

Height Resolved Aerosol Chemical Composition and Sources in Western Europe based on Year-long High Resolution Aerosol Mass Spectrometry Observation

Caiqing Yan¹, Thorsten Hohaus¹, Sebastian H. Schmitt^{1,§}, Avtandil Turdziladze¹, Qi Zhang², Defeng Zhao^{1,†}, Astrid Kiendler-Scharr¹, Andreas Petzold¹ and Thomas F. Mentel^{1*}

¹Institute of Energy and Climate Research (IEK-8): Troposphere, Forschungszentrum Jülich GmbH, Jülich 52425, Germany

²Department of Environmental Toxicology, University of California, 1 Shields Ave., Davis, CA 95616, USA
[§] now at TSI GmbH, Aachen 52068, Germany

[†] now at Department of Atmospheric and Oceanic Sciences and Institute of Atmospheric Sciences, Fudan University, Shanghai 200433, China

Keywords: NR-PM₁, chemical composition, source, height stratification

Contact: t.mentel@fz-juelich.de

Introduction

Atmospheric aerosols play vital roles in air quality, climate change and human health. Comprehensive and reliable long-term observations of chemical, physical and optical characterizations of atmospheric aerosols as well as understanding of their sources, which are highly dependent on observation locations and seasons, are necessary for better assessing their effects on air quality, climate and human health. Up to date, the knowledge of long-term chemical characterization and sources of atmospheric aerosols is still limited in Germany. We conducted a year round observation with a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) at a 124m high meteorological tower at the Forschungszentrum Jülich GmbH, in western Germany, with primary goals including (1) to identify the chemical compositions of non-refractory submicron PM (diameter <1 µm, NR-PM₁), (2) to assess the sources of organic aerosols, and (3) to evaluate the height stratification influences on PM composition and sources.

Methods

The year-round comprehensive observation was conducted for an approximately full calendar year (from September 16, 2016 to September 26, 2017). Ambient atmospheric aerosols were collected sequentially at three heights (e.g., 10m, 50m, and 120m) automatically switching between the heights every 39 min. Besides PM₁ chemical compositions, a variety of parameters were measured simultaneously and continuously, including meteorological parameters such as wind speed, wind direction, temperature, relative humidity and precipitation, as well as some physical and optical parameters using a suite of state-of-the-art instrument. To ensure reliable results, several well established procedures including ionization efficiency calibration and filter measurement were routinely carried out during the observation period. AMS data was processed and analyzed with standard ToF-AMS data analysis software package SQUIRREL (version 1.60P) and

PIKA (version 1.20P). Composition dependent collection efficiency (Mensah et al., 2012) was applied. Co-located real time gas-phase CO₂ observation data was used to correct and improve the measurement of organic aerosol mass concentrations. Furthermore, positive matrix factorization (PMF) was performed to the high-resolution mass spectra using CU PMF Execute Tool (PET, version 3.05) to identify sources of organic aerosols.

Conclusions

The average mass concentration of NR-PM₁ was $5.54 \pm 3.83 \mu\text{g}/\text{m}^3$ during the whole observation period, composed of $47 \pm 16\%$ organic aerosol (OA), $20 \pm 14\%$ nitrate, $20 \pm 9\%$ sulfate, $12 \pm 4\%$ ammonium, and $1.3 \pm 0.8\%$ chloride. The compositions of NR-PM₁ showed monthly variations, with OA being a major fraction throughout the whole year especially in summer. Nitrate exhibited strong seasonality, with the highest contributions observed in winter and early spring, and the lowest in summer. The fractional contributions of particulate organic nitrate to total nitrate decreased with the increase of total nitrate mass, with the highest contributions found in summer. Observations at three heights indicated that stratification (influence of vertical mixing processes) was more pronounced in winter, with enhanced ground-level source emission contributions. PMF analysis suggested that oxygenated OA (OOA) dominated OA at the observation site, while hydrocarbon-like OA (HOA) accounted for around 20% of the measured OA mass. Biomass burning related sources were identified during the wintertime, accompanying with increased contributions of m/z 60 (C₂H₄O₂⁺) and 73 (C₃H₅O₂⁺). More detailed chemical characterization and source apportionment results will be presented and further discussed.

Mensah, A. A., Holzinger, R., Otjes, R., Trimborn, A., Mentel, Th. F., Brink, H., Henzing, B. & Kiendler-Scharr, A. (2012). *Atmos. Chem. Phys.*, 12, 4723-4742.