Atmospheric Abundances of Nitrogen Species in Rain and Aerosols Over a Semi-arid Region: Sources and Deposition Fluxes

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Abstract

The temporal variation for a three-year period (2000-2002) in the atmospheric abundances of principal nitrogen species (NH_4^+ and NO_3^-) has been studied in rain and aerosols from an urban city (Ahmedabad, 23.0^oN, 72.6^oE) located in a semi-arid region of western India. Their concentrations in ambient aerosols over the annual seasonal cycle exhibit large variation [NH_4^+: < 0.001 to 1.3 (GM = 0.25 $\mu g/m^3$); NO_3^-: 0.09 to 4.4 (GM = 1.3 $\mu g/m^3$)]; with systematically higher concentrations during Nov-Feb (drier period) and relatively low during Jun-Aug (wet season). In comparison, abundances of NH_4^+ and NO_3^- in individual precipitation events (n = 91) collected during the southwest monsoon (Jun-Aug) for three years varied as NH_4^+: <1.0 to 220 (VWM: 30 $\mu eq/L$) and NO_3^-: 1.3 to 115 (VWM: 13 $\mu eq/L$). Using corresponding rainfall data, the wet-deposition fluxes of NH_4^+ during 2000, 2001 and 2002 have been derived: 364, 327 and 297 $mg/m^2/y$, respectively; which are considerably higher than the dry-deposition fluxes (16, 11 and 16 $mg/m^2/y$). The wet deposition of NO_3^- over the three years varied as 392, 569 and 487 $mg/m^2/y$ in comparison to dry-deposition fluxes averaging as 201, 134 and 137 mg /m^2/y. These results also indicate that both dry and wet deposition of NO_3^- accounts for as much as ~65% of the total inorganic-N species ($NH_4^+ + NO_3^-$), possibly mediated via in-situ chemical reactions of acidic species (NO_3^- and its precursors) with mineral aerosols. Such regional scale studies are important for quantitative assessment of atmospheric deposition of biogeochemically important trace species and bring to focus the role of semi-arid regions where current knowledge is limited.

Keywords: Dry and wet deposition; NH_4^+; NO_3^-; Aerosol; Precipitation; Semi-arid region; India.

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INTRODUCTION

Nitrogen is a major nutrient in terrestrial ecosystem and an important catalyst in tropospheric photochemistry. Atmospheric nitrogen compounds affect Earth's radiation budget, acidify ecosystems, and cause degradation and eutrophication of lakes, estuaries and coastal oceanic regions (Galloway, 1995; Galloway *et al*., 2004; Holland *et al*., 2005). Among inorganic nitrogen species, NH_4^+, NO_3^- and their precursors are the most dominant species present in the atmosphere. In the present-day scenario, the major sources of atmospheric NH_4^+, NO_3^- and their precursors (NH_3 and NO_x) are attributed to vehicular emissions, biomass and fossil-fuel burning, human and animal excreta, microbial decomposition of biomass etc. The typical residence time of nitrogen species in the atmosphere of about a week is similar to that of ambient aerosols. During their long-range transport, these species undergo a variety of physico-chemical transformation processes and are ultimately removed from the atmosphere via dry and wet deposition. The wet deposition is considered to be the major removal pathway of N-species, accounting for $> 80\%$ in many regions. However, in arid and semi-arid regions, the dry deposition can be substantial; but the relative significance of dry and wet deposition processes are poorly documented. In this context, regional-scale programs are the key components of global nitrogen budget and to predict the effects of nitrogen cycle on climate change (Galloway *et al*., 2004; Holland *et al.*, 2005). The present study reports on the atmospheric abundances of NH_4^+ and NO_3^- in rain and aerosols, their seasonal variability and deposition fluxes (dry and wet) from an urban area located in a semi-arid region of western India.

