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Volatility of water-soluble organic carbon in ambient aerosols

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ABSTRACT

To examine the evaporation and its application to understand the chemical properties, formation mechanisms, and sources of the water-soluble organic carbon (WSOC) in ambient aerosols, field measurements of carbonaceous compounds in the aerosols were conducted at an urban site in a provincial city, Kofu, Japan. The WSOC in the fine particles evaporated with heating at lower temperature than that in the coarse particles, whereas the water-insoluble organic carbon (WIOC) in the fine particles remained at higher temperature compared with that in the coarse particles. In the fine particles, the WSOC would be more volatile than the WIOC. In the coarse particles, on the other hand, the volatility of the WSOC would be similar with that of the WIOC, and both would have the volatility between the fine-mode WSOC and WIOC. Significant parts of the WSOC were not evolved at 340 °C in air. Adsorption of gaseous organic compounds on quartz fiber filters influenced the measurements of the total carbon and WSOC, especially the carbon evolved at and lower than 120 °C. The evaporation of single standards of four typical water-soluble organic compounds was also examined, of which the results indicated a significant contribution of low molecular weight organic compounds to the WSOC, especially the fine-mode WSOC.

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1. Introduction

Water-soluble organic carbon (WSOC) constitutes a major fraction (up to about 30%) of atmospheric aerosol mass in many areas of the world including urban, rural, forested, and marine atmospheres (e.g., Sempéré & Kawamura, 1994; Zappoli et al., 1999; Kundu et al., 2010). Many previous studies have targeted the WSOC in aerosols due to its significant influences on the regional air quality and global climate, because the hygroscopicity and cloud condensation nuclei activity of the aerosols are critically dependent on the amount and chemical properties of the WSOC in the aerosols (e.g., Saxena et al., 1995; Matsumoto et al., 1997; Dick et al., 2000; Ervens et al., 2005). The chemical composition of the WSOC in the aerosols should be clarified in order to discuss the impacts of the WSOC on the atmosphere, but many uncertainties still remain. Although a chemical analysis of individual organic compounds that constitute the WSOC is expected to resolve these uncertainties, more than 100 organic compounds have been detected in the WSOC fraction (Facchini et al., 1999). Qualitative chemical analysis of individual organic compounds would be of limited effectiveness for understanding the chemical properties of the WSOC.

Categorization of organic compounds in the aerosols into several groups with similar chemical properties has been widely conducted in past studies. For instance, organic compounds in the aerosols have often been classified into several

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groups with similar polarities by the extraction with different organic solvents (e.g., Daisey et al., 1984; Zappoli et al., 1999). Water has also been used as a solvent that can classify the organic carbon (OC) into two subcategories; WSOC and water-insoluble organic carbon (WIOC) (e.g., Zappoli et al., 1999; Sullivan & Weber, 2006). The alkaline solubility of the OC has been examined for classifying certain parts of macromolecular organic compounds into humic acid-like substances (e.g., Mukai & Ambe, 1986; Feczko et al., 2007). Recently, categorizations of the organic compounds in the aerosols into several functional groups have been conducted by Fourier transform infrared (FTIR) spectroscopy or proton nuclear magnetic resonance (HNMR) methods (e.g., Decesari et al., 2002; Liu et al., 2012). These categorizations have significantly contributed to improving our understanding of the chemical properties, formation mechanisms, sources, and environmental impacts of the OC in the aerosols.

Volatility is also an important property for the OC in the aerosols because the volatility reflects the chemical properties, formation mechanisms, and sources of the OC. Miyazaki et al. (2007) found from their laboratory experiments using single standards of organic compounds that the volatilities of the organic compounds reflect their molecular weights; higher volatile compounds tend to have lower molecular weights. An et al. (2007) investigated the volatilities of artificially-generated aerosols in air with a thermodenuder-tandem differential mobility analyzer (TDMA) system and concluded that a large amount of secondary organic compounds can vaporize at temperatures lower than 100 °C. High volatilities of secondary OC in the aerosols were also found in the field experiments by a stepwise heating of collected aerosol samples in the air (Matsumoto et al., 2004, 2007). On the other hand, Huffman et al. (2009) investigated the volatilities of the ambient aerosols in the air using a thermodenuder-aerosol mass spectrometer system and concluded that secondary organic compounds has similar or lower volatilities compared with other organic components in the aerosols, whereas primary organic compounds from biomass burning and anthropogenic emissions would be more volatile. In their study, 50% of the oxygenated organic compounds in the aged air masses evaporated at 133–154 °C, but that of the hydrocarbon-like organic compounds evaporated at 85–94 °C. High volatility of primary organic aerosols has been also reported by other investigations (May et al., 2012, 2013; Presto et al., 2012).

