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Elemental composition of different air masses over Jeju Island, South Korea

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ABSTRACT

We investigated the characteristics (concentrations and compositional changes) of atmospheric elements in total suspended particulates through source-receptor relationships using cluster analyses to classify air mass back-trajectories arriving at Gosan, Jeju Island, South Korea, from October 2003 to December 2008. Five trajectory clusters were chosen to explain the transport regimes. Continental outflows of natural and anthropogenic aerosols from Asian dust source regions and eastern China during the colder period could increase element concentrations at Gosan. Elemental levels at Gosan decreased in air masses that passed over marine regions (East China Sea, Pacific Ocean/southern side of Kyushu Island in Japan, and East Sea/southern side of South Korea) during the warmer rainy period due to lower source intensity and dilution by the marine air mass. Anthropogenic pollutants were often major components in air masses passing over marine regions. Air mass characterization by elemental concentration and composition revealed that enrichment by non-sea-salt sulfur in the air mass originated from eastern China, indicative of the main sulfur emitter in northeast Asia. The apportionment of V and Ni by principal component analysis as a marker of heavy oil combustion suggested different residence times and deposition rates from other anthropogenic components in the air. Regionally intermediate concentrations of pollutants were found in the atmosphere over the Korean peninsula.

1. Introduction

Atmospheric transport of particulate elements has been extensively investigated for continentally bounded seas (e.g., Herut et al., 2001; Koçak et al., 2005) to evaluate the relative importance of source intensities to marine aerosol concentration and composition. In East Asia, Asian dust originating from arid and semi-arid northeastern China and Mongolia increases atmospheric aerosol concentrations, especially for components of soil origin, and significantly affects the composition of atmospheric particulate matter (Kim et al., 2003; Kang et al., 2011). During Asian dust events, anthropogenic components are found at lower altitudes (Kang et al., 2011). In addition, dust particles can be transported with primary pollutants containing particles of anthropogenic origin, such as road dust and fly ash (Guo et al., 2007; Han et al., 2007; Y.J. Kim et al., 2009). Portions of natural and anthropogenic components are subjected to long-distance transport and may affect downwind regions in East Asia (Y.-J. Han et al., 2004; Kim and Chung, 2008; J. Kang et al., 2009).

Receptor models such as chemical mass balance (CMB), positive matrix factorization (PMF), and principal component analysis (PCA) use the chemical components of aerosols collected at the receptor to infer the contribution of possible sources to the atmospheric concentration and chemical composition (Viana et al., 2006, 2008; Moon et al., 2008; Pandolfi

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et al., 2008). To identify sources, various elements, ions, and organic and elemental carbon (OC/EC) fractions were selected for the model analysis based on the most abundant parameters and typical tracers of various emission sources (Chow et al., 2004; Guo et al., 2009; Khan et al., 2010). PCA is an established and widely used receptor model that requires relatively little quantitative knowledge about source profiles, although it does require some initial qualitative information about sources in the study area, and is generally comparable with PMF and CMB results (Song et al., 2006; Pandolfi et al., 2008; Viana et al., 2008).

Atmospheric research at Gosan (Fig. 1), located on the western tip of Jeju Island, South Korea, has made important contributions to our understanding of airborne soil and pollution emissions from northeast Asia (Kim and Chung, 2008; Y.J. Kim et al., 2009; Jin et al., 2010). Gosan is a relatively clean area in terms of air quality (J. Kim et al., 2005). The application of receptor models using PMF and PCA at Gosan has revealed a mixture of various sources, such as natural emissions (e.g., soil dust, fresh sea salt, volcanic emissions) and anthropogenic emissions produced by local combustion processes (e.g., vehicles, coal/fuel oil combustion, biomass burning, incineration, metallurgical emissions) (Han et al., 2005b; Moon et al., 2008). Previous studies of aerosol sources at Jeju Island (Han et al., 2005b; J.S. Kang et al., 2009; Nguyen et al., 2009) found that major elements (e.g., Al, Fe, Ca, K, Mg) were at higher concentrations in soil dust and were composed mostly of coarse aerosol particles (>1.15 μm), in comparison with other trace elements (e.g., V, Cr, Ni, Cu, Zn, As, Pb) transported primarily from anthropogenic sources, which were predominantly fine particles (<1.15 μm). Soil components enriched the air mass passing over Asian dust source regions, whereas anthropogenic pollutants (e.g., secondary aerosols, carbonaceous materials, toxic trace metal components) likely originated from distant sources and were transported to the site from different source locations according to wind direction (Han et al., 2005a; J. Kim et al., 2005; K.-H. Kim et al., 2005; Nguyen et al., 2009). Therefore, the identification of aerosol sources by tracking air-mass pathways using an objective methodology is important.

