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Air Quality Profile of Inorganic Ionic Composition of Fine Aerosols at Two Sites in Mumbai City

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The levels of PM_{10}, $PM_{2.5}$, and NO_2 were studied at a kerbsite and ambient site in Mumbai. Measurements were also made for eight inorganic ions (F^-, Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, NH_4^+, Ca^{2+}, and Mg^{2+}) in the $PM_{2.5}$ fraction. During the study period, $PM_{2.5}$, PM_{10} and NO_2 levels ranged between 11–91, 18–125, and 8–64 μg m^{-3} at a ambient site whereas at the kerbsite the ranges were 10–176, 21–189, and 4–55 μg m^{-3} respectively. Average $PM_{2.5}$ values were 42 μg m^{-3} at ambient and 69 μg m^{-3} at the kerbsite. The measured ions accounted for about 50% of the $PM_{2.5}$ mass. Non-sea-salt (nss) sulfate contributed 91% and 85% of the ionic mass at the ambient and kerbsite sites respectively. Due to biomass sources of K, only about 5% of K^+ was from seas salt. The average equivalent ratio of NH_4^+ to nss- $SO^{2}{}_{4}^{-}$, and NO_3^- was over 1, indicating high source strength of ammonia.

INTRODUCTION

Particulate Matter (PM) is the general term used for a mixture of solid particles and liquid droplets found in the air and are produced by a wide variety of natural and manmade sources. The particulates consist of variety of inorganic and organic components. Experience in India as well as in some other countries has indicated that in atmosphere about 70–80% of PM is inorganic in nature and remaining 20–30% is organic fraction (NEERI 2000) Airborne fine particles smaller than 2.5 μm are responsible for haze formation, visibility reduction and also affect global radiation budget. $PM_{2.5}$ particles are known to have adverse impact on health. Increased levels of mortality and morbidity have been associated with respirable particulate air pollution. Epidemiological studies also find an association between fine particles concentration and increased human health effects (Pope et al. 1995). Atmospheric particulates, especially secondary anthropogenic fine particles have been proved to have a major impact on human health (Dockery and Pope 1996).

Each 10 μg m^{-3} elevations in fine particulate air pollution was associated with approximately with 4%, 6%, and 8% increased risk of cardiopulmonary and lung cancer mortality, respectively (Pope et al. 2002). Average high values have been recorded for PM_{10} in various cities in South Asia, values as high as 900 μg m^{-3} for Lahore; up to 225 μg m^{-3} in Nepal, up to 300 μg m^{-3} of $PM_{2.5}$ concentration has been recorded in Dhaka, Bangladesh, whereas moderate levels of about 60 μg m^{-3} have been found in Sri Lanka (World Bank 2004).

The information available on fine particulate air pollution in India is very limited. In India, it is not yet mandatory to monitor $PM_{2.5}$ levels and very limited information is available on $PM_{2.5.}$ Therefore there is need to understand issues of the fine particle pollution by Characterization and also correlation with PM_{10} and NO_2, which are monitored regularly for National Ambient Air Quality Standard (NAAQS) of India.

As per a limited study carried out by the World Bank, average $PM_{2.5}$ levels in Delhi, Kolkata and Mumbai during winter season was 231, 305, 89 μg m^{-3}, respectively (Chowdhury 2004).

Comparison of $PM_{2.5}$ values recorded at various places in India indicate that it constituted about one half of PM_{10} (TERI 2001).

Some of the characterization studies carried out in India are mainly with respect to TSP, PM_{10} and size distributions of aerosols. Over the last decade, 15–20 studies have addressed the problem of pollution by heavy elements. A lead level along with other heavy metals in PM_{10} was studied by (Gajghate and Hasan 1999; Wilson 1998). In an another study for levels of carbon in RSPM (PM_{10}) were studied by (TERI 2001) and the results indicated that Total Carbon and Black Carbon mass concentration were as high as 341 and 216 μg m^{-3}, respectively. The study of Size fractionated aerosols in Agra reported that 52% of the particles were smaller than 2 μm (Kulshreshtha et al. 1995) whereas in Delhi it was found to be 68% of the PM_{10} mass (Gadi et al. 2000).

