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Baseline

Atmospheric deposition of trace elements to Daya Bay, South China Sea: Fluxes and sources

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ABSTRACT

This study was conducted from October 2015 to March 2017, with the aim of providing the first data on the fluxes and sources of wet and dry deposition of trace elements (TEs) in Daya Bay, South China Sea. Wet deposition flux of TEs was always preponderant and orders of magnitude higher than that of dry deposition owing to the high rainfall frequency in Daya Bay. The total deposition fluxes of TEs in the target area were higher than in most places worldwide, but at a moderate level within China. Wet deposition was highest in summer and lowest in winter, whereas dry deposition showed an opposite seasonal trend. The main sources of TEs in wet deposition were seasalt/dust, fossil fuel combustion, and crustal sources, and in dry deposition, they were dust/metallurgic, fossil fuel, petrochemical industry and crustal sources.

In the past 20 years, anthropogenic activities, such as industrialization and urbanization, have significantly increased trace elements (TEs) emissions and altered the airborne particles (Bacardit and Camarero, 2009; Hsu et al., 2010). Atmospheric deposition is considered as an essential process for the transfer of TEs, some of which have caused profound environmental problems and was recognized as a potential threat to human health (Galloway et al., 1982; Niu et al., 2015; Pan and Wang, 2015). Moreover, rain scavenging and gravity sedimentation, which are considered wet and dry deposition, respectively, are the main two transport mechanisms of TEs in the air to aquatic ecosystems (Connan et al., 2013; Herut et al., 2001; Lynam et al., 2015). For example, 26–83% of TEs inputs were from atmospheric deposition in the Black Sea (Theodosi et al., 2013), and wet deposition has contributed 20–70% of the total input of TEs (from riverine, direct and atmospheric inputs) on the coast of Northern France (Deboudt et al., 2004). Atmospheric deposition of TEs contributes significantly to the biogeochemical cycle of coastal waters (Duce et al., 1991).

TEs from dry and wet deposition have been proven a significant source of TEs in coastal waters (Chen et al., 2008). Because of the dynamic nature of the atmosphere, TEs in aerosol particles could be deposited from areas that are adjacent or remote to the initial sources. Natural sources of TEs include mineral dust, crustal species, and biomass burning, whereas anthropogenic sources are mainly fossil fuel combustion, incineration and metallurgical industries (Pacyna and Pacyna, 2001). TEs including Na, Mg, Al, K, Ca, Mn, Fe, and Cr are associated with natural emissions. Elements like Cr, Ba, Mo, and Cu are typically associated with fossil fuel combustion, e.g., motor vehicle emissions and coal combustion, whereas Fe, Pb, and Zn are mainly from municipal waste incinerators (Adgate et al., 2007; Marett, 2003)., A recent study found that marine primary productivity was newly discovered as an indirect source of TEs (Se, V, Co, Ni, Zn, and Al) in the marine environment (Blazina et al., 2017). However, most of the TEs deposited to the marine or terrestrial environment are emitted from a combination of different sources (Polidori et al., 2009). Among these elements, some, e.g., As, Cd, Ni, Cr (hexavalent chromium (VI)), Pb, and Co, can be accumulated by marine organisms and thus cause harm to human beings through seafood consumption (Abernathy et al., 2003; Lin et al., 2016).

Daya Bay, located in the northern South China Sea (SCS), has been under sustained and rapid industrialization. In the past decade, the petrochemical and aquaculture industries have developed rapidly owing to the economic development. Daya Bay is on the edge of the Pearl River Delta (PRD), which is one of the largest economic zones in China. Air pollutants can be spread by transport agents when moving atmospheric air masses pass over the PRD and petrochemical zone, and then partially deposited (rain scavenging or natural deposition) in Daya Bay. These deposition processes of both TEs and nutrients could have a substantial effect on the biogeochemistry of Daya Bay, such as algae bloom and biological cycling of elements. Wang et al. (2013) have reported that the sediments and marine organisms in the coastal area of the SCS, e.g., Daya Bay, Hong Kong, and the Pearl River Estuary,

