



Atmospheric Wet and Dry Depositions of Ions over an Urban Location in South-West India

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ABSTRACT

Wet deposition (WD) and Dry deposition (DD) samples were collected during a period of 4 year (2006 to 2009), at four different sites representing different surroundings around Pune city in southwest India. The samples were collected on a daily basis for WD and weekly basis for DD. These samples were analyzed for major ionic components e.g., Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , NH_4^+ , Ca^{2+} and Mg^{2+} . Both the WD and DD were alkaline ($\text{pH} > 5.6$) at all the four sites. The WD fluxes of all the ionic components were higher than the DD fluxes, except at the traffic junction Swargate, where majority of the species appeared with much higher DD fluxes than WD fluxes (68% for NO_3^- , 63% for Ca^{2+} , 60% for Mg^{2+} , 57% for K^+). WD flux of NH_4^+ is higher (64–80%) than the DD flux at three locations and slightly lower (48%) at a high altitude location. In case of sea salt (Na and Cl), WD fluxes were higher (63–90%) than the DD fluxes at all the four locations. The dominant ion in DD was NO_3^- at Pashan (semi-urban) and Sinhagad (high altitude), Ca^{2+} at Swargate (traffic junction) and SO_4^{2-} at Bhosari (industrial). The difference in deposition fluxes between the four sites was attributed to the effect of the local sources. Deposition velocities of SO_4^{2-} and NH_4^+ were < 1 cm/s while Ca^{2+} , Mg^{2+} , Na^+ , K^+ , NO_3^- , and Cl^- exhibited deposition velocities ≥ 1 cm/s. At one of the sites Pashan, where the earlier data is available; DD rates showed increase in all the chemical components, except for NH_4^+ after a period of about 2 decades.

Keywords: Soil dust; Dry deposition; Long-range transport; Anthropogenic sources; Deposition velocity.

INTRODUCTION

Atmospheric deposition is the process by which airborne particles and gases are deposited to soils, vegetation, waters, and other surfaces either through precipitation (rain, snow, clouds, and fog), known as wet deposition (WD) or as a result of complex atmospheric processes such as settling, impaction, and adsorption, known as dry deposition (DD). Wet or dry deposition of nitrogen and sulfur compounds can result in acidification of freshwaters, loss of aquatic species, eutrophication of estuarine and near-coastal waterways, soil nutrient and base cation leaching, and vegetation changes.

The deposition of dust and other water-soluble chemical species is directly connected to the changes in the various biogeochemical cycles in the atmosphere, and are also related to the Earth's radiation budget (Tegen *et al.*, 1996). The main processes removing the chemical species from the atmosphere are wet and dry depositions (Sehmel, 1980).

WD is the removal of substances from the free troposphere and the boundary layer by precipitation. DD is the direct transport of gases and particulates on to land and water surfaces. DD occurs by several processes, including Brownian motion of particles, settling by gravity, and wind impaction. Both DD and WD depend strongly on meteorological conditions and local emissions (Inomata *et al.*, 2009; Cheng *et al.*, 2010).

DD appears to be an important process for Indian climatic conditions as dry conditions prevail for most part of the year, while rains are confined to a short monsoon period for four months from June to September. Yet, literature on DD is scarce and only a few studies are available (Tripathi *et al.*, 1991; Rao *et al.*, 1992; Saxena *et al.*, 1992, 1997; Singh *et al.*, 1999; Satsangi *et al.*, 1999, 2002; Kumar *et al.*, 2003; Das *et al.*, 2005; Kumar *et al.*, 2006). One of the major aspects for deposition studies is the acidity of depositing components that has a critical impact on the receiving surfaces. The degree of the acidity of deposition depends on the neutralization by certain alkaline components, such as NH_4^+ , Ca^{2+} (in the form of CaCO_3 or Ca(OH)_2), Na^+ and Mg^{2+} . Wet and dry depositions are essential for understanding the regional variations in local influences by anthropogenic

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or natural sources and long-range transport phenomena of air pollutants. The different climatic conditions, the topography of the area and the elevation above sea level also influence these mechanisms. The DD fluxes are marked by seasonal variation, being generally high in summer, and low in monsoon (Kumar *et al.*, 2003). The present work focuses on annual and seasonal trends of wet and DD fluxes over Pune region.

DESCRIPTION OF SAMPLING SITES

Pune is one of the fastest developing urban agglomerations in Asia. It is the eighth largest metropolis in India, and the largest city in the Western Ghats, about 100 km off the West coast. Presently the population of the city is around 3.1 million. Pune has a tropical wet and dry climate with average temperatures ranging between 20 to 28°C. There are three distinct seasons namely summer, monsoon and winter. Typical summer months are February to May, with maximum temperatures ranging from 30 to 39°C. The warmest month is April. The city often receives heavy pre-monsoon thundershowers in May. Even during the hottest months, the nights are usually cool at Pune due to its altitude (559 m amsl). In monsoon, it receives almost 80% of the annual rainfall and July is the wettest month of the year. Average annual rainfall is about 722 mm. Winter begins in November and lasts up to January end. The average temperature in winter is around 29°C and the minimum values range between 10°C and 15°C.

WD and DD samples were collected at four locations, representing distinct surroundings in Pune. All these four locations are shown in the Fig. 1.

