



Effects of Asian dust on the atmospheric input of trace elements to the East China Sea



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ABSTRACT

The long-range transport of Asian dust (AD) can carry large quantities of trace elements to the marginal seas of the Northwest Pacific, and the deposition of soluble fractions of aerosol elements may significantly impact the primary productivity and ecosystem structure. Samples of total suspended particulates (TSP) and size-segregated aerosols were collected synchronously at Huaniao Island in the East China Sea (ECS) between April 2010 and March 2011 and then analyzed for the total and soluble fractions of 14 trace elements. Twelve AD events were identified in the spring and autumn of the sampling year based on official records of AD storms from the China Meteorological Administration and verified by the high Al concentration in the aerosol samples. The dry deposition flux of the total or soluble fraction of each analyzed aerosol element was estimated using its concentration in the TSP samples and dry deposition velocity derived from the size-segregated samples. The dry deposition velocity varied from 0.7 cm s^{-1} for total Pb to 1.5 cm s^{-1} for total Al based on the mass median diameters (MMD) of 1.6 and $4.9 \mu\text{m}$, respectively, of associated particles. During AD events, the mean concentrations of the total and soluble fractions of Al, Fe, Mn, P and Ba increased significantly and led to enhanced dry deposition fluxes of these elements to the ECS (over a factor of 3 for the total and over 30% for the soluble fraction). The concentrations of the soluble fractions of anthropogenically derived elements (As, Cd, Cu, Pb, V and Zn) during AD events were lower than at other times and resulted in lower dry deposition fluxes. Asian dust events were associated with higher than average dry deposition fluxes of soluble Fe and P and lower fluxes of soluble Cu and Zn.

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1. Introduction

The long-range transport of dust carries large amounts of continental materials to the open ocean (Duce et al., 1991), which may lead to changes in primary productivity with potential implications for the oceanic carbon cycle (Mahowald, 2011). Transition metals such as Fe, Mn, Co, Ni, Cu and Zn are essential nutrients for marine phytoplankton (Butler, 1998; Whitfield, 2001), and atmospheric deposition may be an important source of these elements to surface seawater (Chen et al., 2008). Specifically, the atmospheric input of Fe has been suggested as a factor that controls primary productivity in high nitrate low chlorophyll (HNLC) regions of the ocean as well as a critical nutrient for nitrogen fixation in oligotrophic seas (Martin and John, 1990; Falkowski, 1997). Other elements such as Mn (de Baar et al., 1990) and Zn (Bruland, 1989) may also be limiting factors for phytoplankton growth in certain situations. High deposition of aerosol Cu may result in toxicity effects in specific microalgae with implications for the structure of phytoplankton communities (Paytan et al., 2009; Jordi et al., 2012).

The soluble fractions of aerosol elements are believed to be the form that can be directly assimilated by marine phytoplankton. However, there are many factors that may affect the solubility of various elements in aerosols, including their source, associated minerals or compounds, particle size, presence of organic acids, atmospheric processing, etc. (Chen and Siefert, 2004; Mackie et al., 2005; Aguilar-Islas et al., 2010). Differences in leaching methods also contribute to the reported variability in solubility (Chen and Siefert, 2003; Buck and Paytan, 2012; Morton et al., 2013). For example, previous studies reported a range of solubilities for Fe in marine aerosols between 0.01% and 80% (Mahowald et al., 2009; Baker et al., 2006). The atmospheric dry depositions of soluble elements can be estimated by multiplying the concentrations of soluble elements in aerosols by their deposition velocities. However, assuming a constant deposition velocity for all of the elements associated with coarse or fine particles may overestimate or underestimate the dry deposition fluxes.

