Journal of Aerosol Science 87 (2015) 102–110

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

Analysis of the mixing state of airborne particles using a tandem combination of laser-induced fluorescence and incandescence techniques

Fumikazu Taketani [a,*], Yugo Kanaya [a], Takayuki Nakamura [b], Naoki Takeda [b], Kazuhiro Koizumi [b], Noritomo Hirayama [b], Takuma Miyakawa [a,d], Xiaole Pan [a,1], Nobuhiro Moteki [c], Nobuyuki Takegawa [d,2]

[a] Department of Environmental Geochemical Cycle Research, Japan Agency for Marine-Earth Science and Technology, 3173-25 Showamachi, Kanazawa, Yokohama 236-0001, Japan

[b] Fuji Electric Co. Ltd., Fuji-machi 1, Hino-city, Tokyo 191-8502, Japan

[c] Graduate school of science, University of Tokyo, Hongo 7-3-1, Bunkyo, Tokyo 113-0033, Japan

[d] Research Center for Advanced Science and Technology, University of Tokyo, Komaba 4-6-1, Meguro, Tokyo 153-8904, Japan

ARTICLE INFO

Article history:
Received 2 March 2015
Received in revised form
22 April 2015
Accepted 1 May 2015
Available online 21 May 2015

Keywords:
Black carbon
Fluorescent particles
Mixing state
Single particles

ABSTRACT

We have developed a novel system for real-time measurement of the mixing state of aerosol particles using a tandem combination of laser-induced fluorescence (LIF) and incandescence (LII) techniques. The tandem analysis system comprises two chambers connected in series; particles are analyzed with LIF in the first chamber and LII in the second chamber. We analyzed identical particles using the two methods as judged by the time intervals of detection in the two chambers. This system provides information on the mixing state of fluorescent compounds and black carbon in single particles. Ground-based measurements of ambient particles were performed in Tokyo during October 26–29, 2012. We analyzed 43,881 particles with optical diameters greater than 0.4 μm. The fractions of particles with fluorescent composition, black carbon, and both were 14.2%, 2.3%, and 0.3%, respectively, which indicates the presence of internal mixtures of black carbon and fluorescent species in the ambient air for the first time. Mixtures of biological materials (estimated from fluorescence patterns) and black carbon were also detected. The fluorescence patterns of single particles with and without black carbon were almost identical, suggesting that particles with both black carbon and fluorescent composition might be formed by aggregation in ambient air.

1. Introduction

Atmospheric aerosols play a significant role not only in solar radiation, through scattering and absorption, but also in agricultural and public health (Finlayson-Pitts & Pitts, 2000). Aerosol particles are made up of a wide variety of components

* Corresponding author. Tel.: +81 45 778 5722; fax: +81 45 778 5706.
E-mail address: taketani@jamstec.go.jp (F. Taketani).

[1] Now at Research Institute for Applied Mechanics, Kyushu University, Japan.
[2] Now at Graduate School of Science and Engineering, Tokyo Metropolitan University, Japan.

http://dx.doi.org/10.1016/j.jaerosci.2015.05.002
0021-8502/

including carbonaceous materials such as black carbon (BC), which has light-absorbing properties, and organic carbon (OC), which has light-scattering characteristics. These components comprise major fractions of atmospheric aerosol particles (Bond et al., 2013). These varieties of atmospheric aerosols influence climate change by heating and cooling the atmosphere. It is important to understand the optical and chemical properties of aerosols to adequately estimate their present and future influence on radiative budgets.

The mixing state of single particles also significantly influences their lifetime, hygroscopicity, optical properties, and cloud nucleating properties in the atmosphere. For example, coatings on biogenic particles, which are of particular interest because of their high ice nucleating abilities and their significant contribution to ambient OC quantities (Despres et al., 2012; Huffman et al., 2013; Möhler, DeMott, Vali, & Levin, 2007; Murray, O'Sullivan, Atkinson, & Webb, 2012; Tobo et al., 2013), could lead to changes in their characteristics, such as protection of cell viability and high efficiency as cloud condensation nuclei (CCN) (Huffman et al., 2012). On the other hand, enhanced absorption of radiation by coatings has been reported for BC particles; furthermore, coated particles are efficiently removed by wet deposition compared with non-coated BC particles (Jacobson, 2001; McMeeking, Good, Petters, McFiggans, & Coe, 2011; Riemer, West, Zaveri, & Easter, 2010; Weingartner, Burtscher, & Baltenspergert, 1997; Zhang et al., 2008). Even if particles are individually pure when initially generated, there are multiple processes that take place in the atmosphere that convert an external mixture to an internal mixture, leading to complexities in estimating the influence of BC properties on climate. Although internally mixed BC particles have been studied (Adachi & Buseck, 2008; Cappa et al., 2012; Liu et al., 2013; Moteki et al., 2007; Moteki & Kondo, 2007; Shiraiwa et al., 2004, 2008; Ueda, Osada, &Takami, 2011), there are still large uncertainties relating to estimating the influence of BC properties on climate due to a lack of detailed measurements of mixing states (Cappa et al., 2012; Möhler et al., 2007).