1) Sampling site at Swargate, a traffic junction, is about 15 m above the ground, approximately 200 m away from the main traffic center. Swargate is one of the major and most

busy bus depots in the city. It is very crowded and smoke belching place. 2) Bhosari is located in an industrial area, about 25 km to the north-northwest of the main Pune city. There are establishments of various small and large scale industries like steel, rubber, electronic items, agricultural products, glass works and many others. The sampling site is the terrace of a building at about 20 m above the ground level. 3) Another sampling site in Pashan, a semi urban location is at about 12 m above the ground level. It is about 8 km to the west- northwest of the main city of Pune. The site is surrounded on three sides by hillocks of variable heights (up to 200 m), forming a valley like appearance. 4) The sampling site at Sinhadgad, a rural high altitude (1450 amsl) location, is about 2 m above the surface. Sinhadgad is a hill station on a mountaintop in the Western Ghat mountain ranges. It is located at about 18 km southwest of Pune. Its top is flat with an area of about 0.5 km² (Budhavant, 2009). This location represents a rural background site.

METHODOLOGY

There are different sampling techniques for the collection of DD samples (Saxena *et al.*, 1997; Morales *et al.*, 1998; Tanner *et al.*, 2001; Odabasi and Bagiroz, 2002; Lestari *et al.*, 2003; Tasdemir and Gunez, 2006). WD and DD samples were collected using bulk collector at all the four locations, using standard rain collection gadget (Polyethylene funnel of diameter 18 cm and 2 lit collection bottle) at four locations during 2006–2009. The samples were collected on a daily basis for WD and weekly basis for DD in dry periods by washing the funnel with 80 mL deionised water. The samples were filtered through Whatman-41 filter paper. Also Total Suspended Particulates (TSP) were collected during March 2007 to February 2008 using High Volume Air Sampler (Envirotech, India Model 410) at an average flow rate of

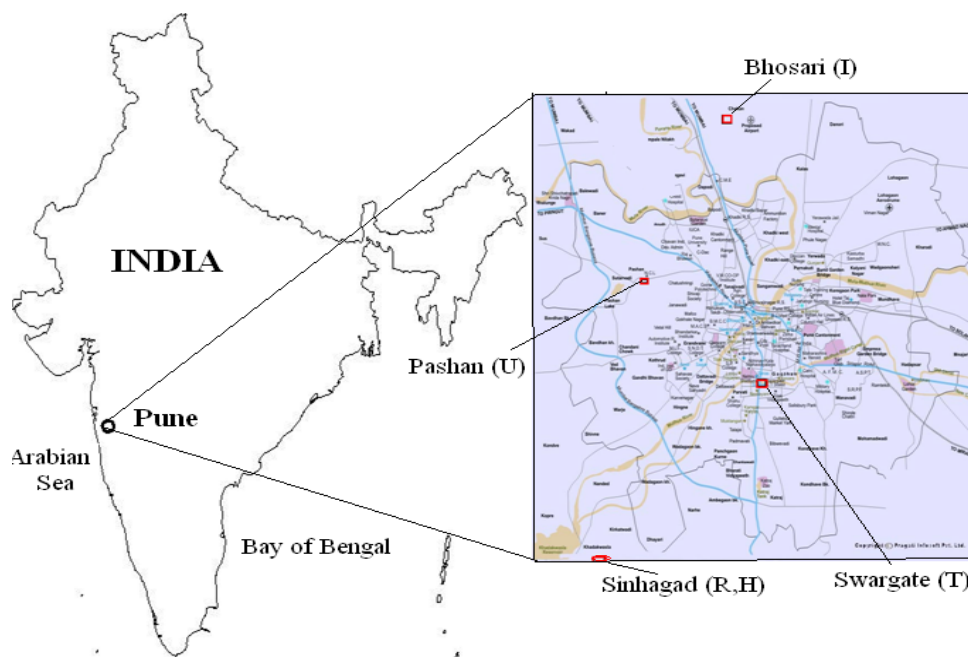


Fig. 1. Location of Pune in India and Sampling locations in Pune city.

1.1 m³/min. Whatman-41 filter papers of 8" × 10" size were used for collection of TSP. Sampling duration for each TSP sample was about 8 h. The net aerosol load on the filter papers was determined by weighing the filter papers before and after sampling with a microbalance in humidity-equilibrated environment. The pH was measured with a digital pH meter, standardized at pH 4.0 and 9.2. Conductivity was measured with a digital conductivity meter calibrated against a reference KCl solution. Chemical species were analyzed with an Ion Chromatograph (IC100). An IonPacAS4A-4 × 125 mm Analytical, column (Dionex Co. Ltd.) was used for anion measurements; sodium bicarbonate (NaHCO₃, 1.7 mM)/sodium carbonate (Na₂CO₃, 1.8 mM) buffer was used as the eluent. The major cations (Na⁺, K⁺, Ca²⁺ and Mg²⁺) were analyzed by Atomic Absorption Spectrophotometer (Perkin Elmer-AAAnalyst 400) with an air acetylene flame. The NH₄⁺ ion was measured by Indo-phenol Blue method (Weatherburn, 1967). The detection limit for ion chromatographic analysis was about 0.01 ppm and that for the atomic absorption spectrophotometric analysis varied from 0.002 to 0.02 ppm.

The Ion balance technique is widely used to check the quality of the data. Under this check, ratio between the sum of cations to the sum of anions both in µeq/L was estimated. The samples which fail this quality check are removed from the data sets. The average ratio for all the locations is 1.02 ± 0.13 for WD and DD samples it is 1.13 ± 0.28. A significant correlation was observed between the sum of anions and sum of cations as shown in Fig. 2. This indicates the completeness of the analyses. Another technique used for the data quality check was the comparison between measured and theoretically computed conductivities (EMEP/CCC-Report 1/95, 1996). Also, this Institute has been participating in the international inter-comparison studies i.e., EANET's Inter-laboratory comparison projects and WMO's Laboratory

Inter-comparison studies (LIS). The results from these comparison studies showed good agreement between the standards and our chemical analyses.

