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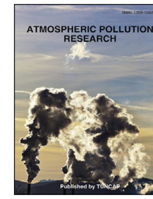

Wet deposition fluxes of atmospheric inorganic reactive nitrogen at an urban and rural site in the Indo-Gangetic Plain

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ABSTRACT

Excess nitrogen deposition is a matter of concern for sensitive ecosystems. However, understanding the sources and transport of Nr species has been a challenge due to limited observations of atmospheric deposition of the key Nr species across India. In this study, wet deposition of atmospheric inorganic Nr species was investigated during the year 2013 at two regionally representative sites: Delhi (an urban site) and Jaunpur (a rural site). These sites are located in the Indo-Gangetic Plain (IGP) region, which is one of the most populated and fertile regions of India. The average NH_4^+ concentrations in rain water were found to be 25.4 μeql^{-1} and 98.5 μeql^{-1} at the rural and urban sites, respectively, whereas average NO_3^- concentrations were 12.4 μeql^{-1} and 28.7 μeql^{-1} at the rural and urban sites (respectively). The annual average wet deposition fluxes of NH_4^+ and NO_3^- at Delhi were calculated as 10.45 and 3.05 kgN ha^{-1} yr^{-1} respectively, whereas at Jaunpur the fluxes were 3.19 and 1.56 kgN ha^{-1} yr^{-1} respectively. In order to assess the Nr deposition, our estimates showed 486% increase in NO_3^- (from 0.52 to 3.05 kgN ha^{-1} yr^{-1}) while 283% NH_4^+ (2.72–10.44 kgN ha^{-1} yr^{-1}) between 1994 and 2013 at Delhi, clearly indicating the effect of urbanization and Land Use Land Cover (LULC) change. Reduced versus oxidized N deposition contribution was also estimated. This study provides key quantitative information to support regional nitrogen budget estimates in south Asia.

1. Introduction

Nitrogen oxides (NO_x) and ammonia are the key reactive nitrogen (Nr) species in atmospheric chemistry and ecosystem productivity. Although the major source of ammonia in the atmosphere is agriculture, other sources can include industry, landfills, household products, biomass burning, motor vehicles, and even wild animals (Ianniello et al., 2010). Before 1995, the contribution of vehicles to non-agricultural NH_3 emissions was negligible, but in recent times NH_3 concentrations in urban environments have also increased due to the over-reduction of nitrogen oxide compounds in catalytic converters of automobile exhaust and industrial power stations (Sutton et al., 1995; Sutton et al., 2000). Fossil fuel emissions and intensive agricultural activities have drastically accelerated the nitrogen (N) cycle, resulting in increasing deposition rates of N species (Galloway et al., 2008; Penuelas et al., 2012; Canfield et al., 2010). Increasing Nr deposition has many adverse effects on human health, vegetation health, air quality, soil acidification, eutrophication, and most importantly global climate change (Sutton et al., 1994; Sun et al., 2004; Ianniello et al., 2011). Further, it affects the global carbon cycle (Goulding et al., 1998; Galloway et al., 2004). Once the reactive nitrogen species are released into the atmosphere, their fate is decided by several processes such as transformation, transport, and deposition. Nr species are deposited either through dry deposition or wet removal processes. Despite a long history of precipitation composition measurements (almost 150 years), the spatial and temporal variations of atmospheric Nr deposition remain unclear due to the complexity of reactions during transport and deposition (Park and Lee, 2002). Nevertheless, the wet deposition of Nr species has been studied by a number of groups as part of acid deposition studies. Available research indicated N deposition reached 70 Tg yr^{-1} N and

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averaged 5 kg ha^{-1} yr^{-1} N at the global scale (Holland et al., 1999; Galloway et al., 2004). Asia including India has the third highest N deposition rates, following North America and Europe (Holland et al., 1999; Galloway and Cowling, 2002).

The reported studies suggest that the Indo-Gangetic Plain (IGP) is a major hotspot of Nr deposition in India (Galloway et al., 2004; Kulshrestha et al., 2005; Van et al., 2011; Vet et al., 2014). The IGP is one of the most densely populated and polluted regions in the world, consequently having increasing emissions. Use of traditional biofuels for cooking and heating purposes in rural areas in the IGP leads to significant air pollution (Singh et al., 2014). One of the most powerful causes of high pollution load in the IGP is rapid urbanization, which results in high emissions of N-containing pollutants and affects levels of atmospheric N deposition (Lovett et al., 2000; Liu et al., 2008). In Europe and the United States, urban areas tend to receive higher rates of N deposition than rural areas (Lovett et al., 2000; Power and Collins, 2010). Similar to many Indian cities, Delhi (the urban site of the present study) is experiencing a high growth rate. During the last two decades (from 1991 to 2011), the population of Delhi is increased by approximately 169% (Table 1).

