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Atmospheric dry and wet deposition of nitrogen species and its implication for primary productivity in coastal region of the Yellow Sea, China

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HIGHLIGHTS

- Size-fractionated dry deposition fluxes of nitrogen species over the sea.
- Combined deposition (dry plus wet) of total dissolved nitrogen for the coastal waters.
- Contribution of organic nitrogen to total dissolved nitrogen.

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ABSTRACT

The size-fractionated dry deposition fluxes of aerosol particles and nitrogen species were estimated in coastal region of the Yellow Sea from July 2005 to March 2006 using size-dependent particle dry deposition velocities and measurements for size-segregated aerosol samples. During the study period, the estimated dry deposition fluxes of aerosol particles, total dissolved nitrogen (TDN), total inorganic nitrogen (TIN), dissolved organic nitrogen (DON), ammonium and nitrate were 1067.5 ± 903.3 mg m^{-2} $month^{-1}$, 15.9 ± 7.2 mg N m^{-2} $month^{-1}$, 12.6 ± 6.6 mg N m^{-2} $month^{-1}$, 3.3 ± 1.5 mg N m^{-2} $month^{-1}$, 6.5 ± 4.2 mg N m^{-2} $month^{-1}$ and 7.2 ± 4.5 mg N m^{-2} $month^{-1}$, respectively. The dry deposition flux for the coarse mode of aerosol particles and nitrogen species consisted more than 66% of the total deposition flux. This result suggests that nitrogen species in coarse particles are important in the estimation of atmospheric deposition input. The estimated wet deposition fluxes of TDN, DON, TIN, ammonium and nitrate were 154.5 ± 178.9 mg N m^{-2} $month^{-1}$, 126.9 ± 152.2 mg N m^{-2} $month^{-1}$, 24.2 ± 34.1 mg N m^{-2} $month^{-1}$, 85.4 ± 98.6 mg N m^{-2} $month^{-1}$, and 40.8 ± 56.1 mg N m^{-2} $month^{-1}$, respectively. The wet deposition contributed 65%, 72%, 55%, and 56% to the sum of the dry and wet flux of TDN, ammonium, nitrate, and DON, respectively. Inorganic nitrogen contributed much more than DON to the TDN, with a percentage of 75% and 86% in the dry and wet deposition, respectively. The dry and wet nitrogen deposition can be converted to a new primary biological productivity of 1.5–30.0 gC m^{-2} yr^{-1} in the Yellow Sea, and the nitrogen input accounts for 0.3–6.7% of the new productivity in the Yellow Sea. As an important nutrient source, the atmospheric deposition of nitrogen could have a significant influence on biogeochemical cycles.

1. Introduction

Atmospheric nitrogen deposition is an important nutritional source for ocean ecosystems (Duce et al., 1991; Castro and Driscoll, 2002; Zhang et al., 2007a). Globally, atmospheric nitrogen input is generally equivalent to riverine nitrogen input (Duce et al., 1991). However, the atmospheric deposition flux and the contribution of atmospheric input to the ocean nitrogen are found to vary in a wide range in different geographic areas. Castro and Driscoll (2002) found that total atmospheric N inputs accounted for 15–42% of the total N inputs to 10 estuaries on the east coast of the United States. The contribution from atmospheric deposition to the ocean nitrogen and productivity is high in many ocean areas (Duce et al., 1991; Zhang, 1994; Baker et al., 2007; Mackey et al., 2010). While in some ocean areas, the contribution from the atmosphere is relatively minor (Rendell et al., 1993). This difference of atmospheric

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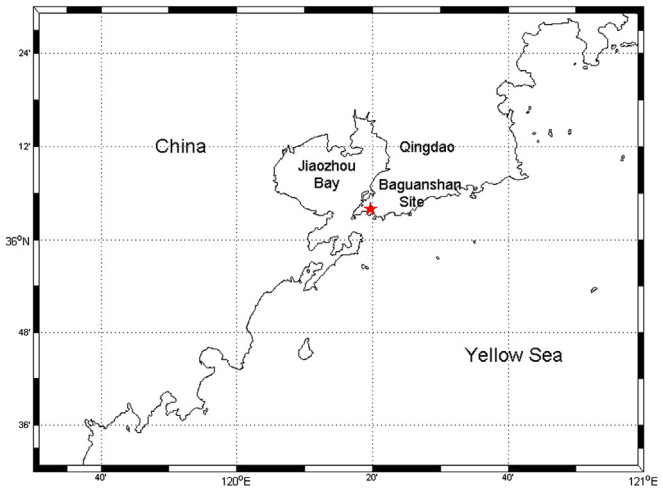

Fig. 1. Location of the sampling site.

deposition flux is commonly believed as a result of different climate and environment in different geographic areas, different estimate methods of flux and lack of observational data. For example, varying nitrogen concentrations and dry deposition velocities in different ocean areas and different seasons could lead to a significant spatial and temporal discrepancy on atmospheric N deposition. Most of the deposition fluxes are short-term measurements in nature and were obtained at a few sites, their spatial and temporal representativeness is limited. Therefore it is necessary to make an intensive study on the atmospheric deposition flux in different ocean environments to understand the contribution of atmospheric input to the ocean nutrient and marine environment.