RESULTS AND DISCUSSION

pH and Ionic Composition of Wet and Dry Deposition

The study based on chemical analyses of WD and DD samples, collected at four locations in Pune city reveals that, on an average, WD and DD was alkaline at all the four locations with pH values for WD were 6.70, 6.16, 5.94, 5.92 for Swargate, Bhosari, Pashan and Sinhagad, respectively, and 7.39, 7.04, 5.97 and 5.94 for DD at these locations respectively. Higher pH value of WD at the traffic location than that at the other locations was mainly due to the abundance of Ca²⁺, a major neutralising cation that is originated by vehicle-driven road-side dust.

The average annual WD and DD fluxes (g/m²/y), of major ionic components calculated from 4-year data are shown in Fig. 3. Except NO₃⁻ and K⁺, WD fluxes of the all the ionic components were higher than the DD fluxes at Pashan, Bhosari and Sinhagad. It is reported that the WD fluxes are proportional to the precipitation amounts and hence the WD will be higher than DD (Galloway, 1985; Chan *et al.*, 1986; Mosello *et al.*, 1988). The dominant ion in the DD was NO₃⁻ at Pashan (0.99 g/m²/y) and Sinhagad (1.97 g/m²/y), Ca²⁺ (2.75 g/m²/y) at Swargate and SO₄²⁻ (1.69 g/m²/y) at Bhosari. The high concentration of SO₄²⁻ at Bhosari may be due to the industrial emissions. WD fluxes of Cl⁻ and SO₄²⁻ showed maximum concentration at Sinhagad (3.54 and 2.62 g/m²/y), Pashan (0.96 and 0.95 g/m²/y), and Bhosari (1.12 and 1.91 g/m²/y) respectively. Where as at Swargate Ca²⁺ (2.75 g/m²/y) showed maximum flux.

Generally DD includes deposition to all different

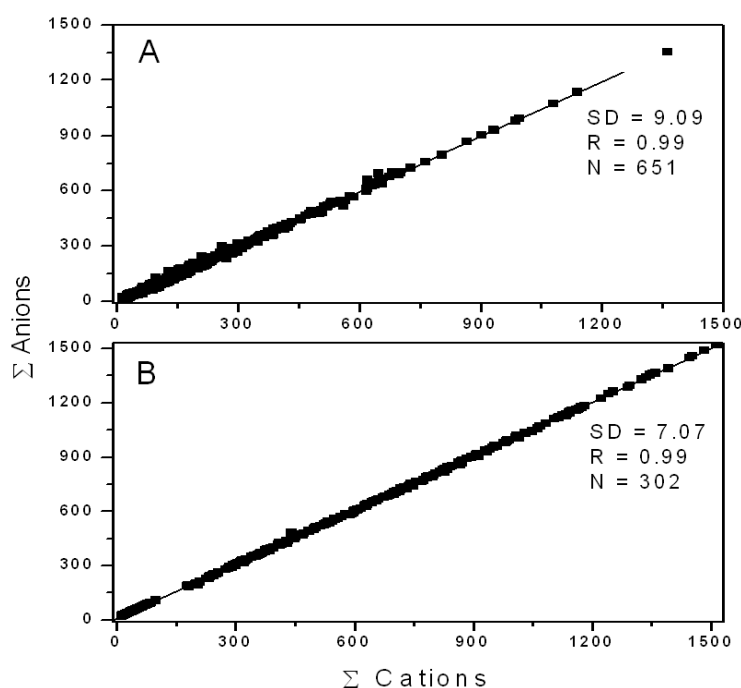


Fig. 2. Relationship between the Cations and Anions in A) Wet, and B) Dry deposition samples, 2006–09.

