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Fluxes, seasonal patterns and sources of various nutrient species (nitrogen, phosphorus and silicon) in atmospheric wet deposition and their ecological effects on Jiaozhou Bay, North China

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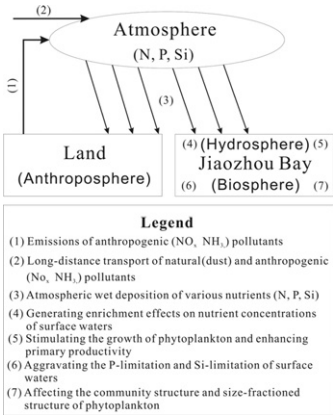
HIGHLIGHTS

- Concentrations and fluxes of various nutrient species were analyzed.
- The impacting factors of seasonal variations of nutrients in AWD were illustrated.
- DON, DOP occupied 24.3% and 62.0% of the AWD fluxes of TDN and TDP, respectively.
- AWD would aggravate the P- and Si-limitation of surface waters.
- Sudden heavy rains affect PP and community structures of phytoplankton.

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric wet deposition (AWD) is an important pathway for anthropogenic and natural pollutants entering aquatic ecosystems. However, the study on the magnitudes and ecological effects of AWD of various nutrient species (nitrogen, phosphorus and silicon) on Jiaozhou Bay is scarce. To address these issues, in this study, wet deposition samples were collected at a coastline site along Jiaozhou Bay from June 2015 to May 2016. Dissolved inorganic nitrogen (DIN, including NH_4–N, NO_3–N and NO_2–N), dissolved organic nitrogen (DON), dissolved inorganic phosphorus (DIP, i.e. PO_4–P), dissolved organic phosphorus (DOP) and reactive silicate (SiO_3–Si) were analyzed. The volume-weighted mean (VWM) concentrations of NH_4–N, NO_3–N and DON in AWD were higher compared with those of NO_2–N, PO_4–P, DOP and SiO_3–Si. The annual influxes of NH_4–N, NO_3–N, NO_2–N, DON, DIP, DOP, and SiO_3–Si via AWD were 92.8, 54.5, 0.427, 47.5, 0.274, 0.448 and 1.73 mmol·m^{-2}·yr^{-1} respectively; NH_4–N and DOP were the dominant species for N and P, and the roles of DON and DOP in AWD could not be neglected. Significant seasonal variations were observed in concentrations and fluxes of all nutrient species owing to the effects of rainfall, the intensities of local emission sources and the long-distance transports of natural and anthropogenic pollutants. The major sources of N, Si and P in AWD were agricultural activities, soil dust and a

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mixing one involving both anthropogenic and natural sources, respectively. Though AWD represents relatively low percentages of external inputs for nutrients and low contribution to primary productivity (PP) of Jiaozhou Bay, large amounts of nutrient inputs originating from sudden heavy rains may enhance PP prominently, as well as aggravate P-limitation and Si-limitation and further affect phytoplankton community structures and size-fractioned structures with the quite high DIN:DIP ratios and extremely low Si:DIN ratios in AWD.

1. Introduction

Nitrogen (N), phosphorus (P) and silicon (Si) are the major biogenic elements that support primary productivity (PP) in aquatic ecosystems. With great enhancement of human activities (Chen and Mulder, 2007; Galloway et al., 2008) and natural changes (Izquierdo et al., 2012; Kang et al., 2009; Min et al., 2011), increasing amounts of airborne anthropogenic and natural nutrients (N, P and Si) are being deposited into oceans through precipitation. Atmospheric wet deposition (AWD) has been proved to be an important pathway for nutrients being transported into marine ecosystem, particularly in the coastal waters (Ayars and Gao, 2007; Duce et al., 2008; Liu et al., 2013; Martinez-Garcia et al., 2015; Paerl et al., 2002; Song, 2010). In the Yellow Sea, AWD supplies 65% and 70% of total inputs for DIN and DIP, respectively (Zhang, 1994; Zhang and Liu, 1994). The wet deposition flux of N occupies ~70% of total inputs of total nitrogen (TN) in the coastal waters of Singapore (He et al., 2011). As a result, AWD of nutrients would strongly affect the marine ecosystem by stimulating the growth of phytoplankton, promoting carbon (C) and N fixation, and eventually enhancing PP (Duce et al., 2008; Martinez-Garcia et al., 2015; Paerl et al., 2002; Srinivas and Sarin, 2013; Zou et al., 2000), even leading to eutrophication and red tide (Bergstrom et al., 2005; Zhang, 1994; Zou et al., 2000). In the open oceans such as the tropical western Pacific Ocean, the nutrients in atmospheric deposition may sustain ~10% of PP (Martino et al., 2014). In the Yellow Sea, due to minor riverine inputs, new productivity (NP) induced by TN deposition accounts for 0.3–6.7% of PP, which is higher than the values of 1.1–3.9% found in the East China Sea (Qi et al., 2013; Zhang et al., 2010). Meanwhile, dissolved inorganic phosphorus (DIP) in atmospheric deposition could account for up to 38% of NP in the eastern Mediterranean (Markaki et al., 2003). Moreover, the increase in N availability resulted by atmospheric deposition has switched parts of the Northwestern Pacific Ocean from being N-limited to P-limited (Kim et al., 2011).