Khemani et al. (1994) studied size distribution of aerosols at Chembur-Trombay of Mumbai an industrial site, results

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indicated that coarse particles (>2.1 μm) accounted for 54.6% of TSP whereas fine particles (<2.1 μm) accounted for 45.4% during the rainy season, whereas (Sharma and Patil 1991) reported the results in general more than 85% of aerosol were small than 12.5 μm in another industrial belt of Mumbai city. (Mahadevan et al. 1984) reported that carbon was 16–25% of the total mass of TSP. (Negi 1987) reveals that major elements accounts for 20–30% of aerosol mass and 68–80% of residual aerosol mass seems to consists of carbon, oxygen and nitrogen in India (Vinod Kumar 2001) characterized SPM near traffic junction showed that sulfate content to be 3.7 and 2.75 $\mu g\ m^{-3}$, Calcium content to be 46.2 and 35.9 $\mu g\ m^{-3}$, magnesium 10.2 and 21.6 $\mu g\ m^{-3}$, sodium 20.7 and 18.2 $\mu g\ m^{-3}$, which shows that it was dominated by crustal origin. PM_{10} aerosols in Mumbai during the INDOEX IFP contains significant amount of carbonaceous 30%, ionic 20% and trace metals 4% (Venkatraman et al. 2002).

Previous studies of aerosols in the Indian region at urban, continental and coastal sites have shown overall alkaline nature, with a predominance of cations over anions (Sharma and Patil 1992a,b; Saxena et al. 1996; Momin et al. 1999; Parmar et al. 2001).

In recent years, investigations of atmospheric aerosols have progressed from relatively simple determination of total mass loading to particle size classification and chemical composition. Water soluble fraction of atmospheric aerosols contains many important compounds, which can change the size, composition, number and life time of aerosols owing to their hygroscopic nature. Most of the previous carried out for Mumbai not addressed the $PM_{2.5}$ ionic composition The present study has made an attempt to monitor PM_{10}, $PM_{2.5}$, and NOx for about one year and especially characterize the $PM_{2.5}$ for anions and cations, to understand their sources and occurrence.

STUDY AREA

Mumbai has a mean elevation of 11 meters above sea level and consists of seven islands situated on the Konkan coast. City is located at 18°59′39N latitutude 72° 48′55E longitude. It has a tropical savannah climate. The annual average temperature is 25.3°C, with a maximum of 34.5°C in June and minimum of 14.3°C in January. Average annual precipitation is 2,078 mm with July having the maximum rainfall. High pollution concentrations usually occur in the winter when adverse meteorological situations with weak winds may prevail. In the winter, the predominant local wind direction is northerly (NW-NE). In the summer and monsoon season westerly and south westerly winds are predominant. Sea breeze is normally witnessed during the day with mean wind speeds between 1.5 and 2 m/s. Calm winds occur at night between 22–06 hours.

Mumbai, the commercial and industrial capital of India with a population of about 15 million has been under stress due to increasing urbanization. Though industries have been declining, the city still has 183 air-polluting industries. The impact of traffic on the local air quality is highly visible. City has witnessed tremendous increase in vehicle population with the total of 1,123,562 vehicles in the year 2003. The percentage increase from 1983 to 1993 was 66% and from 1993 to 2003 was 50% (Transport Commissioners Office Mumbai 2003). The current ongoing air pollution measurement programmes reveal that the city is experiencing substantial particulate pollution problem, with frequent and widespread exceedance of TSP (total suspended particulates) and PM_{10} air quality standards (MCGM 2001). An application of dose response equation to PM_{10} during 2000 showed 600, 2620, 1440 chronic bronchitis cases at commercial, industrial, and residential areas in Mumbai city (Abba et al. 2003). Mumbai is amongst one of three cities of the world with highest level of suspended particulate matter (World Development Report 1992). Road traffic, resuspension of road dust, wood burning, domestic refuse burning and furnace oil used in industry were identified as the major sources of PM emission.