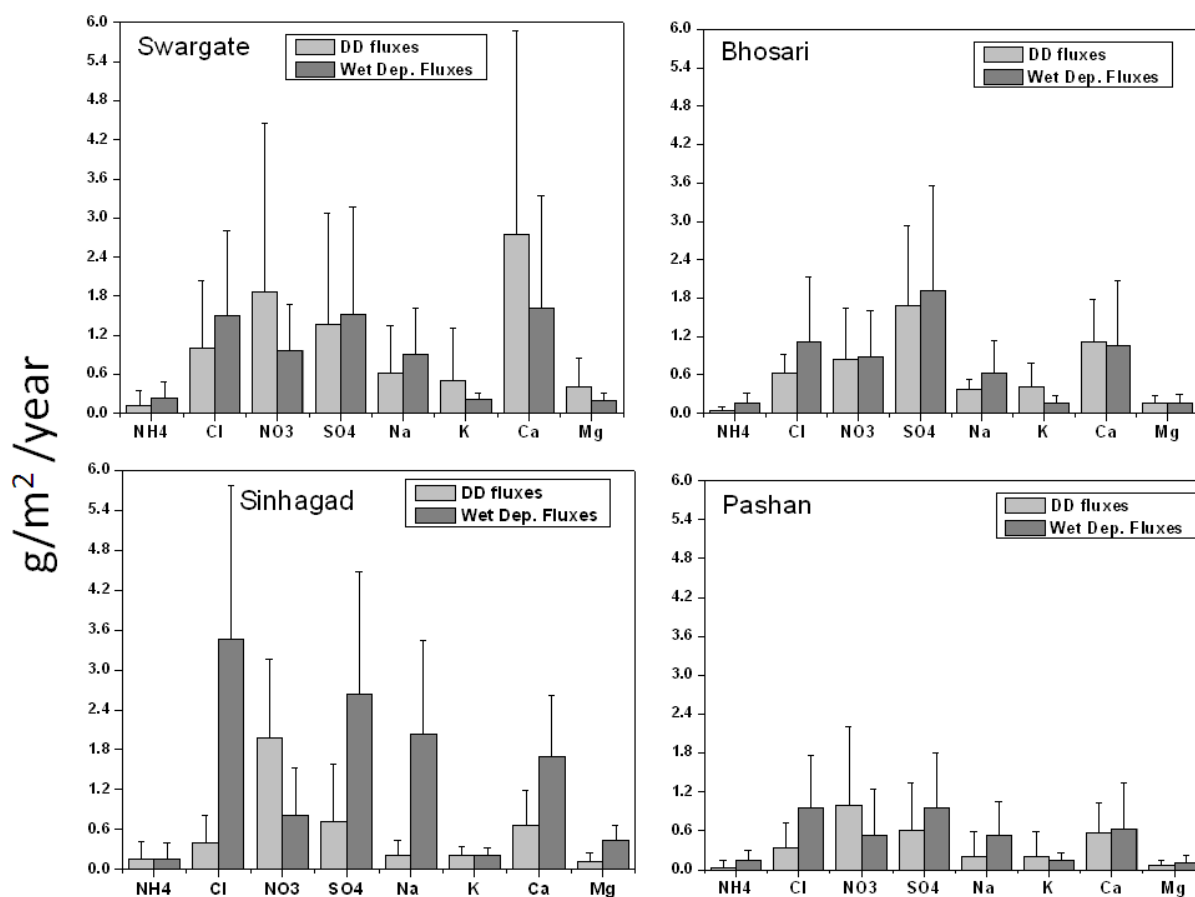


Fig. 3. The average annual wet and dry deposition fluxes of various chemical species at four different locations (2006–09).

underlying surfaces (canopies, soils, water, etc). However, there are many uncertainties in the methods of quantifying DD (Yi *et al.*, 1997; Shahin *et al.*, 2000). To date, there is as yet no accepted technique for evaluating the accuracy of these methods. DD has been primarily based on modeled estimates. A routine direct measurement technique is required for validating these estimates. DD collected in this study is similar to the method using surrogate surfaces. This method might provide reasonable estimation of DD for very large particles considering their DD is dominated by gravitational settling. However, for species mainly in fine particles, DD depends strongly on the characteristics of underlying surfaces. The method used here could provide a low-end estimation of DD for the species having significant fraction of fine particles.

The relative contribution of WD and DD fluxes to total deposition for the four locations are shown in Fig. 4. It can be seen that the WD fluxes were higher for Na^+ , Cl^- , SO_4^{2-} , Mg^{2+} , Ca^{2+} and NH_4^+ than the DD fluxes at Pashan, Bhosari and Sinhagad locations, however at Bhosari and Sinhagad, DD fluxes were significant for Ca^{2+} and NH_4^+ . This result leads to the conclusion that WD is the dominant mechanism for the majority of the ionic species over Pune region. Similar results have been obtained in other investigations where WD of ionic species contributed higher to the total deposition (Mosello *et al.*, 1988; Rao *et al.*, 1992). On the other hand, at traffic junction location Swargate, majority of

the species appeared with much higher DD fluxes than WD fluxes (68% for NO_3^- , 63% for Ca^{2+} , 60% for Mg^{2+} , 57% for K^+) as shown in Fig. 4. This different deposition pattern between the sites in the same region can be attributed to effects of different sources, at these sites. Site of Swargate is mainly affected by the local emissions i.e., particles from wearing of vehicular brake and tire, vehicular exhaust, and track-out soil from unpaved roads which are reported to be the major sources near a traffic junction (Kuhns *et al.*, 2004; Bhaskar *et al.*, 2010; Chakraborty and Gupta, 2010) and is obviously more contributing fluxes to the DD. Chemical analyses of road surface material indicate that most of the particulate matter in it is composed of crustal species (Chow *et al.*, 2004; Balakrishna and Pervez, 2009, Kothai *et al.*, 2011).

Seasonal Variation in Dry Deposition Flux

Seasonal estimates of atmospheric DD fluxes have been prepared to understand the various emission activities and atmospheric processes. However, due to the consistent and frequent precipitation throughout the monsoon season, it was not possible to collect the DD samples in this season. The average fluxes of DD per day of major ionic components during summer and winter seasons are shown in Fig. 5. The DD fluxes varied significantly during these seasons. During the summer season the atmosphere at Pune generally remains unstable due to high convection. Aerosols of local