Recently, a thermal/optical carbon analyzer that can measure OC and elemental carbon (EC) in the aerosols by the stepwise heating of the collected aerosol samples in He and then He/O₂ atmospheres followed by optical corrections has been used to investigate the volatility of the OC. Several heating programs have been used in previous studies. For example, in the IMPROVE protocol that is one of the most representative heating program used in the previous studies, four OC fractions (OC1–OC4) were measured at 120, 250, 450, and 550 °C in a He atmosphere, three EC fractions (EC1–EC3) were obtained at 550, 700, and 800 °C in a 98% He/2% O₂ atmosphere, and the pyrolyzed OC fraction (POC) was determined when a reflected laser light attained its original intensity after O₂ was added to the analysis atmosphere (e.g., Watson et al., 1994; Chow et al., 2004; Gu et al., 2010). These studies demonstrated that the OC in the ambient aerosols volatilized in wide temperature ranges from OC1 to OC4 and POC. These studies also found that the OC fraction evolved at each temperature step can be used for source apportionment; the OC evolved in the OC1, OC2, and OC3–4 in a He atmosphere can be attributed to biomass burning, coal-combustion, and road dust, respectively. On the other hand, the POC is expected to be derived from water-soluble polar organic compounds that are mainly generated by secondary photochemical processes (Yu et al., 2002).

Although these volatility investigations have improved our understanding of the chemical properties, formation mechanisms, and sources of the OC in the aerosols, they have been rarely applied to the WSOC in the aerosols. Yu et al. (2004) classified the WSOC extracted from ambient aerosols into five fractions of carbon that evolved at < 310 °C, 310–450 °C, 450–625 °C, 625–870 °C in a He atmosphere, and at 520–870 °C in a He/O₂ atmosphere using a thermal/optical carbon analyzer, and demonstrated that, as well as for the OC, the WSOC can volatilize in wide temperature ranges from < 310 °C to 870 °C in both He and He/O₂ atmospheres. A similar result was found by Yang et al. (2005). Interestingly, Yu et al. (2004) also demonstrated that the volatility of the WSOC changed with the particle size and sampling season, which indicated that the volatility would reflect the formation processes and/or sources of the WSOC.

The investigation of the volatility could be used to understand the chemical properties, formation mechanisms, and sources of the WSOC in the aerosols. Although, in previous studies, the volatility of the WSOC has been investigated in a He atmosphere (Yu et al., 2004; Yang et al., 2005), it should also be investigated in the air because the volatility in a He atmosphere would be different from that in the actual atmosphere and overestimate the pyrolyzed OC. In this study, we conducted field experiments for the evaporation of the WSOC in ambient aerosols by heating the collected aerosol samples in air. The influence of the adsorption of gaseous organic compounds on the quartz fiber filter for the results of the evaporation, and the evaporation of single standards of typical water-soluble organic compounds that have been widely detected in the ambient aerosols were also examined by field and laboratory experiments. The objective of this study is to improve our knowledge about the volatility of the WSOC in the ambient aerosols and its application to understand the chemical properties, formation mechanisms, and sources of the WSOC in the aerosols.

2. Experiments

2.1. Field experiments of the evaporation of particulate carbon

2.1.1. Sample collection

Samples of the ambient aerosols were collected on the building roof at the University of Yamanashi in Kofu, Japan, (about 17 m above the ground surface). Kofu is a provincial city with a population of about 200 thousand in a basin surrounded by

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