Lagrangian trajectories have been considered in combination with observational data for a range of atmospheric species to identify potential sources [see review by Fleming et al. (2012) and studies on aerosols by Virkkula et al. (1999), Güllü et al. (2004), Kang et al. (2010), and Salvador et al. (2010)] or to establish source-receptor relationships (e.g., Y.-J. Han et al., 2004; Jin et al., 2010). Another approach using cluster analysis has been developed to determine relationships between air-mass origins, physicochemical characteristics of aerosols, and associated meteorological conditions (e.g., He et al., 2003; J. Kim et al., 2005; Kim, 2008). In this study, we applied cluster analysis to back-trajectories calculated in consecutive time steps, twice per day, during a long-term observation period (>5 years) to determine the regional contributions of natural and anthropogenic emissions that affect the elemental characteristics (atmospheric concentrations and composition changes) of total suspended particulates (TSPs) at the Gosan site. For this purpose, we considered the application of PCA for the

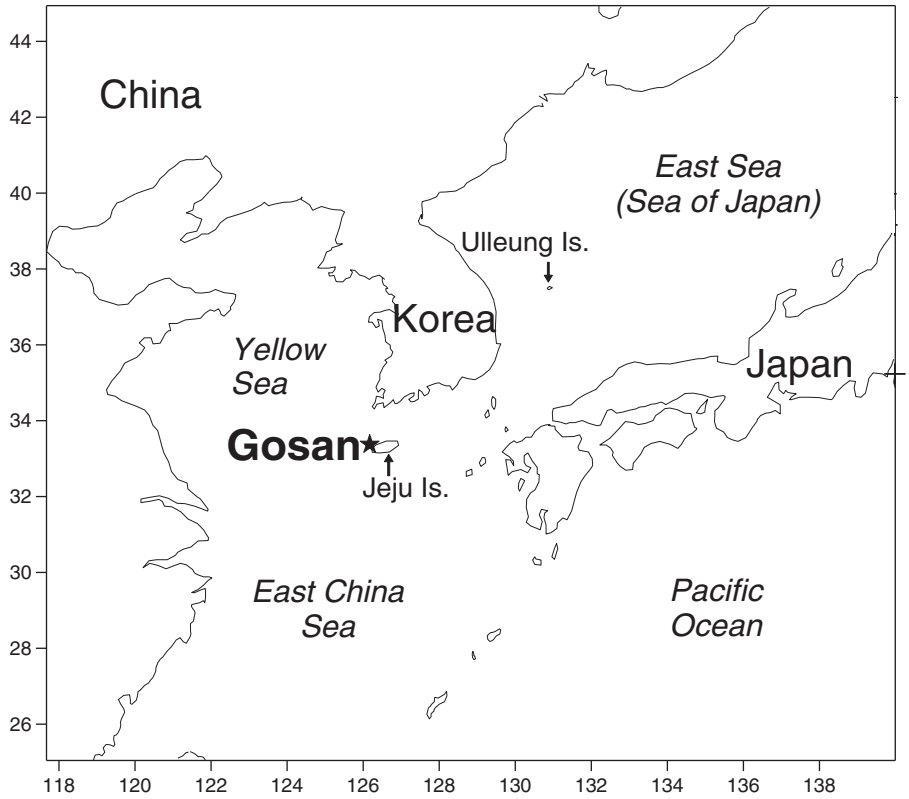

Fig. 1. Map showing the location of the aerosol sampling site at Gosan on Jeju Island, South Korea.