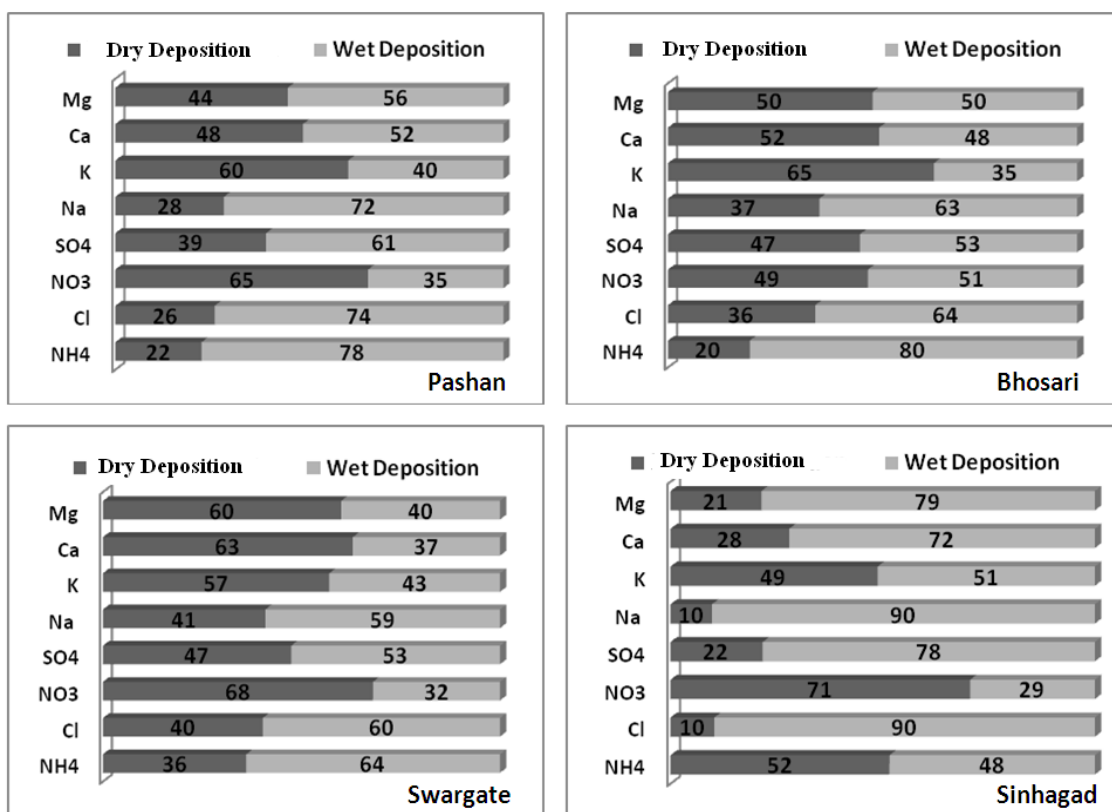


Fig. 4. Percentage contribution of wet and dry deposition to the total deposition over Pune.

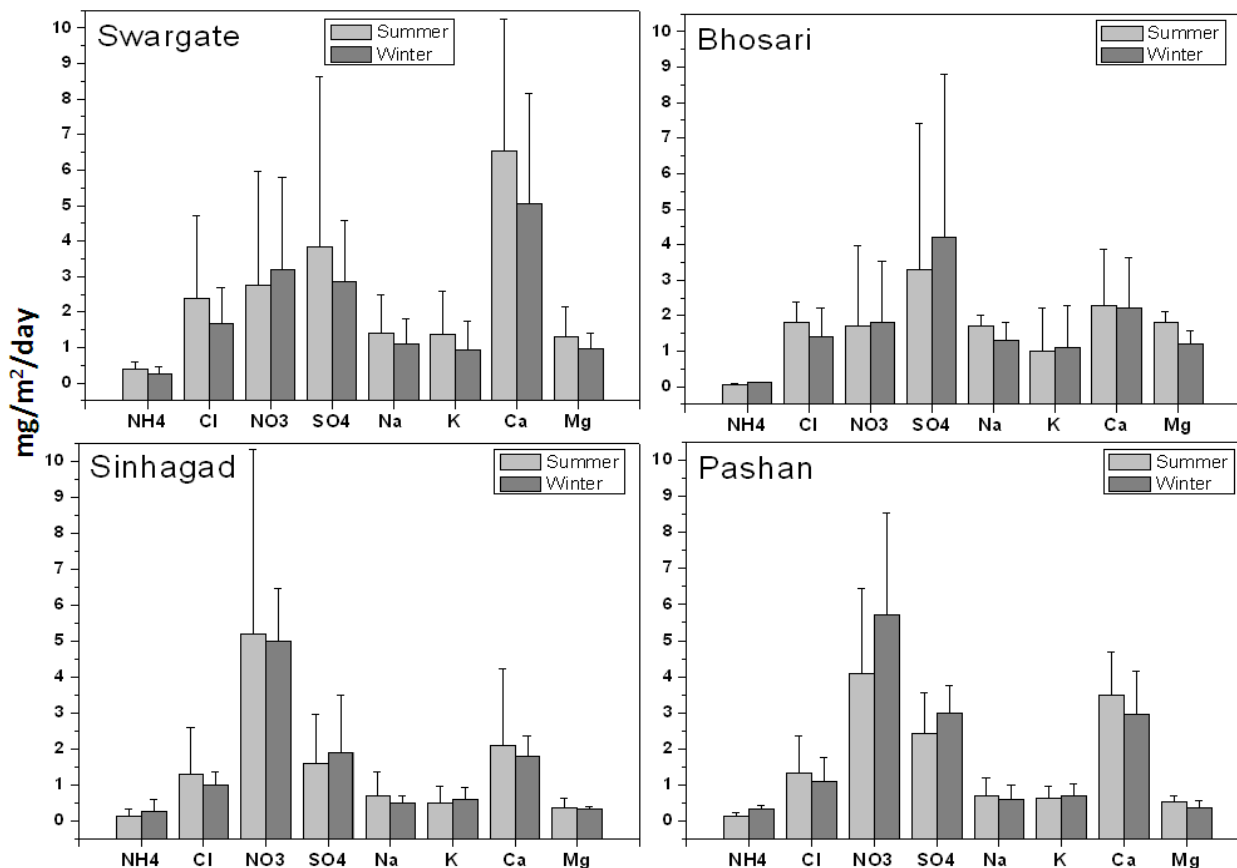


Fig. 5. Seasonal variation in dry deposition fluxes at four different locations over Pune region, 2006–09.