Jiaozhou Bay, located in the southeastern of Shandong Peninsula and western of the Yellow Sea, is surrounded by Qingdao, Jiaozhou and Huangdao on three sides with an area of 370 km^2 and an average depth of 7 m (Fig. 1). Jiaozhou Bay is a typical semi-enclosed bay under the interactions between natural changes and human activities in northern China, which is influenced by the natural factors such as East Asia Monsoon, and the continually enhancing anthropogenic activities in coastal regions such as terrigenous pollutant emissions and mariculture etc. (Sun and Sun, 2015). Owing to the collective influences from the rapid-advancing economy, exploded population, and the frequent haze/fogs and sandstorms, the role of AWD in the inputs of terrigenous nutrients into Jiaozhou Bay may be increasingly prominent. Particularly in the recent years, the runoff and sediment discharges of the rivers around Jiaozhou Bay have been reducing gradually (Liu et al., 2005); in addition, the discharge amounts of nutrients e.g. NH_4–N, total phosphorus (TP) derived from industrial and agricultural wastewater, domestic sewage have decreased over the past few years (Qingdao Municipal Ocean and Fisheries Administration, 2014, 2015). Thus, atmospheric deposition, especially AWD, will be more significant relative to riverine input and discharge of non-point source pollution. Hence, the study on AWD of nutrients is becoming more urgent. Moreover, reversal of eutrophication in Jiaozhou Bay requires the identification of pollution sources and the reduction of nutrient inputs. However, the data on AWD concentrations, fluxes, sources, transport and ecological effects of various nutrients species (N, P and Si) in this region are quite scarce and remain uncertain, although some similar studies have been conducted in the adjacent Yellow Sea (Chung et al., 1998; Zou et al., 2000) and Qingdao City (Zhang et al., 2011).

In this paper, dissolved inorganic nitrogen (DIN, including NH_4–N, NO_3–N, NO_2–N), dissolved organic nitrogen (DON), DIP, i.e. PO_4–P (He et al., 2011), dissolved organic phosphorus (DOP), and reactive silicate (SiO_3–Si) in AWD collected at a coastal site along Jiaozhou Bay were determined, with an aim of the analysis of seasonal patterns and impacting

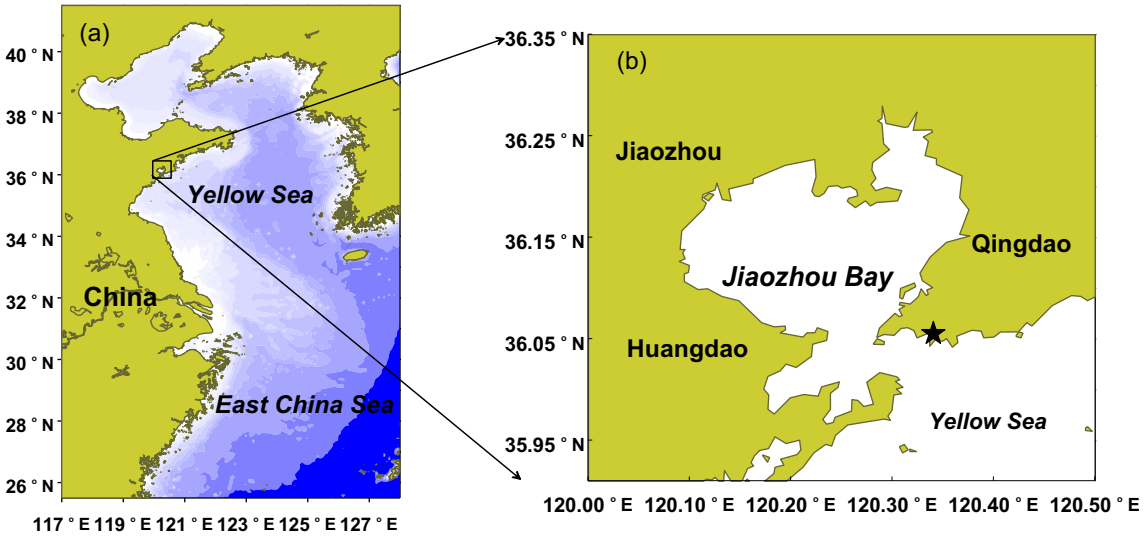

Fig. 1. The location of Jiaozhou Bay (a) and sampling site for atmospheric wet deposition (b) (★ represents the sampling site).