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Correction in aerosol mass concentration measurements with humidity difference between ambient and instrumental conditions

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Abstract

The influence of humidity is considered on the concentration of the suspended particulate matter (SPM) measured with a β-ray counter. The humidity condition inside a small observatory where the counter is located is, in general, different from the ambient condition outside the observatory. From the measured values, the ambient SPM concentration is derived considering the hygroscopic effect of common aerosol species of sea salt (SS), $(NH_4)_2SO_4$, NH_4NO_3 and $NaNO_3$. In a case study conducted during September 2005, temperature and humidity were measured both inside and outside the observatory. The average value of the relative humidity is 48% for inside and 78% for outside, resulting in approximately 53% larger SPM mass concentration after the correction. Accordingly, the value of mass extinction efficiency, which is given by the ratio between the optically measured extinction coefficient and the mass concentration, becomes lower after the correction.

Keywords: Suspended particulate matter; Mass concentration; Mass extinction efficiency; Hygroscopic aerosol; Portable automated lidar

1. Introduction

Aerosol is an important factor in Earth's radiation budget. It is involved in the feedback to global warming and occasionally, it is an important part of the chemical deposition budget for certain chemical species in the ecosystem (Seinfeld and Pandis, 1998). Aerosol concentration depends on factors such as atmospheric conditions, geographic locations, location of sources and atmospheric cycles. The terminology of suspended particulate matter (SPM) is used to refer to particles present in the atmosphere with sizes less than 10 μm in diameter. Larger particles with sizes more than 10 μm hardly travel far distances and those that reach a human body would be filtered by our nose. Smaller particles, on the contrary, can directly enter human lungs (Wilson and Spengler, 1996). Among PM_{10} (with diameters smaller than 10 μm at 50% cut-off) and $PM_{2.5}$ particles, the latter species are well known to cause respiratory illnesses and cardiac disorders.

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A significant interaction exists between aerosols and the Earth's climate (Hobbs, 1993). The Intergovernmental Panel on Climate Change (IPCC, 2001) estimated the direct and indirect impacts of aerosols on the global radiative forcing at the top of the atmosphere to be of the order of magnitude which would almost compensate the CO_2 effect. Knowledge of variations in aerosol concentration is indispensable for understanding solar radiation transmission through the atmosphere.

Recently, we have reported long-term, air-sampling measurements in the Chiba area, about 30 km southeast of Tokyo, Japan (Fukagawa et al., 2006). As for the chemical constituents, the measurement indicates that a significant fraction of atmospheric SPM is composed of inorganic species. Ammonium sulfate aerosols alone account for almost 40% of the fine particles (with diameters $D < 2.1\,\mu m$), while sea salt (SS) makes up about 42% of the coarse particle ($2.1\,\mu m < D < 10\,\mu m$). These inorganic species are known to deliquesce in humid conditions (Tang, 1980). Hygroscopicity (i.e., water vapor affinity) of atmospheric aerosol particles is one of the key factors in defining their impacts on climate (Petäjä et al., 2004), since changes in the water content of hygroscopic aerosols can significantly change the light scattering properties of the atmosphere (Tang, 1996).

Laboratory experiments on aerosol deliquescence have been reported in several literatures. Tang and Munkelwitz (1993, 1994) and Tang (1996) reported growth, water activities, densities and refractive indices over extended concentration ranges for solution droplets containing single and mixed salt. Tang's (1996) paper reported chemical and size effects of hygroscopic aerosols on light scattering. Day and Malm (2001) pointed out the necessity for the consideration on the aerosol property changes due to relative humidity (RH) changes during the sampling process. They employed a humidifier/dehumidifier system to compare aerosol light scattering as a function of RH at different sites. Their study showed that growth curves at all the sites were similar in shape but the magnitude of growth varied from day to day. Condensation of sulfuric acid onto less hygroscopic particles is expected to increase their hygroscopicity. Petäjä et al. (2004) examined the hygroscopic and ethanol uptake properties of ultrafine aerosol particles in the Arctic air masses.

The Mie-scattering arising from these particles can be detected by lidars. In fact, when lidar backscattered signals are correlated with the SPM amount measured at the ground level, a strong correlation can often be found between the two quantities. Del Guasta and Marini (2000) showed an agreement between lidar-derived data and ground-based PM_{10} pollution data. Also, Lagrosas et al. (2005) reported the correlation between SPM mass concentration and lidar-derived extinction coefficient. These observations can lead to the calculation of a conversion factor between the backscattered lidar signal and the SPM concentration. In order to associate this correlation to the optical conditions of the atmosphere, a relevant quantity can be obtained if the extinction coefficients (expressed in units of m^{-1}) are extracted from the optical measurement and compared with the SPM concentration (in units of gm^{-3}). This conversion factor is called the mass extinction efficiency (MEE) (in units of $m^2\,g^{-1}$), and this is an essential parameter linking the mass concentration to light scattering.

The MEE parameter is often used in the study of radiative forcing, since this parameter connects the mass of the scatterers to the extinction coefficient of light. For example, Di Girolamo et al. (1999) used an MEE value of $5\,m^2\,g^{-1}$ to approximate the water content in the planetary boundary layer. Stratospheric aerosols can have MEE values ranging from 1.2 to $3.4\,m^2\,g^{-1}$ at high altitudes (15–20 km) (Di Girolamo et al., 1999). Husar and Falke (1996) showed that the MEE for fine particles ($PM_{2.5}$) varied from 4 to $12\,m^2g^{-1}$ for several different sites, with an average of $7.4\,m^2g^{-1}$. However, their results are higher by a factor of two compared to other literature values. Feczkó et al. (2002) measured the MEE of ammonium sulfate, $(NH_4)_2SO_4$, to be $6\,m^2g^{-1}$, and used this value in the estimation of regional climate forcing of aerosols using box model in a rural site in Central Europe. Mallet et al. (2003) calculated the MEE value for $(NH_4)_2SO_4$ in wet state to be $2.6\,m^2\,g^{-1}$. Their work also showed that 40% of the light extinction from anthropogenic sources was due to elemental carbon (EC) and organic particles. Lagrosas et al. (2005) used a lidar and a ground-based SPM counter and found that the mean value of MEE in Chiba area ranges from 4 to $12\,m^2\,g^{-1}$, where smaller and larger values occurring for size distribution dominated by coarse and fine particles, respectively. In view of the importance of obtaining MEE values that are pertinent to the ambient aerosol particles, the aim of the present study is to experimentally and