources of the elements as mentioned in the literature

Element	*Sites*								*LRV*	*Source*
	A		*B*		*C*		*D*			
Average concentration (range) (ng/m^3*)*										
As	2.2	(0.49–13.8)	2.0	(0.43–12.0)	1.7	(0.10–12.0)	0.82	(0.02–6.6)		Coal burning
Cd	0.88	(0.22–2.1)	0.69	(0.20–2.3)	0.59	(0.15–1.5)	0.3	(0.02–1.1)	10	Waste incineration
Cu	7.9	(2.0–16.9)	4.5	(0.62–11.1)	3.4	(0.79–9.4)	2.7	(0.08–8.0)		
Mn	31.7	(8.3–132)	28.8	(5.5–166)	25.4	(5.6–70)	24.1	(3.0–264)		Soil
Na	170	(49.5–612)	154	(41.4–521)	138	(2.8–349)	81	(16.1–247)		Sea spray, soil
Pb	134	(37.8–382)	55.5	(14.0–154)	39.1	(9.8–99.8)	17.9	(1.1–43.6)	1000	Traffic
Zn	65.0	(12.9–211)	47.7	(12.6–177)	34.9	(9.10–135)	16.7	(2.9–56.6)		Waste incineration
V	3.9	(0.69–16.5)	3.6	(0.80–13.2)	3.1	(0.75–12.7)	1.5	(0.18–7.4)		Oil burning
Average concentration (range) (μg/m^3*)*										
Mass	37.1	(8.7–127)	30.8	(6.2–110)	25.0	(5.2–74.4)	12.6	(0.51–35.5)	70	

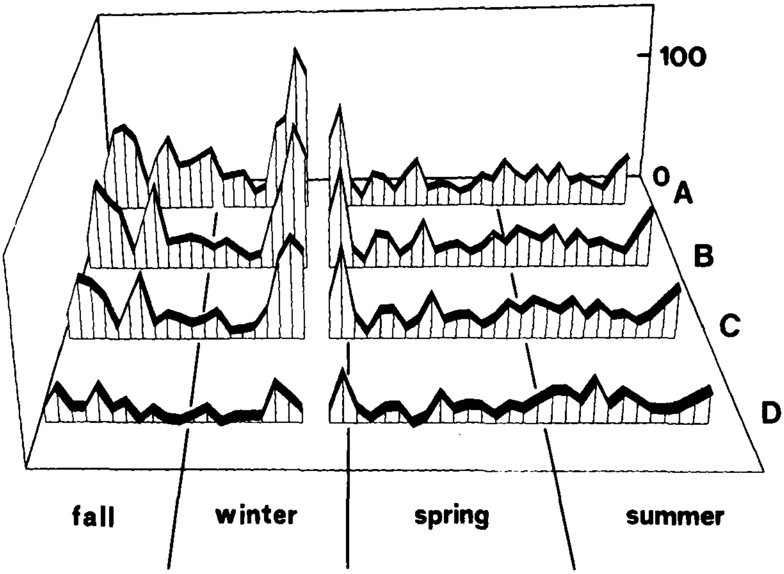

Figure 2 Temporal variation of the particle mass in $\mu g/m^3$ (sum of stages 1 to 7). One point represents the mean of 1 week.

resulted in a much higher particle concentration and those samples also showed a different elemental pattern than normally observed, especially characterized by very high As and V levels.

Other factors influencing the aerosol concentration are windspeed and long-lasting dry periods.

In Figure 3 the size distributions of the annual average of particle mass and of some elements are given. The mass size distribution is very similar at the four sites, indicating that the important sources of mass are either the same everywhere, or that the sources at every site have the same size distributions and relative intensities. A clear decrease of mass concentration with increasing elevation is observed in every particle size class (as a result of the strong height dependence during fall and winter).

Most of the mass is contained in the particles below 1 μm aerodynamic diameter. The elements we measured add up to about 1 % of total aerosol mass.

Very similar features to those of particle mass are observed for cadmium and zinc as well as for arsenic and vanadium (the last two elements were not measured in all the samples).

Manganese exhibits different features: The size distribution is changing from site to site and the concentration reduction is not as clear as for the other elements. This important local influence is expected if soil is an important Mn source.

