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### Interannual variability of estimated monthly nitrogen deposition to coastal waters due to variations of atmospheric variables model input

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#### ABSTRACT

In this work, the influence of the interannual variation of meteorological input data on the nitrogen (N) deposition estimated applying atmospheric dispersion models is discussed. As a case study, the deposition of atmospheric oxidized N coming from the NO<sub>x</sub> generated in the Metropolitan Area of Buenos Aires to waters of de la Plata River is evaluated for three years, considering high spatial (1 km<sup>2</sup>) and temporal (1 h) resolutions. The interannual variation of monthly N dry deposition is in the range 10-160%, being mostly controlled by the photochemical activity of the atmosphere and the frequency of winds towards the river. The variation of monthly wet deposition values between years is in general greater than a factor of 2. It is mainly affected by the frequency of rainy hours and the precipitation rate during offshore wind conditions, and reaches a factor of 124 as a result of the variation of these variables in a factor of 4 and 3, respectively. These results show that estimated monthly N deposition to coastal waters may vary significantly between years. The evaluation of the atmospheric N that can be transferred to coastal waters can therefore be improved considering several years of atmospheric input data.

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#### 1. Introduction

The continuous increase of the population at coastal urban zones leads to greater emissions of air pollutants, which not only affects the human health and welfare but also produces diverse impacts on the environment. In urban areas, the major atmospheric N emission source is given by fossil fuel combustion that releases nitrogen oxides  $(NO_x)$ . In an urban atmosphere, the  $NO_x$  are oxidized to form other nitrogen species such as nitrogen dioxide (NO<sub>2</sub>), gaseous nitric acid (HNO<sub>3</sub>) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) aerosol. These

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compounds can be transferred to the aquatic surface through dry and wet deposition processes (Poor et al., 2001; Pryor et al., 2001; Gao, 2002; Whitall et al., 2003; Schlünzen and Meyer, 2007; Bencs et al., 2009). In the absence of precipitation, dry deposition occurs when species are transported downward mainly by atmospheric turbulence and then absorbed or adsorbed by the surface. Wet deposition is produced when the precipitation scavenges substances being present in the air column, transferring them to the surface. These processes depend on the physical and chemical characteristics of the substance (e.g., the diffusivity of the species in air, its solubility and reactivity in water) and meteorological conditions (e.g., atmospheric stability, wind speed, wind direction, and precipitation rate), which not only affect the chemical transformation rates between the different N species and hence their ambient concentrations, but also their deposition velocities. The complex relationships between dry and wet deposition processes and atmospheric

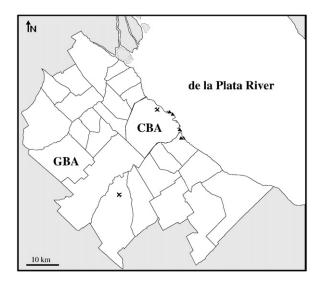
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and emission conditions make the deposition of nitrogen to coastal waters to vary considerably with time.

In previous papers (Pineda Rojas and Venegas, 2008, 2009), we estimated oxidized nitrogen (NO<sub>2</sub>, gaseous HNO<sub>3</sub> and NO<sub>3</sub><sup>-</sup> aerosol) deposition fluxes to coastal waters of de la Plata River (Buenos Aires, Argentina). The atmospheric dispersion model DAUMOD-RD (version 3) (Pineda Rojas and Venegas, 2009) was applied to area source emissions and CALPUFF model (Scire et al., 2000) to main point sources of  $NO_x$  in the Metropolitan Area of Buenos Aires (MABA). Maximum total oxidized N dry deposition fluxes  $(7-13 \text{ kg-N km}^{-2} \text{ month}^{-1})$  to the de la Plata River were within the range of values obtained in other coastal sites of the world, and N wet deposition fluxes (1-4 kg- $N \text{ km}^{-2} \text{ month}^{-1}$ ) were consistently lower than values reported by other authors given the very low frequency of rain events with offshore wind and that only source emissions in the MABA were considered. The objective of this study is to discuss the variations of monthly N deposition values estimated considering three different years of meteorological input data. Interannual variability of monthly meteorological variables in the area for the period 1999-2001 is presented. The seasonal and interannual variations of estimated monthly dry and wet deposition of oxidized N species generated from NO<sub>x</sub> emissions in the MABA to de la Plata River are discussed focusing on the influence of atmospheric variables on chemistry, transport, dispersion and subsequent deposition.