EXPERIMENT

Site description

Our sampling site (located in Ahmedabad city: 23.0^oN, 72.6^oE, 49 m asl, population ~5.5 million) represents a typical urban atmosphere of a semi-arid region in western India. To its northwest lies the great Thar Desert. In the southwest is the Arabian Sea and to its northeast is the continental area inhabited by a large human population and diverse industrial activities (Fig. 1). The contrasting wind patterns during Nov to Feb (representative of NE winds from large-scale pollution sources that affect the air quality) and May to Aug (southwesterlies transporting sea-salts) dictate the aerosol composition over the study site (Rastogi, 2005). The wind fields during Mar-Apr and Sept-Oct represent transition phase in the circulation pattern. The annual rainfall averages ~650-700 mm, all of which occurs during the SW monsoon (Jun-Aug). The monthly average temperature from Jan to Dec varies between 21 and 34^oC with relatively high

temperatures (32 to 34°C) during summer months (Apr-Jun) and low (21 to 23°C) during winter months (Dec-Feb).
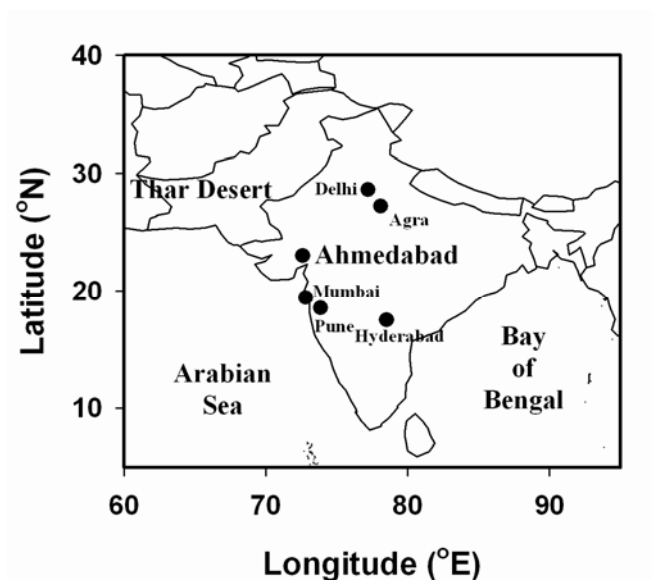

Fig. 1. Site map of Ahmedabad.

Sampling and analyses

The sampling gear for aerosol and rainwater collection was set-up at a height of ~40 m above the ground level in order to avoid near-surface local sources. Bulk-aerosols (n = 99) were collected, from Jun 2000 to Jan 2003, onto clean precombusted tissuquartz filters (PALLFLEX^{TM}, 20 × 25 cm) by operating a high-volume air sampler (flow rate: ~1.2 m^3/min). On an event basis, "wet-only" precipitation samples (n = 91) were also collected during the SW monsoon (Jun-Aug) for three successive years (2000-2002). The total rainfall during 2000, 2001 and 2002 was 730, 680 and 400 mm, respectively; all of which occurred during the SW monsoon. The rainwater samples and water extracts of aerosols were analyzed for NH_4^+ and NO_3^- along with other major ions (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-} and HCO_3^-) by standard techniques. Further details on analytical procedures are found in Rastogi and Sarin (2005) and Rastogi (2005). The cation/anion ratio (on equivalent basis) in water-extracts of the aerosols ranged from 0.80 to 1.14 (average: 1.02, sd: 0.08). The chemical data in precipitation events also follows the 1:1 trend, except in a few heavier precipitation events when the solute concentration (cation + anion) is less than 100 µeq/L.

RESULTS AND DISCUSSION

Source apportionment for NH_4^+ and NO_3^- in aerosols and rainwater

In order to decipher the dominant sources of NH_4^+ and NO_3^- in aerosols and precipitation events, a linear regression analysis was performed among various ions measured in water-extracts of aerosols and in rainwater samples (Fig. 2). We have considered nss-SO_4^{2-} as a constituent of anthropogenic origin, abundance of nss-Ca^{2+} signifies its dominant contribution from mineral dust, and that of nss-K^+ from biomass burning sources (Rastogi, 2005). Unlike rain events occurring during SW monsoon (Jun-Aug), the chemical data on aerosol samples (collected around the year) has been split into three seasons based on prevailing wind patterns (Fig. 2).