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currently were contaminated. However, very little knowledge on the amount and sources of TEs in the aerosol particles and atmospheric deposition is documented in this area, even though Daya Bay is situated between a severely polluted area and the SCS. Because some of TEs present high toxicity and high lability, toxic TEs deposited to this embayment ecosystem have led to considerable concernowing to their transfer and accumulation in the food web. Therefore, atmospheric deposition monitoring is crucial for improving our understanding of the magnitude and spatial distribution of TEs deposition and assessing the efficacies of emission regulations (Azimi et al., 2003; Connan et al., 2013).

To our best knowledge, this study provides the first direct measurements of TEs entering Daya Bay from the atmosphere via wet and dry deposition. In this study, dry and wet deposition and aerosol particle samples were collected synchronously over one and a half years in aim to quantify the input fluxes of TEs from atmospheric deposition and to identify the possible sources of TEs in the study region.

Daya Bay is a semi-enclosed subtropical embayment located at 22.4–22.83°N, 114.50–114.89°E in the northern SCS (Fig. 1). It is one of the largest bays in the SCS, with an area of ~600 km^2. The annual temperature and precipitation in Daya Bay are 22 °C and 1948 mm, respectively, which are mainly controlled by subtropical and monsoonal climate. It is also under the influence of the East Asian monsoon system, whereby northeast winds prevail in winter and southwest winds in spring. > 10 typhoons, on average, traverse the SCS in summer and autumn annually (Huang and Guan, 2012), half of which invade or influence Daya Bay. In this area, March to May is considered spring and June to August summer, all of which are considered rainy months. September to November is considered autumn, and December to February winter, all of which are considered dry months (Fig. 2). There are petrochemical bases in the north and aquaculture areas in the northeast and southwest. Two large nuclear power stations (NPS) and the Pinghai coal-fired power plant in Daya Bay are operating in this area. Since 1985, there has been a rapid expansion of aquacultural, industrial, and agricultural activities in the area, with simultaneous developments in tourism and in harbor and highway constructions.

In this study, dry deposition particles and wet deposition rainwater were collected separately using a custom dry-wet automatic sampler (ZJC-III, Zhejiang Hengda Scientific Instruments Co., Ltd., China). This kind of equipment has been applied successfully to collect wet deposition of TEs. The sampler immediately opens its lid when it detects the rain, and closes the lid and seals the cylinder automatically once the rain event ends. Collection of dry deposition of TEs was consistent with the method deployed by Pan and Wang (2015) using a 707 cm^2 aperture, which is close to the real deposition condition due to the efficient polyurethane foam (PUF) filter. By this separate collection method of wet and dry deposition, the daily rainfall and monthly particulate dry deposition were collected in separate bucket containers. Because the sensing time of the sampler detector is when the rain begins/ceases, dry and wet deposition samples were collected separately with little mixing. The automatic dry-wet sampler (1.5 m in height) was deployed on the roof of the second-floor building (5 m above the ground to avoid collecting re-suspended particles) 50 m away from the seaside at the Daya Bay Marine Biology Research Station, Chinese Academy of Sciences (MBRS) (Fig. 1) from October 2015 to March 2017. Field blanks of wet deposition also were collected simultaneously in another pre-cleaned cylinder by rinsing with ultra-purewater when the rain event stopped at the sampling site, but the sampling duration period was only 5 min. Field blanks of dry deposition were handled identically to the samples, using a glass bucket that was placed for 5 min.

Aerosol samples (n = 109) were collected synchronously using a medium-flow aerosol particle sampler (Zhejiang Hengda™, 100 $L{\cdot}min^{-1}$) with glass fiber filters (Whatman, 0.1–100 μm). Filters were precombusted in an oven at 450 °C for 3 h and packaged it into sealed bags before being placed on the sampler. Sample durations ranged 8–24 h.