In order to sustain the increasing population, humans are drastically changing land use land cover (LULC) as well. LULC change has been linked to high dust fall fluxes in Delhi, ultimately affecting the air quality (Kumar et al., 2014). Urbanization and land use changes have resulted in temperature increases in the USA during the last several decades, ultimately affecting the climate (Kalnay and Cai, 2003). In China, diurnal temperature differences have decreased due to land cover change leading to change in regional climate (Zhou et al., 2004). These land use practices also affect air quality by altering emissions and changing the atmospheric conditions that affect reaction rates, transport, and deposition of pollutants (Sillman, 1999).

Delhi is facing an issue of increased traffic emissions due to a tremendous increase in the number of vehicles on the road (Goyal et al., 2013). Being capital, Delhi city has been a location of common concern but unfortunately, the city is not covered by any systematic and long term measurement network of wet deposition of reactive nitrogen species. This is also true for rural areas. Along with highly populated cities, rural sites in the IGP are important contributors to Nr emissions due to extensive agricultural practices and use of synthetic fertilizers. Hence, the present study is an attempt to measure the wet N deposition levels in one of the world's most polluted cities (Delhi) along with the measurements at a rural site in Jaunpur district of Uttar Pradesh state. This study reports the measurements of wet deposition of inorganic Nr species at an urban and a rural site in Indo-Gangetic plain, providing Nr estimates from a rather large area through these regionally representative sites. In addition, in order to observe the influence of two decades of land use change and urbanization on Nr fluxes, this study compares the present deposition fluxes of Nr species at Delhi with observations reported for 1994.

Table 1
Descriptive statistics of demographic change of Delhi between 1991 and 2011.

Area type	Census year	
	1991	2011
Population	6,220,406	1,67,53,235
Rural	452,206	4,19,319
Urban	5,768,200	1,63,33,916
Population density (person km^2)	6352	11,297
Category	1994	2013–14
Motor vehicle No.*	24,32,295	82,93,167

Source*: Transport department, Govt. of NCT of Delhi.

2. Methodology

2.1. Site description

Two sites having different land use, land cover, sources and population density located in Indo-Gangetic plains in India were selected for this study. The Delhi site is representative of urban areas whereas the Jaunpur site, in the adjacent Uttar Pradesh state, represents rural areas (see Fig. 1). Basic information regarding both the sites is given below.

2.1.1. Delhi (urban site)

In Delhi, Jawaharlal Nehru University (latitude 28°31′30″–28°33′30″ and longitude 77°9′0″ –77°11′0″) was selected as the urban representative site. According to several reports, Delhi is the most polluted city in the world (Gargava et al., 2014; Pant et al., 2015). The main sources of pollution in Delhi are thermal power plants, on-road transport, small-scale industries, and domestic cooking/heating activities (Khillare et al., 2004; Chowdhury et al., 2007; Lelieveld et al., 2001; Chelani et al., 2010; CPCB, 2010). The air quality of Delhi is most affected by vehicular exhaust (Verma and Kulshrestha, 2015; Bisht et al., 2013; Dholakia et al., 2013). The sampling site was located at the School of Environmental Science (SES) building, Jawaharlal Nehru University, New Delhi. The JNU campus lies in the southernmost section of Delhi, and is surrounded by a very small forested area.

2.1.2. Juanpur (rural site)

Mai village (25°37′23.5″N, 82°51′10.1″E), in Jaunpur, was selected for the study, which is 885 km from Delhi. The total area of the village is 617 ha, out of which 385 ha land (approximately 62%) is used for agricultural purpose. The area around the site is dominated by agricultural activities, thus it is an excellent representative location for the study of reactive nitrogen emissions from urea and DAP fertilizer applied in agricultural fields. A few industrial establishments are located at a distance of 13 km from the village including one vegetation oil production factory to the southeast (Singh and Kulshrestha, 2014).

2.2. Sample collection

Sampling was conducted at the terrace of the School of Environmental Science building (~23 m height) at the university campus in Delhi, and on a house roof (~20 m) at the rural site, Jaunpur. An assembly of polythene funnel of 20 cm diameter fitted onto 2 L capacity polythene bottle was used for rain water collection at both the locations. The sampling occurred during the southwest monsoon (SW monsoon) of 2013. The SW monsoon occurs from July through September. Monsoon rainfall accounts for about 80% of the total annual precipitation in India and is a major factor controlling water resources, agriculture, and ecosystems (Gadgil and Kumar, 2006). It is characterized by huge spatial variability, which is largely a function of topography. The samples were collected manually during rainfall events using pre-washed plastic bottles and funnels (Kulshrestha et al., 2003a). The collection assemblies were placed at about 1.5 m above the roof and were deployed as soon as the rain began, and were retrieved immediately after the rain stopped. After each sample, the bottle and funnel were cleaned with deionized water to avoid any residual contamination.

Collected samples were stored in prewashed small polythene bottles of 60 ml volume. A small amount of thymol was added to each sample to prevent biological degradation (Gillet and Ayers, 1991; Granat et al., 1992; Koshy et al., 1993; Kulshrestha et al., 2005). For each event, two samples of 30 ml volume were kept