and land origin are continuously lifted up and therefore, higher deposition fluxes of soil-derived elements are observed. However, sometimes DD fluxes get reduced in this season due to the occurrence of occasional rain showers. The air flow during the summer period is predominantly westerly and large amount of moisture is brought inland from the Arabian Sea. This is responsible for the observed high DD fluxes of sea salt ions Cl^- , Na^+ and Mg^{2+} . The DD flux of NH_4^+ was found to be low among all the ions in both the seasons. Also, the comparatively high DD flux of NH_4^+ in winter may be due to the formation of stable NH_4NO_3 aerosol because of low winter temperatures (Safai *et al.*, 2010). Both SO_4^{2-} and NH_4^+ showed higher DD fluxes during winter than summer, except at Sinhgad.

In the winter period, the wind in the lower atmosphere reverses with the withdrawal of the monsoon and easterly flow sets in from October. As a result, the continental, air mass, rich in aerosols of continental origin, passes over the region (Fig. 6(A) and (B)). Also, occurrence of rain is less than that in the summer season. These features account for the high DD fluxes of the major ionic components, except sea salt and Ca^{2+} , during the winter. However, in case of Ca^{2+} , the major source lies in the W/NW of sampling locations (arid zones in NW India) and therefore due to westerly winds (Fig. 6), DD fluxes for Ca^{2+} are more during summer at all the sites.

Dry Deposition Velocities

The DD rate of pollutants at the surface is usually expressed in terms of the deposition velocity. The deposition velocity (V_d) of the measured ions can be used as a measure of mass transfer onto the receiving surface (Hosker and Lindberg, 1982) and is calculated according to the formula $V_d = F/C$. In the present study DD velocities of major ions are determined by taking ratio of the fluxes of major ions (F) to the simultaneously measured air concentrations (C) of the respective ions. DD fluxes were obtained using the bulk collector (polypropylene surface) while air concentrations were obtained from TSP samples collected by a high-volume air sampler. The DD velocities of the major water-soluble components at Pashan for summer and winter seasons (2007–08) are shown in Fig. 7. The variation in the DD velocities may be a function of particle size distribution and the meteorological conditions (i.e., wind direction, wind speed, temperature, atmospheric stability) (Finlayson-Pitts and Pitts, 1986; Saxena *et al.*, 1997; Zeller *et al.*, 1997; Caffrey *et al.*, 1998; Tanner *et al.*, 2001; Zhang *et al.*, 2001; Odabasi and Bagiroz, 2002). The average annual DD velocities are given in Table 1. DD velocities were found to be > 1 cm/s for NO_3^- , Ca^{2+} , K^+ , Cl^- and Mg^{2+} , whereas they were < 1 cm/s for NH_4^+ and SO_4^{2-} . In case of Na^+ , DD velocity was nearly 1 cm/s. The DD velocity was highest for NO_3^- and lowest for NH_4^+ . This suggests that velocities are directly proportional to the size of the aerosols, i.e. coarse size aerosols show high DD velocity. It is reported (Momin *et al.*, 1999) that NO_3^- over Pune exists in coarse size due to formation of $\text{Ca}(\text{NO}_3)_2$ originated from reaction of HNO_3 vapours with soil derived Ca^{2+} or marine Na^+ (Wolff, 1984).

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model developed by the National Oceanic and Atmospheric Administration Air Resources Laboratory, USA, was used to calculate the backward trajectories of air masses reaching the Pune region. The 7 day backward trajectories were calculated for DD at 600 m beginning at 00 UTC. The 7 days backward trajectories for the WD were calculated at 1500 m, beginning at the precipitation start time. The air masses that brought dust to Pune region were transported mainly via two paths. One path leads directly from the adjoining Arabian Sea during summer, through southwesterly winds (Fig. 6). The other path shows air masses associated with heavy dust deposition were transported from North West part of India by northwesterly winds. Most of the air masses following the other path are transported from the African continent and over the Arabian Sea before arriving over Pune indicating mixed continental-maritime air mass (Fig. 6).

Comparison with Reported Studies

The present results on atmospheric concentration, DD flux and DD velocity at one of the location, Pashan have been compared with those earlier reported (Rao *et al.*, 1992) for the same site (Table 1). After a period of 20 years, the concentrations and DD flux of major acidifying components, i.e. SO_4^{2-} (77%) and NO_3^- (46%) showed substantial increase. This is attributed to the rapid increase in urbanization and industrialization during the two decade period over this site. Anthropogenic air pollution caused by residential heating, industrial activities and transportation is one of the main reasons for high sulfate concentrations in the city atmosphere because the burning of sulfur-containing fuels emits sulfur oxides, which are converted to sulfates (Seinfeld and Pandis, 1998). Concentration of other alkaline component such as Ca^{2+} and NH_4^+ also showed 3.1 times and 3.2 times increase respectively. The Sea salt components i.e. Na^+ , Cl^- and other components like K^+ and Mg^{2+} have not shown significant increase. The deposition velocities of all the ionic components, except for NH_4^+ , have not changed much.

