CHEMICAL COMPOSITION AND MASS CLOSURE OF THE SIZE-SEGREGATED ATMOSPHERIC AEROSOL IN FALKENBERG (BERLIN, GERMANY) DURING LACE

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INTRODUCTION

The chemical composition of the atmospheric aerosol is a key parameter for understandding atmospheric chemical processes and aerosol climate forcing. For strongly anthropogenic influenced air masses in Central Europe there are still considerable gaps of knowledge with regards to different aerosol types and their respective size distribution. Especially size-resolved and simultaneous measurements of the main constituents carbon and ionic species are needed. To confirm reliability the efficiency of a given sampling method has to be determined. This can be done by comparing the mass size distributions determined by gravimetric analysis with the number-derived mass size distribution.

METHODS

During the Lindenberg aerosol characterisation experiment (LACE) in Falkenberg in July/August 1998, two 5-stage low pressure cascade impactors (Berner-type) were used to determine the mass and chemical composition of the atmospheric aerosol. Both weighing and sampling were performed at 60% relative humidity. The time resolution was approximately 12 h.

One impactor (Aluminum-foils) was used to determine carbon by a thermographic method. Two steps (590 °C N_2 ; 650 °C O_2) were chosen to separate volatile (organic) carbon and nonvolatile (elemental) carbon.

The second impactor equipped with Tedlar foils was used to determine ionic species. The foils were leached in 1.5 ml deionised water and analyzed for anions by capillary electrophoresis using indirect UV-detection. Concentration of cations were determined by ion chromatography.

Number size distributions of dry particles were measured with a DMPS system and converted to mass size distributions using hygroscopic growth factors obtained by means of a HTDMA and an estimated density.

RESULTS

Since air masses changed several times during the field campaign the particle mass concentration varied between 2.3 μ g/m³ (Dp < 3.5 μ m) for air masses from the North and 19.3 μ g/m³ (Dp < 3.5 μ m) for air masses from west. Primary calculations of the mass concentration derived from number size distributions show about 33 % higher values compared to the gravimetrically determined for Dp < 3.5 μ m. This is within the uncertainties of 20 % for each method. Anyway, there are several possible reasons to explain these differences: (i) losses of particles during impactor sampling due to evaporation of volatile species such as ammonium nitrate or (ii) a density (1.4 g/cm³ at 60 % RH) estimated too high. The chemical composition depends strongly on air mass history. Fig. 1 on the right hand shows an example for marine air (the 5-day backward trajectory originates in Canada and passes the northern part of the British

Islands). On the left hand the chemical composition of a continental air mass is presented (here the 5-day backward trajectory originates only from a few hundred kilometers eastwards in Poland). The mass distribution differs as well as, e.g., the nitrate distribution. In the marine case sea salt aerosol is abundant in the coarse mode range, which irreversible absorbs nitrate from the gas phase.



Fig. 1 Size distribution of gravimetric mass and chemical species for a continental (left part) and a marine case (right part). The legend belongs to both parts.

An important parameter of aerosol particles is the content of carbon. The following Tab. 1 shows mean values and one standard deviation of atmospheric concentrations and fractions of gravimetric mass for both volatile and nonvolatile carbon. Concentration of volatile carbon shows maximum values in the two size ranges between $0.14 < Dp < 1.2 \mu m$, whereas the nonvolatile carbon has a more distinct maximum between $0.14 < Dp < 0.42 \mu m$. The mass fraction of volatile carbon is almost independent of particle size, whereas the fraction of nonvolatile carbon decreases with particle size. Generally the variation is larger for volatile compared to nonvolatile carbon, which might be due to more differentiated sources for organic species (both primary and secondary).

	atmospheric concentration in [µg/m³]		fraction of gravimetric mass in [%]	
particle size range [µm]	vol C	nonvol C	vol C	nonvol C
0.05 - 0.14	0.12 ± 0.15	$\textbf{0.10} \pm \textbf{0.07}$	12.8 ± 10.5	13.4 ± 6.8
0.14 - 0.42	0.44 ± 0.32	0.44 ± 0.36	10.8 ± 6.1	10.0 ± 4.4
0.42 - 1.2	$\textbf{0.47} \pm \textbf{0.43}$	$\textbf{0.28} \pm \textbf{0.20}$	13.6 ± 13.6	7.7 ± 3.3
1.2 - 3.5	$\textbf{0.27} \pm \textbf{0.12}$	0.08 ± 0.06	11.9 ± 5.5	3.5 ± 2.4
3.5 - 10	0.23 ± 0.22	0.08 ± 0.12	11.1 ± 4.7	3.6 ± 2.5

Tab. 1 Concentration and gravimetric mass fraction of volatile and nonvolatile carbon (mean and one standard deviation for all 26 samples.