The total area source emissions based on population distribution was 10.77 tones/day for SPM. The total emission of SPM from point sources was about 26.82 tonnes/day (NEERI 2004). A recent study on emission inventory of PM sources for Mumbai city (Kumar 2003) shows that industries still dominate the PM emissions load with a contribution of about 54%. The other sources, though appear small, but contribute significantly towards higher exposures to human population due to low elevation of emissions.

Mumbai is turning to a city with mainly commercial activities compared to an industrial city known in the past. This transformation has led to high vehicular usages; we wanted to examine the effect of increase traffic on air quality and compared to the same with an ambient location. In the present study, ambient air quality monitoring for $PM_{2.5}$ PM_{10}, and NO_2 was carried out at Worli (Figure 1). The sampler was placed at 3 m height above the ground on a rooftop. Worli is a residential cum commercial area and very close to the coast. Kerbsite was near the traffic junctions of arterial road in Worli and is influenced by high traffic density of about 78,000 vehicles per/day (NEERI 2004). Monitoring was carried out for 3 seasons: summer (March to May), monsoon (July to September), and winter (November to January) at both sites during April 2003 through March 2004. Sampling was carried out for one year during different seasons. Twelve samples were generated for the whole one year out of which six samples (2 samples for each season) were characterized for ionic composition. Representative samples were chosen from each season to get average values for one year. Sampling days were not consecutive.

EXPERIMENTAL

A Caltech-built $PM_{2.5}$ filter sampler was used to determine the levels of $PM_{2.5}$. Fine particulate matter was collected on one PTFE filter (PALL Life Sciences, 47 mm, 1 μm). The $PM_{2.5}$ samples were collected on 24-hour basis. At ambient site, 2 additional pre washed nylon filters were used to measured fine particle nitrate by the denuder difference method. The

