



Mass size distribution of carbon in atmospheric humic-like substances and water soluble organic carbon for an urban environment



Imre Salma^{a,*}, Tímea Mészáros^a, Willy Maenhaut^b

^a Eötvös University, Institute of Chemistry, P.O. Box 32, H-1518 Budapest, Hungary

^b Department of Analytical Chemistry, Ghent University, Proeftuinstraat 86, B-9000 Gent, Belgium

ARTICLE INFO

Article history:

Received 1 March 2012

Received in revised form

6 June 2012

Accepted 12 June 2012

Available online 26 June 2012

Keywords:

Cascade impactor

Inversion

Condensation mode

Droplet mode

HULIS

WSOC

ABSTRACT

Aerosol samples were collected with a micro-orifice uniform deposit impactor in an aerodynamic diameter (AD) range of 0.5–10 μm in central Budapest, Hungary for 12 days. Aqueous aerosol extracts and atmospheric humic-like substances (HULIS) were obtained from the combined aerosol samples for each impactor stage. Water-soluble organic carbon (WSOC) and carbon in HULIS (HULIS-C) were measured in the samples with a total organic carbon analyzer. The analytical data were inverted into semi-smooth mass size distributions, and modal parameters were derived. The size distributions for both WSOC and HULIS-C consisted of three peaks: a coarse mode and two accumulation submodes. The geometric mean AD for the coarse mode was 6.4 μm for both WSOC and HULIS-C. Contribution of the coarse mode to the total concentration of WSOC and HULIS-C were substantial and similar for both components, i.e., approximately 20%. The splitting of the accumulation mode into condensation and droplet submodes was explained by water processing of aerosol particles in the air. The geometric mean ADs of the condensation submode for the WSOC and HULIS-C were 0.37 and 0.31 μm , respectively, and the droplet submode appeared at 1.72 and 1.22 μm , respectively. The condensation submode was larger than the droplet submode by similar ratios of 1.7–1.8 for both WSOC and HULIS-C. The relative concentrations of the two submodes were likely influenced by local meteorology, in particular by relative humidity, pollutant gases, and water uptake properties and aging of fine particles.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction and objectives

Water-soluble chemical compounds including both inorganic and organic constituents in fine-sized atmospheric aerosol particles play an important role in the biogeochemical cycling of water and other substances (Ramanathan, Crutzen, Kiehl, & Rosenfeld, 2001; Mahowald, 2011), climate (Facchini, Mircea, Fuzzi, & Charlson, 1999), and several other environmental processes (Fuzzi et al., 2006). The effects of inorganic salts are well documented, while the organic compounds still represent a challenge. This is caused by the large number (up to several thousands), structural complexity and similarity of the molecules involved, and by the difficulties associated with the collection, separation and analytical methods available. Instead of the quantification at the molecular level, the organic matter is usually divided into groups

* Corresponding author. Tel.: +36 1 3722615.

E-mail address: salma@chem.elte.hu (I. Salma).

which are subsequently utilized to characterize the bulk organic material or its specific chemical fractions. Organic carbon (OC) and elemental carbon (EC) are the main groups. The former is frequently divided further into water-soluble and water-insoluble chemical fractions. The water-soluble organic fraction is usually quantified by water-soluble organic carbon (WSOC). Some water-soluble organics have unique features with respect to inorganic salts, e.g., they can substantially influence the surface tension of the gas-liquid interface of solution droplets in the air. It is a crucial property in cloud droplet formation and water uptake. One of the most important atmospheric surfactants are humic-like substances (HULIS; for a review, see Graber & Rudich, 2006). They are defined operationally as a subgroup of water-soluble organic matter. HULIS are regarded to be polymeric material comprised of aromatic compounds that have aliphatic chains with oxygenated functional groups. Formation mechanisms, physicochemical properties and chemical composition of HULIS and water-soluble organic matter are being studied increasingly (e.g., Decesari, Facchini, Fuzzi, & Tagliavini, 2000; Decesari et al., 2001; Decesari et al., 2006; Kiss et al., 2003; Samburova, Zenobi, & Kalberer 2005; Tagliavini et al., 2005; Salma, Ocskay, Chi, & Maenhaut, 2007; Salma, Ocskay, & Láng, 2008; Salma et al., 2010; Ziese et al., 2008; Claeys et al., 2012).