The most striking differences between the sites can be seen for lead. The concentration reduction with elevation is much larger than for every other element. The second observation is the change in the size distribution: At the site

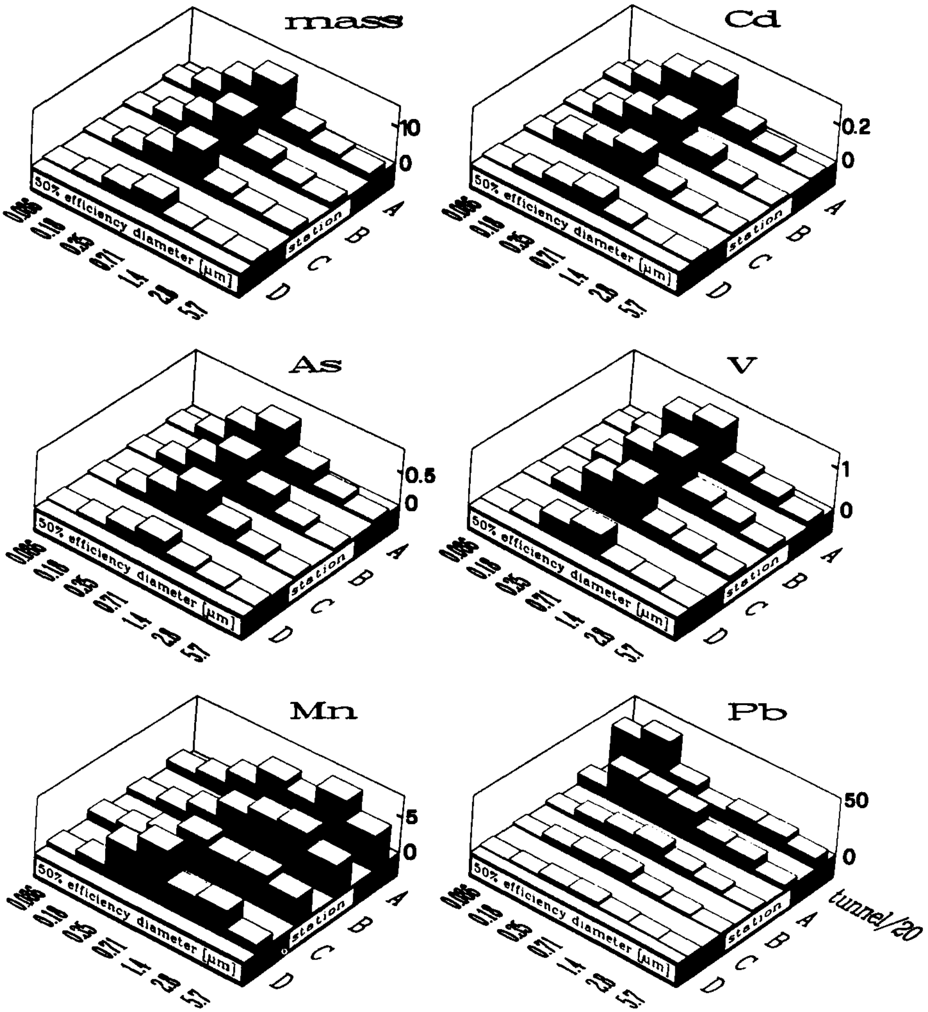

Figure 3 Size distribution of the annual average concentrations of mass and some elements at the four sites. The z-axis unit for the mass concentration is $\mu g/m^3$, for the element concentrations ng/m^3. The lead values measured in the St. Gotthard tunnel are also included (concentrations divided by 20).

near the emission source the maximum mass is found in the particle size class between 0.125 μm and 0.25 μm aerodynamic diameter, while at sites further away the maximum is shifted to about 0.7 μm. This trend of the shift of mass to larger particles is also consistent with the results found for very freshly emitted aerosol as measured in the St. Gotthard tunnel (highway N2): Most of the lead is found on particles below 0.25 μm aerodynamic diameter. Table 3 lists the mass median diameter of lead at the source and the sites and of some other elements at the sampling locations.

Table 3 Mass median diameters of several elements determined from the annual average size distribution at the four sites. For lead and particle mass, the MMD measured in the St. Gotthard tunnel is also given

Element	*Mass median diameter* (*μm*)				
	A	*B*	*C*	*D*	*Tunnel*
Mass	0.63	0.65	0.65	0.63	0.19
Pb	0.43	0.54	0.54	0.54	0.19
Cd	0.54	0.60	0.54	0.58	-
Mn	1.37	1.48	1.06	0.73	-
As	0.77	0.68	0.66	0.66	-
V	0.60	0.64	0.63	0.60	-

CONCLUSION

The results have shown that most of the elements possibly dangerous to human health are concentrated in the inhalable fraction on particles below about 1 μm aerodynamic diameter. Especially lead is able to enter the alveolar region and a part will be deposited there.

The concentrations measured in Switzerland are comparable to results obtained in rural areas of other European countries and of the U.S. and are well below the standards imposed by the federal government.

The composition of the aerosol reaching the sites is quite similar at the different locations with the exception of lead and manganese. Presumably most of the measured elements are emitted in the densely populated Swiss Plateau and get well mixed before they reach the sampling sites. In contrast, when the aerosol is imported from the Eastern part of Europe a different elemental composition is observed.

The factors controlling the concentration and composition of the aerosol are the origin of the air masses, windspeed, the vertical stability of the atmosphere, rainfall and dry periods, and the distance to the major sources.

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