Among the water-soluble aerosol species, a significant linear co-variance ($r^2 = 0.38$) is observed between nss-SO_4^{2-} and NO_3^- (excluding the wet season data, Jun-Aug), suggesting a contribution of NO_3^- from anthropogenic sources. Furthermore, linear regression parameters for nss-K^+ versus NO_3^- scatter plot (slope = 1.54 and $r^2 = 0.45$, Fig. 2) addresses the issue that biomass burning could also be a dominant source of NO_3^- as KNO_3 (expected NO_3^-/K^+ wt ratio = 1.59, based on stoichiometry). However, a significant intercept (c = 0.98) in the above plot would imply NO_3^- contribution from anthropogenic sources as discernible from co-variance between nss-SO_4^{2-} and NO_3^- (Fig. 2). Thus, multiple sources can be invoked for atmospheric NO_3^- aerosol. The scatter plot between nss-Ca^{2+} and NO_3^- also exhibit a linear trend (except for a few samples marked by high abundance of Ca^{2+}, > 4 $\mu g/m^3$) (Fig. 2). This is attributed to neutralization reaction between HNO_3 and mineral dust ($CaCO_3$) (Laskin *et al.*, 2005; Rastogi, 2005). The outlier (Ca^{2+} > 4 $\mu g/m^3$) corresponds to aerosol samples collected during high-dust events. It is noteworthy that regression analysis of NH_4^+ with SO_4^{2-} shows a large scatter (Fig. 2); the lowest concentrations of NH_4^+ occurring during the May to Aug (a period dominated by SW winds of marine origin), and highest during Nov-Feb (NE winds transporting pollutants from large continental areas) (Fig. 3). A linear trend observable between NH_4^+ and nss-SO_4^{2-} for the data set with NH_4^+ abundance exceeding 0.5 $\mu g/m^3$ (Nov-Feb, Fig. 2) suggests its occurrence as sulphate salts. A large seasonal variation in the abundance of aerosol NH_4^+ is attributed to its partitioning between gaseous and particulate phase; the contribution from the latter phase is relatively more pronounced during winter months (Nov-Feb) under the influence of NE winds. In the case of precipitation samples, the regression analysis for NH_4^+ and NO_3^- with K^+, Ca^{2+} and SO_4^{2-} exhibits characteristics trends somewhat similar to those observed for water-soluble species of aerosols (Fig. 2).
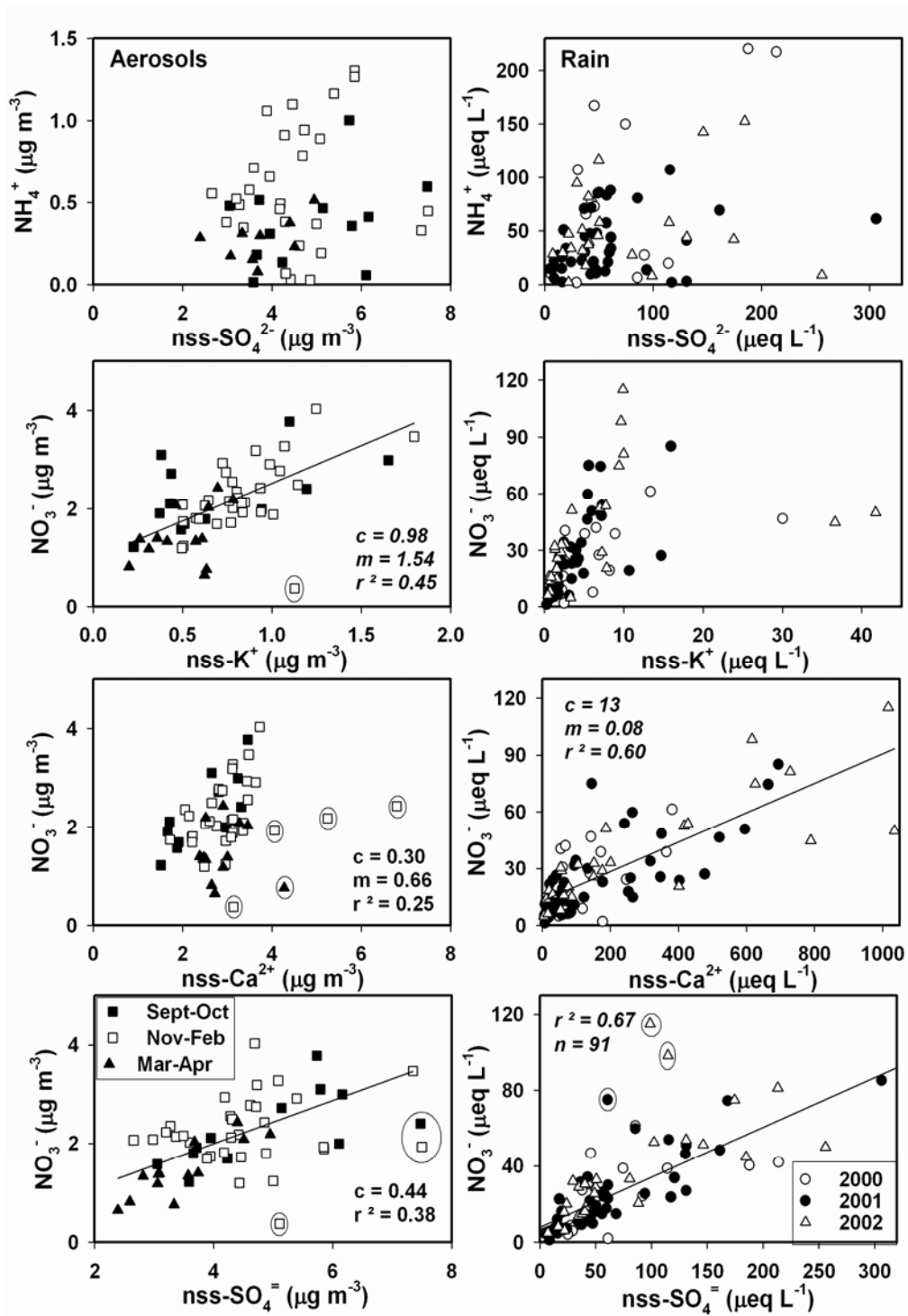
Fig. 2. Source apportionment of NH_4^+ and NO_3^- in rain and aerosols based on scatter plots with major ions. Encircled points are not included in regression analyses.
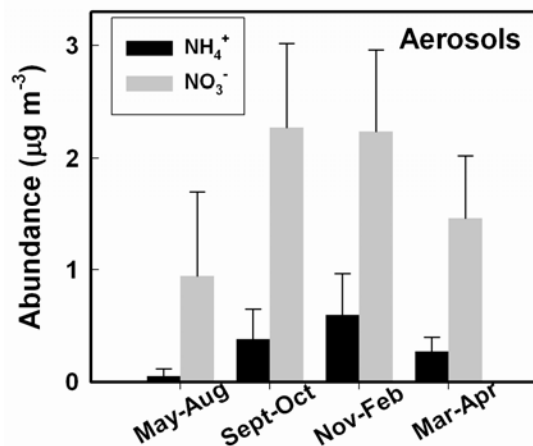
Fig. 3. Seasonal average abundances of NH_4^+ and NO_3^- in aerosols exhibits relatively high values during Nov-Feb due to dominance of polluted continental air-mass (NE winds) and low during May-Aug (SW winds). Error bar represents the standard deviation.