## 2. Estimation of N deposition to coastal waters of de la Plata River

The Metropolitan Area of Buenos Aires (MABA) is considered one of the ten greatest urban conglomerates in the world and the third mega-city in Latin America, following Mexico City (Mexico) and Sao Paulo (Brazil). It is composed of the city of Buenos Aires (200 km<sup>2</sup> – 2.8 million inhabitants) and the Greater Buenos Aires (3600 km<sup>2</sup> – 8.7 million inhabitants) (Fig. 1). In the urban area, there are numerous



**Fig. 1.** Area of study: waters of de la Plata River in front of the Metropolitan Area of Buenos Aires (MABA) formed by the city of Buenos Aires (CBA) and the Greater Buenos Aires (GBA); domestic airport ( $\bigstar$ ); international airport ( $\bigstar$ ); power plants ( $\blacktriangle$ ); oil company ( $\bigtriangleup$ ).

 $NO_x$  emission sources coming from road traffic, residential, commercial and low industrial activities, and aircrafts at the main airports. Moreover, the main point sources located near the coast are the stacks of four Power Plants and a large oil company. Due to the geographical location of the MABA, pollutants released to the atmosphere are transported towards de la Plata River during great part of the year. De la Plata River is a shallow river-type estuary of 327 km long and with a width varying between 2 and 227 km. In front of the MABA, the river is 42 km width and shows a large zone where depths are less than 5 m. The river constitutes the main source of drinking water for the city of Buenos Aires and surrounding areas.

To estimate the deposition of atmospheric nitrogen (N), coming from the N emitted in the MABA, to surface waters of de la Plata River, the DAUMOD-RD (version 3) model was applied to area source NO<sub>x</sub> emissions and the CALPUFF model to the emissions coming from the main point sources near the coast. Both models were applied over a surface of the river of 2339 km<sup>2</sup> (Fig. 1), considering spatial and temporal resolutions of 1 km<sup>2</sup> and 1 h, respectively. Three years (1999–2001) of hourly surface meteorological information measured in a coastal site within the domestic airport and sounding data from the site located in the international airport (Fig. 1), were used. The  $NO_x$  emission data belong to a high resolution emission inventory developed for the Metropolitan Area of Buenos Aires (Pineda Rojas et al., 2007) and the emissions from the stacks of the four Power Plants and the large oil company. The emission inventory includes area source emissions: residential, commercial, small industries, road traffic and aircrafts landing/take-off at both the domestic and the international airports. The emission factors used in preparing the emission inventory were derived considering: a) monitoring studies undertaken in Buenos Aires (Rideout et al., 2005); b) The EMEP/CORINAIR Atmospheric Inventory Guidebook (European Environment Agency, 2001); c) The US Environmental Protection Agency's manual on the Compilation of Air Pollution Emission Factors (US EPA, 1995). These factors were applied to fuel consumption, gas supply data and vehicle kilometres travelled within each grid square. Data on traffic flow and bus service frequencies was also available. Aircraft emissions were computed knowing the scheduled hourly flights, the type of aircraft, the information available on LTO (landing/take-off) cycles and NO<sub>x</sub> emission factors (Romano et al, 1999, EMEP/CORINAIR, 2001). Table 1 includes

Table 1

Annual  $NO_x$  (expressed as  $NO_2$ ) emission from the main point sources and area sources of the MABA considered for calculations.

Source category	Annual emission $(ton-NO_x yr^{-1})$
Main point sources	
4 Power plants	31484
Oil company	1794
Total point sources	33278
Area sources	
Road traffic	53883
Residential	7520
Commercial	702
Small industries	3839
Aircrafts (airports)	879
Total area sources	66823
Total annual emission	100101

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