The rainwater samples were immediately stored in 100 mL wide-mouth polypropylene (PP) bottles in a freezer at − 20 °C after collection. In the lab, 10 mL rainwater samples were acidified to pH ~ 1 with 0.1 mL concentrated nitric acid (65% HNO_3, Merck, Germany) to dissolve TEs in flocculation status or associated with suspended particles. These rainwater samples were sealed and stored in the dark at 4 °C before analysis (within one month).

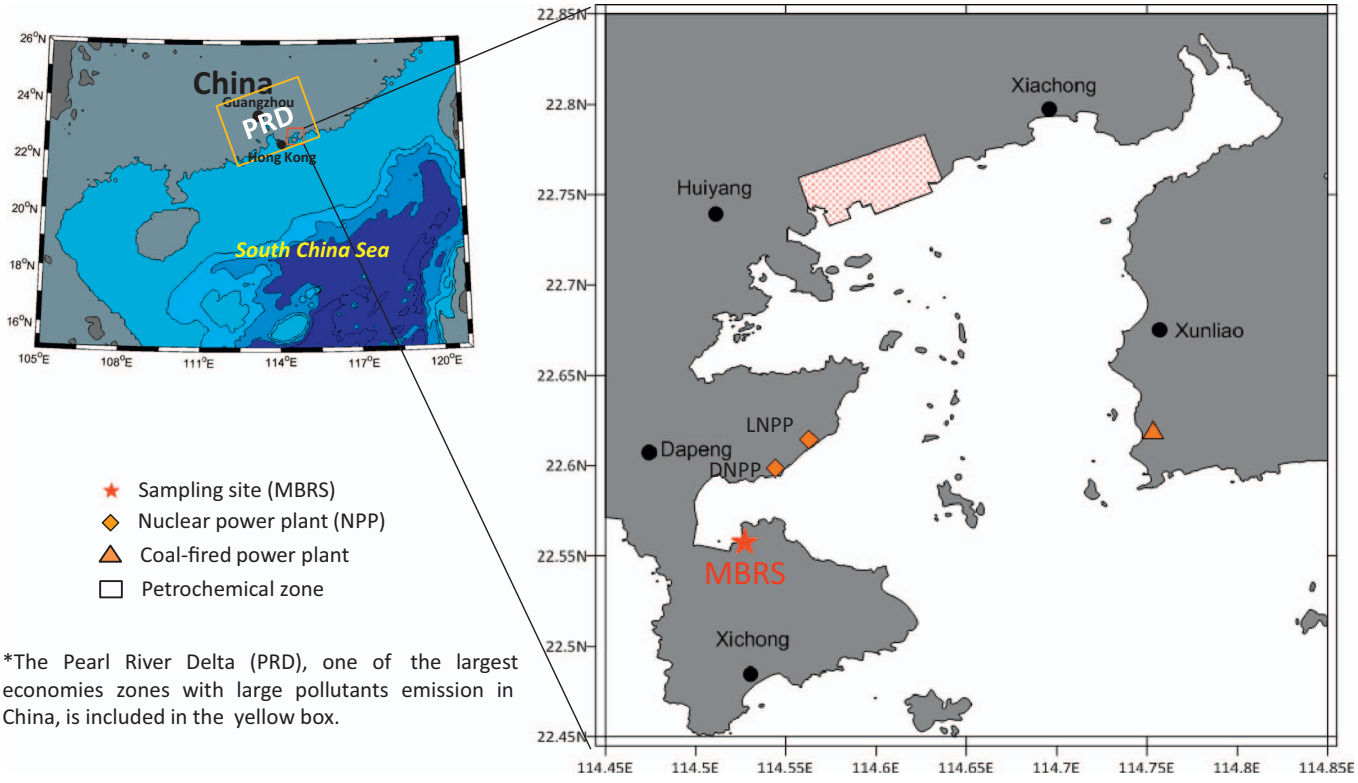

Fig. 1. Sampling site in Daya Bay.