Seasonal and inter-annual variation of NH_4^+ and NO_3^- in aerosols and rain

The abundances of NH_4^+ and NO_3^- measured in aerosols and precipitation samples exhibit large variations (Fig. 2). The aerosol abundance of NH_4^+ ranges from < 0.001 to 1.3 $\mu g/m^3$ and that of NO_3^- as 0.09 to 4.4 $\mu g/m^3$, with relatively high values during Sept-Oct (NH_4^+: Avg. 0.38 ± sd. 0.27; NO_3^-: 2.3 ± 0.74) and Nov-Feb (NH_4^+: 0.60 ± 0.37; NO_3^-: 2.2 ± 0.72) (Fig. 3). The concentrations of both species were relatively low during Mar-Apr (NH_4^+: 0.27 ± 0.13; NO_3^-: 1.5 ± 0.57) and May-Aug (NH_4^+: 0.05 ± 0.06; NO_3^-: 0.94 ± 0.76), with NH_4^+ concentration often below detection limit (0.001 $\mu g/m^3$) (Fig. 3). During May-Aug, relatively low abundances of nitrogen species are attributed to the predominance of SW winds (marine air mass), as well as rapid removal of aerosols (via wet deposition). The volatilization loss of NH_4^+ (if any) present as NH_4NO_3 or NH_4Cl aerosols could also be significant during Apr-Jun before the onset of SW monsoon when ambient temperature over the study site reaches maximum [a mechanism similar to that suggested by Matsumoto and Okita (1998)]. Relatively high concentrations of both species are observed during Nov-Feb (Fig. 3), under the influence of NE winds that dominate the transport of continental pollutants (Rastogi, 2005).

The volume-weighted-mean (VWM) concentrations of NH_4^+ and NO_3^- in precipitation events collected during SW monsoon, for three years, are shown in Fig. 4. The abundance of NH_4^+ ranges from < 1.0 to 220 $\mu eq/L$ (VWM: 30 $\mu eq/L$) and accounts for nearly 20% to the cation balance (Rastogi and Sarin, 2005). The relatively low NH_4^+ concentration (close to detection limit) in aerosols during May-Aug (Fig. 3) and its significant abundance in precipitation events

during SW monsoon period ascertains its dominant occurrence in gaseous phase over the study site. The abundance of NO_3^- ranges from 1.3 to 115 (VWM: 13 μeq/L) and on average contributes ~10% to the anion balance (Rastogi and Sarin, 2005). The large variability observable for NH_4^+ and NO_3^- in rain events is attributed to the variations in the source strength, antecedent dry period between the successive rain events and precipitation amount. A successive increase in VWM of NO_3^- over three-years (Fig. 4) can be viewed as systematic increase in NO_x emission over the Asian region (Van Aardenne *et al.*, 1999; Streets *et al.*, 2003). The VWM of NH_4^+ does not show a significant and systematic inter-annual variability.
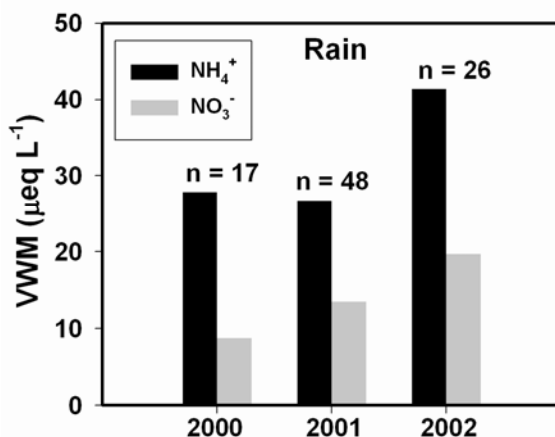

Fig. 4. Volume-weighted-mean (VWM) concentration of NH_4^+ and NO_3^- in rain events during SW monsoon shows linear increase in NO_3^- abundance, attesting to predicted trends over Asia.

Dry and wet deposition fluxes of NH_4^+ and NO_3^-

Based on the average concentration of NH_4^+ and NO_3^- in aerosols and volume-weighted-mean (VWM) concentration in rainwater during the study period, their dry and wet deposition fluxes $(mg/m^2/y)$ have been computed as follows:

$$\text{Dry deposition flux} = [X] \times V_d \qquad (1)$$

$$\text{Wet deposition flux} = C \times P \qquad (2)$$

Where, [X]: Average concentration of a species X (mg/m^3); V_d: deposition velocity (m/y) for that species; C: VWM concentration of species (mg/m^3); P: Precipitation (m/y). V_d is considered as 0.1 cm/s for NH_4^+ and 0.3 cm/s for NO_3^- (Duce *et al.*, 1991).