The East China Sea (ECS) is one of the largest continental shelves in the world, and it is located in the western North Pacific between 25°N and 32°N . This area is heavily impacted by Asian dust (AD) storms that originate primarily from the Gobi Desert, Taklimakan Desert and Loess Plateau. During AD events, large amounts of dust particles are injected into the North Pacific, particularly during late winter and spring (Zhang et al., 1997). The ECS is also influenced by anthropogenic air

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pollution as a result of its proximity to East Asia and the increased numbers of automobiles, industrial development and urbanization in this region (Kaneyasu et al., 2000; Matsumoto et al., 2004). It has been suggested that the high deposition of aerosol constituents, such as trace elements, during AD storms may be one of the factors triggering phytoplankton blooms in the surface ocean of the ECS (Gao et al., 1997; Park and Dam, 2010). Indeed, the timing of AD storms appears to coincide with the occurrences of red tides in the ECS and adjacent oceans (Shi et al., 2012; Tian and An, 2012). Nevertheless, little information has been reported on the atmospheric dry deposition fluxes of the soluble fractions of trace elements over the ECS, particularly during AD events.

This study investigates the frequency of AD storms and their impact on the concentrations, solubility and dry deposition fluxes of trace elements over the ECS; such studies may be helpful for understanding the linkages between AD events and subsequent phytoplankton blooms.

2. Sampling and analytical method

2.1. Aerosol sampling

Aerosols were sampled at Huaniao Island (30.86°N, 122.67°E) in the western ECS (Fig. 1) from April 2 to 30, 2010 (spring), July 29 to August 27, 2010 (summer), November 12 to December 11, 2010 (late autumn) and March 4 to 30, 2011 (early spring). In total, 62 total suspended particulates (TSP) and 24 sets of size-segregated aerosol samples were collected synchronously using a high-volume air sampler (1130 L min⁻¹, Thermo Scientific) and 9-stage cascade impactor (28.3 L min⁻¹, Model FA-3, Anderson Company), respectively. The cascade impactor separates aerosols into nine size bins: 9.0–100 μm (stage 0), 5.8–9 μm (stage 1), 4.7–5.8 μm (stage 2), 3.3–4.7 μm (stage 3), 2.1–3.3 μm (stage 4), 1.1–2.1 μm (stage 5), 0.65–1.1 μm (stage 6), 0.43–0.65 μm (stage 7) and 0–0.43 μm (backing filter). The backing filter was not installed and the missing 0–0.43 μm size-fraction may significantly decrease the concentrations of pollution-related elements (V, Zn and Pb, Morton et al., 2013). However, the elemental concentrations reported in this study are derived from TSP samples, and the size-segregated sample measurements are only used to obtain the size distribution to calculate the dry deposition velocity of each element. It has been suggested that the deposition velocity is controlled primarily by aerosol particles greater than 5 μm, and it could be an order of magnitude higher for stage 0 particles compared to the 0–0.43 μm size-fraction (Chen et al., 2006). Therefore, the lack of backing filter should have a limited impact on the estimate of dry deposition fluxes for anthropogenic elements.

Each sample was collected over 24 h (spring and early spring) or 48 h (summer and late autumn) for TSP and 72 h for the size-segregated samples. Whatman-41 filters (Whatman Inc., Maidstone, UK) were

used for the high-volume sampler (20.3 cm×25.4 cm) and cascade impactor (90 mm diameter). The filters were pre-cleaned by soaking in 3 M (GR), 0.07 M (ultrapure) and 0.02 M (ultrapure) HCl successively for 24 h for each step and rinsed 6 times with Milli-Q water after each step. The filters were dried in a class-100 clean hood and kept at a constant temperature (20 ± 1 °C) and relative humidity (40 ± 1%) for 24 h before weighing (by Sartorius 2004 MP balance with a reading precision of 10 μg). Operational filter blanks were also collected. All of the samples were stored inside a –24 °C freezer prior to chemical analysis.

2.2. Chemical analysis

The sample filter or blank filter was cut by 1/4 (cascade) or 1/32 (TSP) using an acid-cleaned ceramic knife and then placed in Teflon jars with 20 mL of Milli-Q water and sonicated for 40 min (Kersten et al., 1991). The extract (approximately 20 mL) was filtered through a microporous membrane (pore size of 0.45 μm) and acidified with ultrapure HNO₃ (GR HNO₃ purified by sub-boiling distillation) to the final concentration of 0.3 M prior to the analysis of water-soluble trace elements (WSTE). Although the solubility determined here is operationally defined, this “batch” leaching method has been previously used in other studies (Chen et al., 2006; Duvall et al., 2008; Hsu et al., 2010; Kumar et al., 2010); therefore, our data can be compared with other studies.

