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Title: Determination of trace elements in ambient aerosols by synchrotron XRF

Abstract: (min. 300 - max 500 words)
Aerosols from combustion and abrasion processes often contain traces of metallic elements that are difficult to determine with conventional methods, which typically require long sampling times to accumulate enough mass for analysis. Synchrotron x-ray fluorescence (SR-XRF) spectrometry is a very sensitive method for elemental analysis that allows for very small amounts of material to be measured. This in turn can be profitably used for reducing the sampling time of individual samples to hours instead of days such that studies of diurnal variations become feasible.
We collected ambient aerosol samples with rotating drum impactors in different geographic settings (in an alpine valley, close to a freeway, in a city) with time resolutions of two hours in the winter of 2005/2006. The impactors segregated the particles into three size bins (PM1, PM2.5-PM1, and PM10-PM2.5). Accidentally two of the three field campaigns coincided with heavy pollution episodes, where the legal thresholds of 50 µg/m³ (daily average) were exceeded by up to a factor of 3. The samples were then analyzed with SR-XRF at the Swiss Synchrotron Light Source (SLS) at PSI. The elemental compositions did not differ much between the sites in northern Switzerland. Heavier elements were found mainly in the largest size fraction, which is usually considered to originate from (mineral) dust resuspended into the air or from mechanical abrasion. The smaller particle size fraction showed high amounts of carbonaceous material not detectable with our XRF setup and is more typical for combustion processes. K, Si, Fe, Ca, Al, Na, Mg, Ba, Zn were identified as the important metallic contributors, while Ti, Cu and Cr were found only in traces. K is an indicator for wood combustion, but is also a component of mineral dust and de-icing salts applied to roads during wintertime. The elemental fingerprints determined with SR-XRF will be used for source apportionment studies to help determining specific emission reduction strategies.

Short CV:
I studied physics at ETH and acquired a PhD degree in geography (climatology) at the University of Berne in 1990. Since then I have been working at PSI in the fields of air quality and air pollution meteorology in complex terrain, atmospheric fluxes and – more recently – aerosol chemistry. I spent a post-doc year at Pacific Northwest Laboratory in the U.S. in 1992/93. My research emphasis is on experimental work, especially field studies in mountainous terrain.
Determination of trace elements in ambient aerosols by synchrotron XRF

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Motivation and Background

Aerosols from combustion and abrasion processes often contain traces of metallic elements that are difficult to determine with conventional methods, which typically require long sampling times to accumulate enough mass for analysis. Synchrotron x-ray fluorescence (SR-XRF) spectrometry is a very sensitive method for elemental analysis that allows for very small amounts of material to be measured. This in turn can be profitably used for reducing the sampling time of individual samples to hours instead of days such that studies of diurnal variations become feasible.

We collected ambient aerosol samples with rotating drum impactors in different geographic settings (in an alpine valley, close to a freeway, in a city) with time resolutions of two hours in the winter of 2005/2006. The samples were then analyzed with SR-XRF at the Swiss Light Source (SLS) at PSI. The combination of sampling and analysis methods allow for:

- High temporal resolution of measurements (hours)
- Size segregated detection of (metallic) trace elements (in concentrations as low as pg m⁻³)
- Fast analysis of a large number of individual samples

The elemental fingerprints determined with SR-XRF will be used for source apportionment studies to help determining specific emission reduction strategies.

Aerosols from wood burning

It has been realized in recent years in Switzerland, that the combustion of wood contributes significantly to the amount of particulate matter in the air, especially during winter (Szidat et al. 2007). Therefore, rural areas may suffer more from air quality problems than previously assumed. How emissions from wood stoves and open fires influence the composition of particulate matter is the subject of ongoing research.

Average elemental composition of winter smog aerosols in Zurich

Iron (Fe) can be found in significant amounts in all size fractions, although decreasing with decreasing size. This may be a strong indicator of traffic emissions, mainly of abrasion of mechanical parts in cars, engines, and on road surfaces. Coarse particle elements Cl and Ca are probably the result of road dust and of the application of de-icing salts on the roads. In contrast, K increases with decreasing size fraction. The large amount in the smallest size fraction hints towards biomass burning in domestic heating. Sulfur is almost absent in the largest size fraction, but amounts to about half in the B and C stages. Fine mode S can thus be assigned to secondary sulfate aerosol.

Conclusions

The results presented here demonstrate:

- The advantage of size-segregated aerosol sampling with rotating drum impactors
- The applicability of the SR-XRF method for analysing small amounts of aerosol mass
- A gain in information on emission characteristics from various sources in different size fractions

References


Fig. 1: Rotating drum impactor. Left side: Instrument control. Right side: Impactor drums (open) for 3-stage particle size segregation.

Fig. 2: Mylar tape of stage B (1 - 2.5 μm) with collected particles. Each “bar” corresponds to 2 h of sampling.

Fig. 3: Synchrotron-XRF setup showing the sample holder on the goniometer table in the Helium chamber. The x-ray light path is indicated with a red line.

Fig. 4: Element spectra for Roveredo samples of 2005-12-09, 1700 CET. Each spectrum represents a particle size range (or impactor stage). Yellow: calculated peak area. Red line: continuum (background).

Fig. 5: Time series of S in ng m⁻³ for the three different size bins (blue: stage A, red: stage B, green: stage C).

Fig. 6: Trace element composition of aerosols measured in Zurich during the smog episode of January 2006.