Dry deposition fluxes of NH_4^+ and NO_3^- during 2000, 2001 and 2002 are 16, 11 and 16 mg/m^2/y and 201, 134 and 137 mg/m^2/y, respectively, while wet deposition fluxes are 364, 327 and 297 mg/m^2/y, and 392, 569 and 487 mg/m^2/y, respectively (Fig. 5). These results indicate that almost 90% of NH_4^+ removal from the atmosphere occurs via wet deposition; whereas 20-30% of NO_3^- is removed by dry, and 70-80% via wet deposition (Fig. 5). The inter-annual variability in dry and wet deposition fluxes of NH_4^+ and NO_3^- is not significantly pronounced during the three-year study period (Fig. 5). The total deposition fluxes of inorganic nitrogen species (NH_4^+ + NO_3^-) during the year 2000, 2001 and 2002 are 973, 1041 and 937 mg/m^2/y, respectively; similar to the modeled values of ~1000 mg/m^2/y over this region (Galloway *et al.*, 2004). Of the total inorganic-N-deposition, NO_3^- accounts for as much as ~ 65% and that of NH_4^+ is ~35%.

A comparison of NH_4^+ and NO_3^- abundances over different sites in India, as well as from a few global regions, are presented in Table 1, thus representing their dry deposition fluxes (assuming uniform deposition velocities for NH_4^+ and NO_3^- over all the sites). Their abundances over Ahmedabad are similar to those reported over Christchurch (New Zealand) and Dundee (United Kingdom) while significantly lower than those reported over other sites in India and the world (Table 1). The observed differences are attributable to their different source inputs, as well as source strength and meteorological conditions. The wet deposition fluxes of NH_4^+ and NO_3^- over Ahmedabad are comparable to those reported over Pune (Pillai *et al.*, 2001), Delhi (Jain *et al.*, 2000), Hyderabad (Kulshreshtha *et al.*, 2003), Amersfoort and Louis Trichardt (Mphepya *et al.*, 2004), Banizoumbou (Galy-Lacaux and Modi, 1998) and Ankara (Kaya and Tuncel, 1997); but significantly lower than those at Reston (Conko *et al.*, 2004), Sau Paulo (Rocha *et al.*, 2003) (Fig. 6).
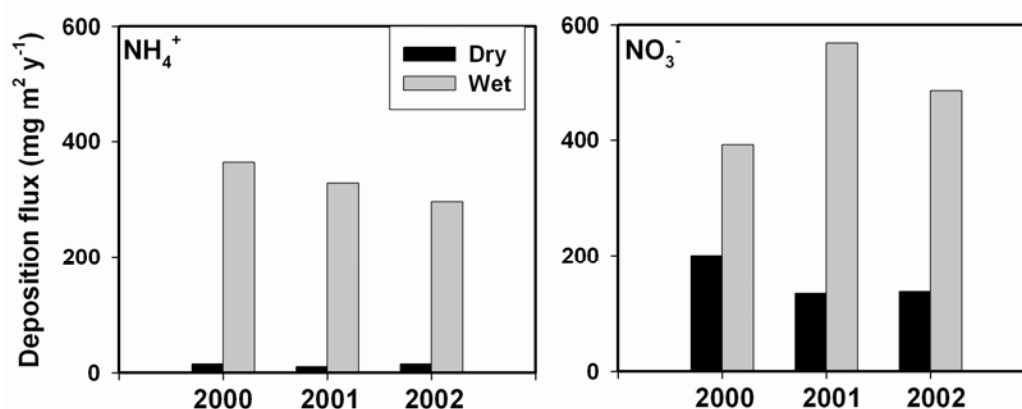