The analysis of total trace elements was conducted on 1/2 (cascade) or 1/16 (TSP) of each filter after a strong acid microwave digestion. The digestion was performed in a MARS Xpress microwave digestion system with 40 Teflon tubes per batch, and then 8 mL of 15 M ultrapure HNO₃ and 0.6 mL of ultrapure HF (GR HF purified by an FEP sub-boiling distiller, Shanghai Longtuo Equipment and Instrument Company) were added to each Teflon tube along with the sample. Approximately 20 mg of standard reference material (SRM, ESS-4, China National Environmental Monitoring Centre) was processed with each batch to evaluate the digestion yield. The amount of reference material was comparable to the aerosol mass collected on the filter sub-sample and sufficient for the detection of certified trace elements. During the digestion, the temperature was ramped to 120 °C at 1600 W, followed by 3 min of condensation and reflux, increased again to 180 °C and held at that temperature for 30 min. After cooling down to room temperature, the solution was transferred into acid-cleaned polypropylene jars and dried by evaporating the liquid at 80 °C. The residue was re-dissolved in 0.2 mL of ultrapure HNO₃ and diluted to a volume of 10 mL. All of the sample handling was performed in a class-100 clean bench.

The concentrations of various elements in the total and soluble fraction extraction solutions were determined by optical emission spectrometer with inductively coupled plasma excitation (ICP-OES, SPECTRO ARCOS). The average repeatability of all of the analyzed elements is approximately 6%, and the digestion recoveries for all of the certified elements range from 86 to 105%. The detection limits (DLs) are calculated as three times the standard deviation of the blank filters and converted to ng m⁻³ using the volume of the extraction solution and sampling duration and flow rate. The DLs for the total fractions of Al, Fe, Ba, Sr, Mn, P, Cu, Pb, Zn, Ni and V in the size-segregated samples are 11, 24, 0.8, 0.8, 0.4, 3.4, 0.4, 0.6, 22, 0.8 and 1.4 ng m⁻³, respectively. The DLs for the soluble fractions of Al, Fe, Ba, Sr, Mn, P, Cu, Pb and Ni in the size-segregated samples are 1.6, 1.6, 0.3, 0.3, 0.03, 1.2, 0.8, 2.0 and 0.08 ng m⁻³, respectively. For the total digestion of the TSP samples, the DLs are 18–20 ng m⁻³ for Al, Fe and Zn, 1.8–6.7 ng m⁻³ for Ti, Cu, P, Pb, Ba, Sr and As, and 0.1–0.4 ng m⁻³ for Mn, V, Ni, Cd and Co. For the soluble fractions of elements in the TSP samples, the DLs are 1.0–1.4 ng m⁻³ for Al, Fe and Zn, 0.1–0.5 ng m⁻³ for Cu, P, Pb, Ba, As, Mn and Ni, and 0.01–0.03 ng m⁻³ for Ti, Sr, V, Cd and Co.

2.3. Enrichment factor (EF) and solubility

Enrichment factor is calculated as the concentration ratio of an element (C_X) relative to the Al (C_{Al}) in aerosols compared to the average

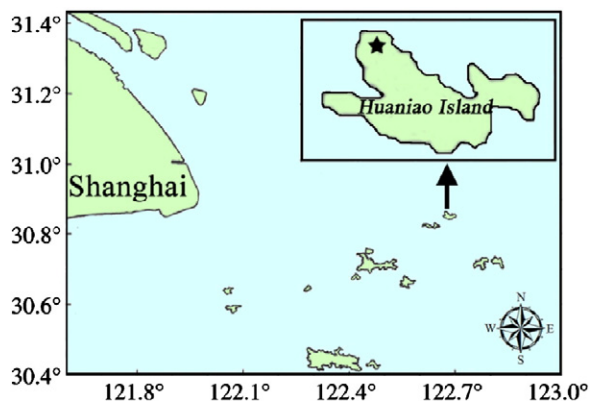


Fig. 1. The sampling site (30.86°N, 122.67°E) at Huaniao Island in the East China Sea.

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