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Chemical apportionment of shortwave direct aerosol radiative forcing at the Gosan super-site, Korea during ACE-Asia

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Abstract

Shortwave direct aerosol radiative forcing (DARF) at the surface as well as aerosol optical depth (AOD) were estimated and chemically apportioned on the basis of ground-based aerosol and radiation measurements at the Gosan super-site in Korea during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) in April 2001. An aerosol optical model and a radiative transfer model (RTM) were employed to calculate the aerosol extinction coefficient and radiative flux at the surface, respectively. The calculated scattering and absorption coefficients for $D_p < 10\,\mu m$ aerosols agreed well with measured scattering and absorption coefficients with root mean square errors (RMSEs) of 23.6 and $3.0\,Mm^{-1}$, respectively. The modeled direct and diffuse irradiances at the surface were also in good agreement with the measured direct and diffuse irradiances. In this study we found that the 17-day mean aerosol radiative forcing of $-38.3\,W\,m^{-2}$ at the surface is attributable to mineral dust (45.7%), water-soluble components (sum of sulfate, nitrate, ammonium, and water-soluble organic carbon (WSOC)) (26.8%), and elemental carbon (EC) (26.4%). However, sea salt does not play a major role. For the cases of Asian dust and smoke episodic events on 26 April 2001, a diurnal averaged forcing of $-36.2\,W\,m^{-2}$ was contributed by mineral dust ($-18.8\,W\,m^{-2}$), EC ($-6.7\,W\,m^{-2}$), and water-soluble components ($-10.7\,W\,m^{-2}$). The results of this study suggest that water-soluble and EC components as well as a mineral dust component are responsible for a large portion of the aerosol radiative forcing at the surface in the continental outflow region of East Asia.

Keywords: Surface aerosol radiative forcing; Extinction; Closure study; Chemical apportionment; Aerosol; ACE-Asia

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1. Introduction

Aerosols have a direct radiative forcing because they scatter and absorb solar and terrestrial radiation in the atmosphere (Yoon and Kim, 2006). Aerosols also alter the formation, lifetime, and precipitation efficiency of liquid-water, ice and mixed-phase clouds, thereby causing an indirect radiative forcing associated with these changes in cloud properties (Intergovernmental Panel on Climate Change (IPCC), 2001). The quantification of aerosol radiative forcing is more complex than the quantification of radiative forcing by greenhouse gases because aerosol mass loading and chemical composition are highly variable in space and time (IPCC, 2001; Yoon and Kim, 2006). Quantifying and assessing the climatic impact of atmospheric aerosols require knowledge of their physical, chemical, optical, and radiative properties, as well as of their spatial and temporal variability (Penner et al., 1994; Huebert et al., 2003; Yoon et al., 2005).

During the last decade, there has been a remarkable improvement in the estimation of direct aerosol radiative forcing (DARF) by means of box models (Charlson et al., 1992; Penner et al., 1994; Nemesure et al., 1995), various measurements at the surface and onboard aircraft (Bush and Valero, 2003; Xu et al., 2003; Conant et al., 2003; Markowicz et al., 2003; Won et al., 2004; Yoon and Kim, 2006), remote sensing by satellites (Christopher and Zhang, 2002; Zhang et al., 2005), and chemical transport models (CTMs) (IPCC, 2001; Takemura et al., 2003; Yu et al., 2004; Yoon et al., 2005). However, there is still a large degree of uncertainty in the aerosol radiative forcing estimation because not only the physico-chemical properties of aerosol particles are not well established but also the complex interactions between atmospheric aerosols and radiation are poorly understood (Penner et al., 1994; IPCC, 2001). The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) campaign was designed to characterize the physical, chemical, and optical properties of aerosols in the atmosphere of the East Asian region and estimate their effect on regional radiative imbalance (Huebert et al., 2003).

Estimation of the light-extinction, optical depth, and radiative forcing due to atmospheric aerosols with chemical apportionment is important toward quantitatively assessing the aerosol impact on visibility degradation and reduction of the incoming solar radiation at the surface (McInnes et al., 1998; Bush and Valero, 2003) and for creating optimal control strategies to mitigate adverse aerosol effects on climate. Up to now chemically specified estimation of aerosol radiative forcing has mainly relied on CTMs (IPCC, 2001; Takemura et al., 2003; Yu et al., 2004; Yoon et al., 2005). In this paper, we present results of aerosol optical closure and radiative forcing studies, performed using aerosol chemical, physical, and optical measurements at Gosan during ACE-Asia together with an aerosol optical model and a radiative transfer model (RTM).

2. Experiments

2.1. Aerosol optical properties and atmospheric radiation measurements

Aerosols and atmospheric radiation measurements were performed at the Gosan super-site, South Korea (126°10′E, 33°17′N, altitude: ~50 m above mean sea level), which was the most heavily instrumented ground site during ACE-Asia (Huebert et al., 2003). Aerosols were sampled using an inlet with a height of ~10 m above ground level. Sample air was heated to achieve a relative humidity (RH) less than 50%. An impactor system was used to enable measurements of two aerosol populations: particles with aerodynamic particle diameters (D_p) less than 1.0 and less than 10.0 μm. Aerosol light scattering (σ_{sp}) and absorption (σ_{ap}) coefficients at 550 nm were measured using a TSI Model 3563 nephelometer and a Particle Soot Absorption Photometer (Radiance Research Inc.), respectively. The uncertainties in the 1-min average optical data are ~10% for $\sigma_{sp} = 33\,\mathrm{Mm}^{-1}$ and ~28% for $\sigma_{ap} = 5\,\mathrm{Mm}^{-1}$ (Kim et al., 2005a).

Direct and diffuse solar irradiances at the site in the broadband wavelength range (0.285–4.0 μm) were measured using a pyrheliometer (Eppley NIP) and shaded pyranometers (Eppley PSP), respectively. Thermal offset errors in the diffuse PSP were corrected. The radiative flux measurements are accurate within approximately 10 W m^{-2} for the 1-min averages recorded at a sampling rate of 1 Hz. In addition, spectral aerosol optical depths (AODs) at 368, 412, 500, 610, 675, 778 and 862 nm were determined once per minute by two Carter–Scott sunphotometers with an accuracy of ~0.02 (Kim et al., 2005b). Cloud screening of the solar radiation data was accomplished by examining the stability of the continuously recorded solar signal so