There are a few established techniques for measuring the mixing state of BC particles. Off-line analyses using electron microscopes are suitable for determining the size, composition, and mixing state of single particles (Abel, Haywood, Highwood, Li, & Buseck, 2003; Adachi & Buseck, 2008; Chen, Shah, Huggins, & Huffman, 2006; Takami et al., 2013; Ueda et al., 2011). However, this technique has limitations, such as the difficulty of analyzing volatile samples and low quantitative capabilities. To quantify the chemical, physical, and optical characteristics of BC-containing particles, direct and on-line measurements are required. Several types of instruments have been developed for real-time measurement. Single particle aerosol mass spectrometry (AMS) with laser ionization provides details of chemical composition including carbon (Gard et al., 1997; Murphy & Thomson, 1995; Narukawa et al., 2007) but cannot provide quantitative information on BC. Single particle soot photometers (SP2) based on laser-induced incandescence can provide quantitative information on external/internal mixtures of BC and coating thickness (Laborde et al., 2013; McMeeking et al., 2011; Moteki & Kondo, 2007; Moteki, Kondo, Takegawa, & Nakamura, 2009; Moteki, Takegawa, Koizumi, Nakamura, & Kondo, 2011; Schwarz et al., 2006, 2010; Shiraiwa et al., 2004, 2008). Although this instrument can measure the coating thickness of BC particles based on the analysis of light scattering and particle incandescence, it cannot directly measure the chemical composition of the coating. Recently, an instrument combining AMS with SP2 was developed, which can measure the chemical composition of particles containing BC (Cappa et al., 2012; Miyakawa et al., 2014; Onasch et al., 2012). While this combined instrument can provide chemical information on BC coatings, it does not provide information of mixing states of individual particles. To obtain more information on the mixing states of particles, it would be better to combine other types of techniques with BC detection.

In this study, we developed a real-time tandem measurement system for suspended single particles by combining the laser-induced fluorescence and incandescence techniques. The fluorescence detection technique has the advantage of being non-destructive, allowing sequential analysis of the same particles. The fluorescence of suspended single particles has been used to identify and classify certain types of organic/biological particles (Pan et al., 2003; Pan, Pinnick, Hill, Rosen, & Chang, 2007; Pan, Pinnick, Hill, & Chang, 2009; Pinnick et al., 1995, 1998; Pinnick, Hill, Pan, & Chang, 2004; Taketani et al., 2013: Kaye, Barton, Hirst, & Clark, 2000, 2005; Kiselev, Bonacina, & Wolf, 2013; Toprak, & Schnaiter, 2013; O'Connor, Healy, Hellebust, Buters, & Sodeau, 2014; Saaria, Reponenbc, & Keskinena, 2014). OC could be directly emitted into the atmosphere from both anthropogenic and natural sources or formed via conversion of volatile precursors. Pöhlker, Huffman, and Pöschl (2012) summarized the fluorescence properties of various types of aerosol particle compositions, including both biological and non-biological compounds. Several instruments have reported real-time monitoring of fluorescence from single particles. Waveband Integrated Bioaerosol Sensor (WIBS) is capable of measuring light-induced fluorescence with two different excitation wavelengths at 280 nm and 370 nm (Kaye et al., 2000, 2005). The ultraviolet aerodynamic particle sizer (UV-APS, TSI, Inc.) (Brosseau et al., 2000) uses a pulsed ultraviolet laser (Nd:YAG) at 355 nm for excitation and measures fluorescence in the 420–575 nm wavelength range. These instruments focus on detection of biological fluorophores (tryptophan, NADPH, and/or riboflavin) with various criteria (Pöhlker et al., 2012). Therefore, on-line fluorescence detection from ambient single particles is of interest as it allows quantification of biological particles.

By detecting BC and/or biological particles based on incandescence and fluorescence signals from single particles, new insights into the mixing of BC and natural sources could be obtained. These mixed particles would have different CCN properties from original ones. Estimating the properties of these particles in the atmosphere would reduce uncertainties in the estimation of the effects of BC properties on climate. Toward this goal, this study aims to build an instrument for the detection of fluorescence and incandescence from the same single particles and to characterize the types of OC fluorophores present as well as the internal/external mixing states of BC particles to understand their optical properties and mixing processes.