The atmospheric nitrogen emissions increased since 1860. The atmospheric nitrogen deposition to the ocean has increased too. When an excess of nutrient-rich particles enters estuaries and oceans through atmospheric deposition, it can lead to ecological problems, such as red tides (Zhang, 1994), eutrophication (Doney et al., 2007), and changes phytoplankton growth rates (IGBP Report 50, 2004). Studies in these regions find that atmospheric inputs have a significant impact on primary productivity (Zhang, 1994; Zou et al., 2001; Nakamura et al., 2005; Zhang et al., 2010). According to the estimates of emissions of atmospheric anthropogenic nitrogen (ANN) for 2030, a ratio of 2030-to-2000 deposition rates of ANN shows up to a factor of 2 increase in Southeast Asia, and up to 30% across essentially all the mid-latitude North Pacific compared with 1860 (Duce et al., 2008). Therefore, atmospheric deposition is very important to the ocean nitrogen and marine ecosystem in these regions.

The Yellow Sea is located in the northwestern Pacific Ocean. The Yellow Sea is an important conduit through which Asian dust and its chemical components move eastward to the Pacific Ocean and beyond, which affects both regional and global biogeochemical cycles (Zhang and Gao, 2007). Prior studies have reported that atmospheric ammonium input is greater than riverine input, but that atmospheric nitrate input is less than riverine nitrate input in the Yellow Sea. The atmospheric deposition of nutrients into the coastal regions of the Yellow Sea was investigated in Zhang et al. (2007a), and between April 1999 and July 2003, the dry deposition fluxes of NO_2^- + NO_3^- and NH_4^+ were 9780 and 9300 μmol m^{-2} yr^{-1}, respectively. The wet deposition fluxes were higher than the dry deposition fluxes for these nutrients with values of 24,600 and 33,500 μmol m^{-2} yr^{-1}, respectively (Zhang et al., 2007a). Earlier studies of the Yellow Sea have reported the atmospheric deposition of nutrients, but there is little or no information provided on the combined deposition (dry and wet deposition) of total dissolved nitrogen to the coastal waters of the Yellow Sea, especially the size-fractionated dry deposition. In the present study, to evaluate the input of nitrogen into the Yellow Sea, we estimated the wet and size-fractionated dry deposition of nitrogen species (total dissolved nitrogen, inorganic nitrogen, and dissolved organic nitrogen) to the Yellow Sea's coastal waters.

Table 1
The detection limits, precision and linear ranges of the IC[a].

	Precision (RSD%)	Linear ranges (mg L^{-1})	Correlation coefficient	Detection limit (mg L^{-1})
NO_2^-	2.58	0.03–200	0.999 7	0.03
NO_3^-	0.19	0.01–200	0.999 9	0.01
NH_4^+	2.61	0.01–100	0.996 9	0.02

[a] These values were calculated by results of four repeatability experiment with sample number of 6.

2. Materials and methods

2.1. Experimental section

2.1.1. Sampling

The sampling site was located in the coastal region of the Yellow Sea (Fig. 1). The sampler was mounted at 1.5 m above the roof of an office building (36° 6′ N, 120° 19′ E, 77 m above sea level) approximately 1.0 km from shore. Aerosol samples were collected at ten-day intervals from July 2005 to March 2006. The sampling duration was 72 h for each sample. Size-segregated aerosol samples were collected using Andersen impactor samplers (Model AN-200, Sibata Scientific Technology Ltd.), and the aerosol particle sizes were fractionated into the following eight stages: >11 μm, 7.0–11 μm, 4.7–7.0 μm, 3.3–4.7 μm, 2.1–3.3 μm, 1.1–2.1 μm, 0.65–1.1 μm and 0.43–0.65 μm. The particles larger than 2.1 μm are referred to as coarse particle in the sampler stages presented later in the discussion.

As long as it was raining, the rainwater samples were collected using a stainless steel container (diameter: 30 cm; height: 20 cm) 1.5 m above the roof. And the rainfall was measured by using an automatic weather station. The rainwater samples were filtered immediately and reserved in refrigeratory before analysis.

2.1.2. Sample analysis

The aerosol samples were ultrasonically extracted with ultrapure water in an ice-water bath, and the inorganic nitrogen concentrations were determined by ICS-1000 ion chromatography (Yu et al., 2007). The average filter blank of nitrate and ammonium was 0.045 mg L^{-1} and 0.052 mg L^{-1} respectively. The detection limits, precision, correlation coefficient and linear ranges for IC listed in Table 1. The water-extracted aerosol samples were filtered and the total dissolved nitrogen (TDN) was oxide to be nitrate, and then the concentration of nitrate was determined using a BRAN + LUEBBE Auto Analyzer 3 (Shi et al., 2010). The filter blank of TDN was accounts for 0.08–3.2% of the extracted TDN in aerosol samples. The detection limit, correlation coefficient and relative standard deviations were 0.03 mg L^{-1}, 0.9995 and 2.7%, respectively (Shi et al., 2010). The concentration of dissolved organic nitrogen (DON) was obtained by subtracting TIN (Total Inorganic Nitrogen) from TDN. The nitrogen species concentrations in these rainwater samples were determined using the above methods after filtration. The average blank of nitrate, ammonium and TDN was 0.066 mg L^{-1}, 0.026 mg L^{-1} and 0.060 mg L^{-1} for rainwater sampler, respectively. The detection limit, correlation coefficient and relative standard deviations were same as aerosols samples due to the same analysis methods.