Fig. 5. Dry and wet deposition fluxes of NH_4^+ and NO_3^- over an urban area (Ahmedabad). Wet deposition seems to be the major removal pathway for both the species and inter-annual variability in their deposition fluxes is insignificant.
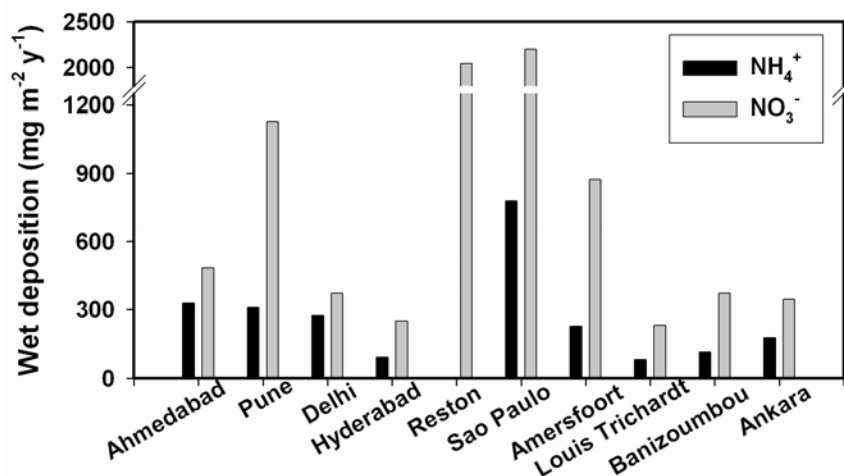
Fig. 6. Wet deposition fluxes of NH_4^+ and NO_3^- over Ahmedabad (3-year average) and comparison with a few other study sites. By and large, wet fluxes are comparable to those reported over other sites.

Table 1. Abundances ($\mu g/m^3$) of NH_4^+ and NO_3^- in aerosols over Ahmedabad and a few sites in India and the world.

City	Country	Year	NH_4^+	NO_3^-
Ahmedabad	India	2000	0.50	2.12
Ahmedabad	India	2001	0.33	1.42
Ahmedabad	India	2002	0.50	1.45
Mumbai[a]	India	1999	-	4.70
Pune[b]	India	1998	2.14	2.91
Delhi[c]	India	1998	-	3.38
Agra[d]	India	1993	6.52	8.37
Dhaka[e]	Bangladesh	2001	1.86	3.63
Erdemli[f]	Turkey	1996-99	2.22	3.34
Shanghai[g]	China	2000	6.60	6.80
Christchurch[h]	New Zealand	2000	0.22	1.03
San Joaquin[i]	USA	1993	5.52	13.5
Dundee[j]	UK	2002	0.67	1.11

[a]Venkatraman et al., 2002; [b]Momin et al., 1999; [c]Parashar et al., 2001
[d]Kulshrestha et al., 1998; [e]Salam et al., 2003; [f]Kocak et al., 2004;
[g]Yao et al., 2002; [h]Wang and Shooter, 2001; [i]Chow et al., 1993;
[j]Qin and Oduyemi, 2003.

CONCLUSIONS

The abundances of inorganic nitrogen species (NH_4^+ and NO_3^-) measured in rain and aerosols over an urban area located in a semi-arid region of western India has brought out the following important conclusions:

1. Their abundances in aerosols exhibit large temporal variation with high concentrations occurring in Nov-Feb (predominance of NE winds during winter time); and low during the wet season (Jun-Aug) when dominant atmospheric removal occurs via wet deposition from gaseous phase. The abundances of NH_4^+ and NO_3^- also exhibit large variability in the individual precipitation events. The inter-annual variability in the VWM of NH_4^+ is not significantly pronounced, but that of NO_3^- is marked by systematic increasing trend.

2. The inter-relationship with major inorganic species points to biomass burning and anthropogenic emissions as the dominant sources of NO_3^- in rain and aerosols. The contribution of NO_3^- from dust sources is not predominant. The NH_4^+ is derived from anthropogenic sources and largely present in gaseous phase.

3. Wet deposition is the dominant mechanism for the atmospheric removal of NH_4^+ and NO_3^- and that contribution of NO_3^- to the total deposition inorganic-N (NH_4^+ and NO_3^-) is dominant (~65%).

Such regional scale studies from semi-arid regions are important where current knowledge must be expanded if the impacts of environmental change are to be properly assessed. Thus, these data on N-deposition make a significant contribution to global programs, such as the International Nitrogen Initiative (INI) and Deposition of Biogeochemically Important Trace Species (DEBITS).

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