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## Characteristics, sources and water-solubility of ambient submicron organic aerosol in springtime in Helsinki, Finland



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## ABSTRACT

In this study the characteristics, sources and water-solubility of submicron organic aerosol (OA) were investigated in Helsinki, Finland. An Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was used to determine the submicron non-refractory aerosol components nitrate, sulfate, ammonium, chloride and organics between April 9 and May 8, 2009. The concentrations of the major water-soluble ions and water-soluble organic carbon (WSOC) were measured by a particle-into-liquid sampler (PILS) combined with a total organic carbon (TOC) analyzer and two ion chromatographs (IC) between April 25 and May 28, 2009. Parallel measurements of the submicron particulate matter (PM<sub>1</sub>), organic carbon (OC), black carbon (BC), meteorological quantities and trace gases were used to complement and validate the AMS and PILS-TOC-IC data.

Sources or atmospheric processes affecting the organic aerosol were investigated by applying the Positive Matrix Factorization (PMF) analysis to the high-resolution mass spectra of the HR-ToF-AMS organics. All together seven factors were needed to describe the variation in the obtained dataset. The factors consisted of two different types of low-volatility oxygenated OA (LV-OOA), local and long-range-transported (LRT) biomass burning OA (BBOA), semi-volatile OA (SV-OOA), hydrocarbon-like OA (HOA), and one local source (coffee roastery). These factors were interpretable and could be connected to specific sources or chemical characteristics (biomass burning, traffic, biogenic emissions, oxidized long-range-transported aerosols, marine-processed aerosols and nearby industrial activity) of ambient aerosols. In order to study the organic fraction and PMF factors further, the elemental ratios OM:OC, O:C, H:C and N:C were calculated. The value of the OM:OC ratio varied between 1.4 and 2.1. A high OM:OC ratio (1.5–2.1) was observed for the highly-oxidized and water-soluble fraction, whereas this ratio was clearly lower (1.2–1.4) for local and fresh sources such as traffic. Two different factors representing local and long-range-transported biomass burning were observed. Local biomass burning emissions had a lower OM:OC ratio, indicating that this factor was less aged and had a different source area compared with the LRT BBOA. The water-solubilities of the OA factors were studied by investigating the

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correlation between these factors and WSOC and by reconstructing the concentration of water-soluble particulate organic matter (WSPOM) from the OA factors. The reconstructed WSPOM had a good correlation with the measured concentration of WSPOM.

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## 1. Introduction

Organic aerosol (OA) comprises a large fraction of the ambient submicron aerosol mass and has therefore a significant impact on climate, visibility and human health (e.g. Kanakidou et al., 2005; Jimenez et al., 2009; Heald et al., 2011). Due to its complex nature, the knowledge about the sources, behavior and chemical composition of the organic fraction is limited. The compounds in the organic fraction are typically divided into primary and secondary organic aerosol (POA and SOA, respectively) according to their origin. POA refers to organic compounds that are directly emitted in a particulate form or vapors that condense onto particles without undergoing gas-phase chemistry, whereas SOA is formed in the atmosphere by gas-to-particle conversion. The fact that the OA consist of thousands of different compounds that continually change both chemically and physically depending on atmospheric conditions (Robinson et al., 2007; Hallquist et al., 2009; Jimenez et al., 2009) makes the characterization of OA challenging.

In order to understand the atmospheric behavior of OA and to determine its health and climate effects, the chemical composition of OA must be known. During the last decade, significant progress has been made in characterizing the organic compounds of atmospheric aerosols on a molecular level. Individual compounds have been shown to be tracers of specific sources, so measuring such tracer compounds makes it possible to get detailed insights into aerosol precursors and aerosol formation processes (Hallquist et al., 2009). The major tracers for biomass burning, biogenic SOA or marine aerosols have been chemically characterized (e.g. Simoneit et al., 1999; Phinney et al., 2006; Hallquist et al., 2009 and references therein; Yasmeen et al., 2011; Zhang et al., 2011 and references therein). However, a full and extensive chemical characterization of organic fraction based on offline sampling and chemical analyses is usually not feasible. Therefore, other approaches have been applied to gather information on the organic fraction of the atmospheric aerosol. A large number of high-time-resolution, online devices, such as the Aerodyne aerosol mass spectrometer (AMS), semi-continuous EC/OC carbon aerosol analyzer and particle-into-liquid sampler (PILS) coupled with either ion chromatographs (IC) or total organic carbon (TOC) analyzer, have been developed and used intensively during the last decade. Compared with offline sampling and subsequent chemical analyses, online methods typically offer artifact-free data with high time resolution. In addition, time-resolved data enable the chemical characterization of OA during fast processes, including different combustion processes (Liu et al., 2011) and evolution and aging of aerosols (Zhang et al., 2007; Jimenez et al., 2009; Heald et al., 2010; Morgan et al., 2010a,b; Ng et al., 2010; Hennigan et al., 2011; Ng et al., 2011a).

The atmospheric organic aerosol can be divided into a water-soluble and water-insoluble fraction. The water-soluble fraction represents the highly-oxidized and typically long-range transported aged fraction of OA. The water-solubility affects the chemical and physical properties of aerosols, such as its acidity and radiative properties as well as its ability to act as cloud condensation nuclei (e.g. Jacobson et al., 2000; Saxena & Hildemann, 1996). The main sources of water-soluble OA are secondary organic aerosol formation and biomass burning (Saxena & Hildemann, 1996; Decesari et al., 2006; Hennigan et al., 2009; Sun et al., 2011). The water-insoluble fraction often represents local or regional fresh emissions, like traffic.

The primary goal of this study was to explore the sources and characteristics of the submicron organic aerosol at an urban background station in Helsinki, Finland. The OA was measured with an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS), and the measured data were analyzed using Positive Matrix Factorization (PMF). The results obtained from the HR-ToF-AMS measurements were compared with simultaneous high-resolution measurements of water-soluble organic carbon (WSOC), trace gas measurements and meteorological quantities. To our knowledge, this is the first study in Europe where highly time-resolved WSOC and inorganic ion data have been available concurrently with the HR-ToF-AMS data, making it possible to investigate the solubility of different fractions of OA. An additional aim of this study was to investigate whether it is possible to reconstruct the water-soluble organic fraction of the aerosol based on AMS data.

## 2. Experimental

### 2.1. Measurement site

The measurements were conducted at an urban background station, SMEAR III (60° 12', 24° 57', 30 m a.s.l., Järvi et al., 2009). The SMEAR III station has been established for conducting long-term measurements of chemical and physical properties of atmospheric aerosols, trace gas concentrations, meteorological quantities and turbulent fluxes. The SMEAR III station has an air-conditioned container for scientific instruments and a 31-m-high measurement tower for flux and meteorology measurements at different heights. The site is situated at the University of Helsinki campus area about 5 km northeast from the city center of Helsinki. Next to the station there are the buildings of the Finnish Meteorological Institute

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