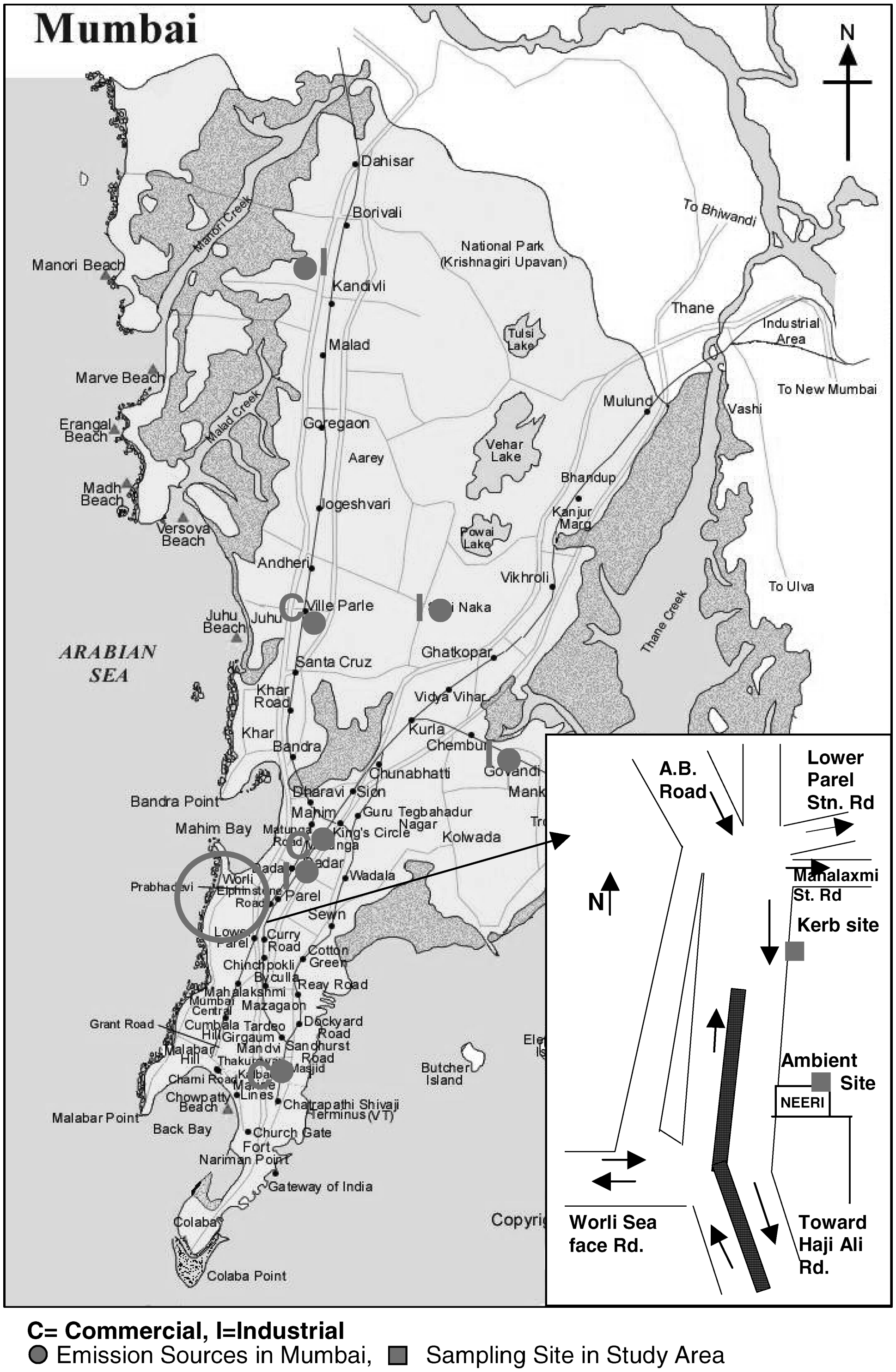

FIG. 1. Study Area (Ambient Site, Kerb Site) Mumbai City, India.

nylon filter located down stream of the magnesium oxide (MgO) recoated diffusion denuder was used in the study. For collection of the fine particles, ambient air was drawn at a rate of approximately 22.5 lpm through a glass inlet line to a Teflon-coated AIHL-designed cyclone separator (John and Reischl 1980) removed large particles according to a collection efficiency curve with a 50% aerodynamic cutoff diameter at 2.5 μm before the air passed through the fine particle collection filters. The air

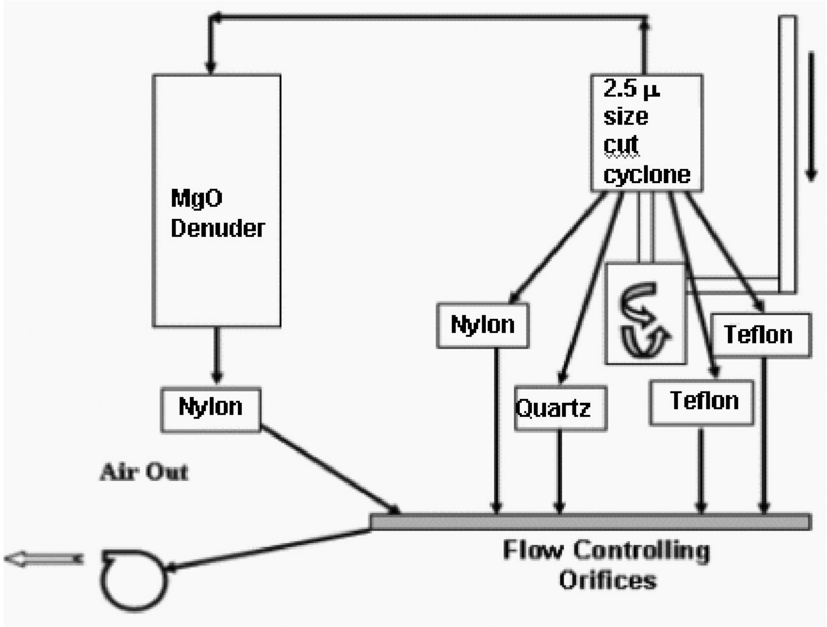

FIG. 2. Schematic Diagram of the Sampling Equipment.