Size distributions belong to the key properties of ambient aerosol particles. They provide valuable information on emission sources, formation and transformation processes in the air, dry deposition, atmospheric residence times, and some surface-related properties. Mass size distribution of WSOC was studied in different environments (Decesari et al., 2001; Matta et al., 2003; Yu, Yang, Zhang, & Lau, 2004; Huang, Yu, He, & Yuan, 2006; Timonen et al., 2008; Aggarwal, Aggarwal, Okuzawa, & Kawamura, 2010; Lin, Huang, He, & Yu, 2010), while there is very limited information on the size distribution of HULIS (Lin et al., 2010). To follow up and complement these studies, mass size distributions of WSOC and HULIS-C were determined as semi-smooth curves in an aerodynamic diameter (AD) range of 0.5–10 μm for an urban environment (central Budapest, Hungary), their fine structure were identified, the modal parameters were derived, and the identified modes were related to production and transformation processes.

2. Methods

2.1. Collection of aerosol samples

An aerosol sample collection and measurement campaign was conducted in central Budapest (5, Rákóczi Street, latitude 47°29'37" N, longitude 19°03'38" E, altitude 111 m above the mean sea level) at a kerbside site within a street canyon from 23 April through 5 May 2002 (Salma et al., 2004, 2005). Mean traffic flow of motor vehicles in both directions at the site (obtained from loop counting) over the campaign was $2.0 \times 10^3 \text{ h}^{-1}$. The samplings and measurements were performed at a height of about 7.5 m above the street level. The location and time period were characterized by median concentrations of particulate matter (PM) mass, OC and EC of 53, 10.6 and $3.5 \mu\text{g m}^{-3}$, respectively in the PM_{10} size fraction. The corresponding median concentrations for the $\text{PM}_{2.5}$ size fraction were 22, 6.9 and $3.3 \mu\text{g m}^{-3}$, respectively.

The samples dealt with in the present study were collected with a micro-orifice uniform deposit cascade impactor (MOUDI, Marple, Rubow, & Behm, 1991). It has 11 impaction stages and a backup filter stage. The cut-off ADs for the impaction stages are 18, 9.9, 6.2, 3.1, 1.8, 1.0, 0.603, 0.301, 0.164, 0.094 and 0.053 μm . The device operates at a flow rate of 30 L min^{-1} . It had no additional inlet, and was operated with its inlet facing up. The samples were collected on ungreased 37-mm diameter Al foils (4 mg cm^{-2}) for the impaction stages, and on Whatman QM-A quartz fibre backup filters. The foils and filters were pre-heated at $550 \text{ }^\circ\text{C}$ for 24 h to remove organic contaminants prior to sampling. The samples were collected separately over daytime periods (from about 7:00 to 19:00 local daylight saving time, UTC+2) and nights (from about 19:30 to 6:30). A total of 11 samples for daytime periods and 12 samples for nights together with 6 field blank samples were obtained. The samples were placed into polycarbonate Petri-slide dishes, and were transported and stored in a freezer. Use is also made of some complementary atmospheric concentrations for comparative purposes that were obtained from collections in parallel with the MOUDI samples by a high volume (HiVol) sampler on pre-heated Pallflex quartz fibre filters. More information on the location, campaign and actual collections can be found in Salma et al. (2004, 2005, 2007) and Maenhaut et al. (2005).

2.2. Sample treatment and chemical analysis

The aerosol samples obtained on the backup filter and on the first impaction stage (with the largest cut-off diameter) were not treated because of the undefined size interval for these two stages, and large relative uncertainty due to sampling artifacts, in particular for the quartz fibre filters. The first impaction stage served as an inlet with a well-defined cut-off value. The aerosol samples collected on the other ten impaction stages were processed. Aerosol mass was obtained by weighing each impaction foil before and after sampling with a microbalance with a sensitivity of $1 \mu\text{g}$. The substrates were pre-equilibrated before actual weighing at a temperature of $20 \text{ }^\circ\text{C}$ and relative humidity of 50% for at least 24 h. Half sections of each foil were treated. The further procedure was performed eight years after their collection. It is expected that the samples can be kept in freezer for many years without substantial changes in their major properties since, e.g., HULIS belong to the most stable organic constituents of aerosol particles due to their aromaticity (Salma et al., 2008). There were 23 sections that corresponded to identical stage numbers. These sections were combined and processed together in order to increase the absolute amount of carbonaceous species of interest for sample treatment and chemical

Download English Version:

<https://daneshyari.com/en/article/4452587>

Download Persian Version:

<https://daneshyari.com/article/4452587>

[Daneshyari.com](https://daneshyari.com)