There are different sampling techniques for the collection of samples of DD for particulate sulfate (Saxena *et al.*, 1997; Morales *et al.*, 1998; Tanner *et al.*, 2001; Odabasi and Bagiroz, 2002; Lestari *et al.*, 2003; Tasdemir and Gunez, 2006). However, there is no generally accepted method for collection and analysis of DD (Odabasi and Bagiroz, 2002). Therefore, there is a wide range of reported deposition fluxes available in the literature (Table 2). The sulfate fluxes measured at Bhosari (3.72 mg/m²/d) were higher than those measured in rural areas (Table 2). However, Tasdemir and Gunez (2006) reported a very high average particulate sulfate flux value (30.9 and 154.9 mg/m²/day) at Bursa, Turkey. The average flux reported from Sinhgad was smaller than the average flux measured in the city area of Pune. This is due to the high level of sulfate in the city due to presence of more anthropogenic sources. Similarly, two sites of rural and sub-urban environment in Venezuela and China showed similar fluxes as reported at Sinhgad whereas a slightly higher sulfate DD flux has been reported from a rural site in Agra mg²/day by Saxena *et al.* (1997).

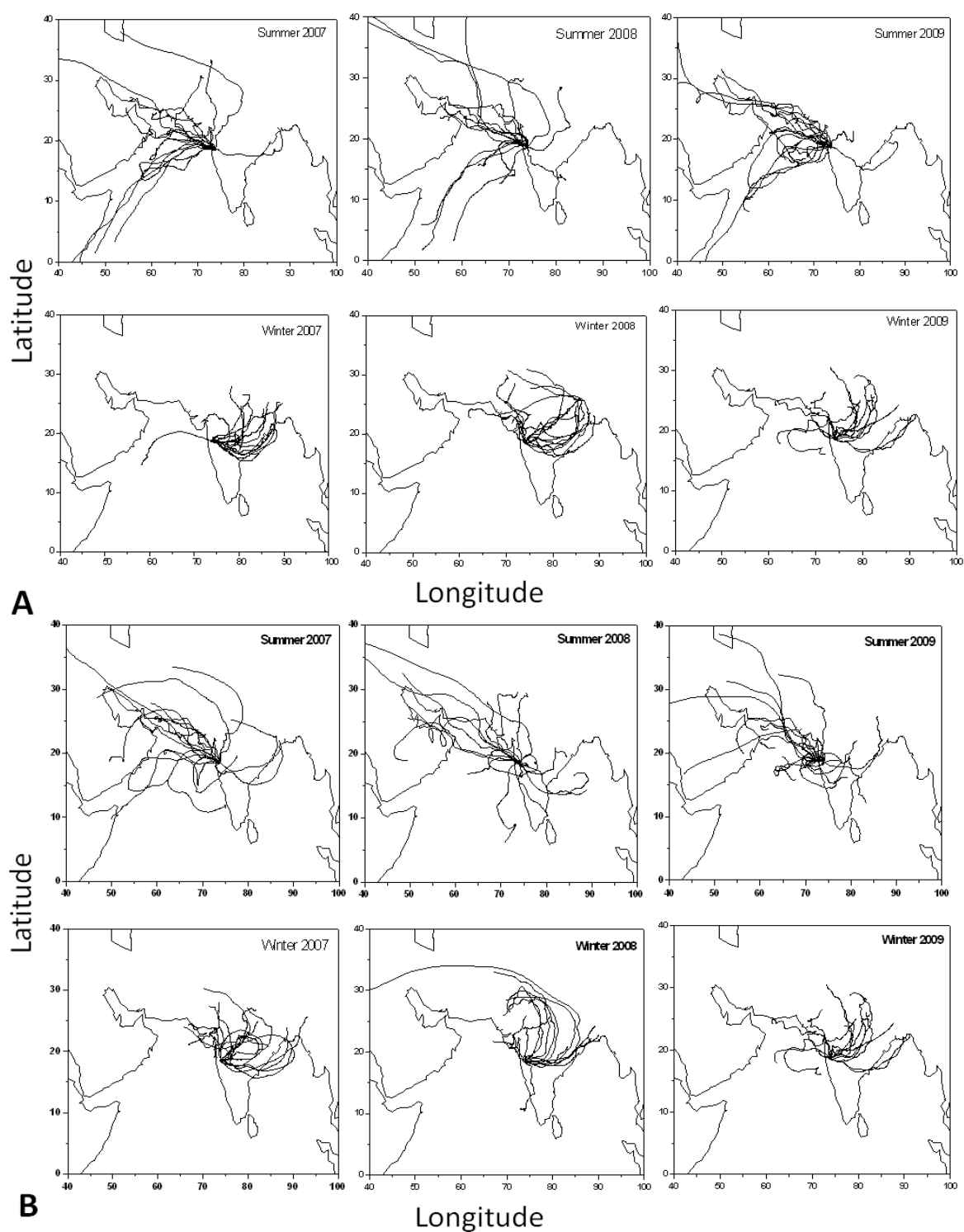


Fig. 6. Air mass backward trajectory for 7 days ending at Pune during summer and winter seasons at 600 m and 1500 m amsl heights.

CONCLUSIONS

In the present study, we measured wet deposition and dry deposition fluxes of major water-soluble chemical species at different locations in Pune, India, during 2006–2009. The major conclusions obtained are as following:

The present study confirms the consideration that DD is

generally more connected with local emissions contrary to the WD which better reflects transport phenomena and other gas-particle reaction formations. Although it was expected that atmosphere of the Pune city would be more influenced from emissions from the traffic and industries. However, due to the significant contribution of alkaline components originating from crustal sources the WD and DD were not

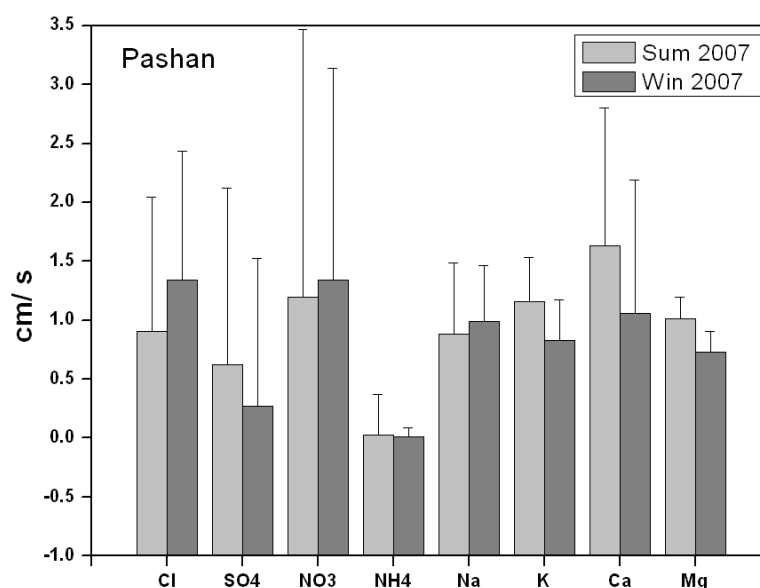


Fig. 7. Dry deposition velocities of different chemical species at Pashan during summer and monsoon seasons (2007–08).

Table 1. Comparison of DD velocities of the major water-soluble components of the DD at Pashan site (cm/s).

Element	Rao <i>et al.</i> , 1992			Present Study		
	Atmos. Conc. ($\mu\text{g}/\text{m}^3$)	DD flux ($\text{mg}/\text{m}^2/\text{d}$)	DD Velocity (cm/s)	Atmos. Conc. ($\mu\text{g}/\text{m}^3$)	DD flux ($\text{mg}/\text{m}^2/\text{d}$)	DD Velocity (cm/s)
Cl ⁻	1.35	1.24	1.10	1.81	1.79	1.15
SO ₄ ²⁻	1.27	0.58	0.53	7.51	2.89	0.45
NO ₃ ⁻	1.21	1.54	1.50	3.23	4.73	1.69
NH ₄ ⁺	0.45	0.07	0.18	1.45	0.03	0.02
Na ⁺	0.78	0.61	0.91	0.95	0.78	0.95
K ⁺	0.35	0.27	0.89	0.46	0.52	1.29
Ca ²⁺	1.01	2.59	2.97	3.18	4.33	1.58
Mg ²⁺	0.37	0.32	1.00	0.49	0.46	1.09

DD- Dust-fall deposition; Atmos. Conc.-Atmospheric Concentration.

Table 2. Particulate sulfate dry deposition fluxes ($\text{mg}/\text{m}^2/\text{day}$) measured at various locations.

Location	Sulfate flux	Deposition Velocity	Method	Period	Reference
Rural, La Esperanza, Venezuela	1.4	0.10	Plexiglass funnel	Annual, Feb.–Aug. 1989	Morales <i>et al.</i> , 1998
Sub-urban, Shenzhen, China	1.3	0.25	Polystyrene Petridishes	April 1999	Tanner <i>et al.</i> , 2001
Urban, Chicago, IL, USA	0.7–4.0	-	KSS with grease	Spring, Summer and Fall 1994–95	Lestari <i>et al.</i> , 2003
Rural, Gopalpura, India	2.1	-	Polypropylene collector	Dec. 1995–Aug. 1997 (Winter)	Satsangi <i>et al.</i> , 2002
Sub-urban, Agra, India	0.6–4.6	0.7	Polypropylene surfaces	Nov.1990–Jan. 1992	Saxena <i>et al.</i> , 1997
Suburban, Uludag Univ.Campus, Bursa, Turkey	30.9	3.0	KSS with GFFs	Sept. 2004–March 2005	Tasdemir and Gunez, 2006
Urban, City of Bursa, Turkey	154.9	7.5	KSS with GFFs	Sept. 2004–March 2005	Tasdemir and Gunez, 2006
Urban, (Pashan) Pune, India	2.66	0.45	Polyethylene funnel	2006–2009	Present study
Industrial ,(Bhosari) Pune, India	3.72	-	Polyethylene funnel	2006–2009	Present study
Traffic, (swargate) Pune, India	3.35	-	Polyethylene funnel	2006–2009	Present study
Rural, Sinhadgad, India	1.72	-	Polyethylene funnel	2006–2009	Present study

highly acidic and so there is no serious environmental threat in the area as far as the acidity of deposition is concerned. WD influenced by the direct continental air mass was characterized with high soil-derived species, whereas that influenced by the mixed continental-maritime air mass exhibited high concentrations of sea-salt derived species. The relationship between soil-derived species in DD and WD were significant. On the other hand, there was no significant correlation between soil derived and anthropogenic pollutants. Local dust around the monitoring sites dominated the DD flux under heavy traffic conditions. Also DD was more than WD at the traffic junction, Swargate. The DD fluxes of water-soluble species were connected with the emission and modification of dust during the long-range transport.

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