flow rate through each filter was measured before and after each 24 hours sampling period with a calibrated rotameter. The schematic diagram is given in Figure 2. Only one Teflon channel and nylon channel were used in the present study. The second Teflon channel was unused. Since the aim of the study was to determine the anionic composition only, quartz filter was not used.

Ambient air quality monitoring for PM_{10} was also carried out on the same day and period for 24 hours. It was collected on 8″ × 10″ Glass fiber filter (Whatman GF/A) using Envirotech APM 460 NL sampler. Glass fiber filters and PTFE filters were equilibrated in desiccators containing silica gel for 24 hours before and after sample collection and weighed on pre-calibrated Afcoset balance (ER182A) measuring accurately upto 10 μg. Samples were stored in refrigerator until chemical analysis. NO_2 was monitored on 4-hour basis for 24 hours using modified Jacob and Hochheiser modified (Na-Arsenite) method. $PM_{2.5}$ samples were extracted in 25 ml deionised water using ultrasonicator for 1 hour, kept overnight and again ultra sonicated for 1 hour and filtered through Whatman 42 filter. The target ions viz. F^-, Cl^-, NO_3^-, K^+, NH_4^+, Na^+, Ca^{2+}, and Mg^{2+} were analyzed by ion chromotragraph, DIONEX-100 with cation exchange column, Ionpac CS12 and anion exchange column; Ionpac AS4 A-SC with conductivity detector. The eluent used for anion was 1.8 mM Na_2CO_3/1.7 mM $NaHCO_3$ and for Cation 18 mM Methanesulphonic Acid (MSA) with the regenerant flow rate of 1 ml/min. Filter blanks were determined and considered for the calculation purposes. The concentrations of chemical species were calculated by subtracting the values of blank filters from field samples.

RESULTS AND DISCUSSION

PM_{10} and $PM_{2.5}$ and NO_2 (24-hour average values) for the three seasons viz. summer, monsoon and winter are presented in Figure 3. The correlation coefficients of $PM_{2.5}$ and PM_{10} showed r^2 value of 0.83 and 0.85 at ambient and kerbsite, respectively. The average ratio of $PM_{2.5}$ to PM_{10} was 0.68 and 0.70 at the ambient and kerbsite respectively. The major sources present within 1 km radius of sampling stations vehicles, biomass burning, crematoria and bakeries. The source identification in the study area was covered for entire city in a 2 km × 2 km grid by the authors in a separate study (NEERI 2004). $PM_{2.5}$ levels in Mumbai exceeded USEPA, $PM_{2.5}$ standards of 65 μg m^{-3} during the winter season indicating unhealthy air quality during winter time; however, in other season values were lower. Though Mumbai's climate is highly dominated by coastal wind, the impact due to wind is minimal during winter leading to higher levels of PM during this season. Period of monsoon extends for about four months (June–September) during which the high wind along with rains brings the particulate levels down.

The NO_2 levels recorded at Worli site during study ranged between 8–64 μg m^{-3} at ambient site whereas at kerbsite it was found to be in the range of 4–55 μg m^{-3}, respectively. Regression analyses carried out for $PM_{2.5}$ levels and NO_2 showed correlation coefficient for ambient and kerbsite locations 0.85 and 0.84 respectively indicating that NO_2, which is mainly associated with vehicular pollution and responsible for nitrate aerosols has same origin as that of $PM_{2.5}$. It also indicates that ambient site is equally impacted by vehicular emissions. Similar correlation has been established in an another study carried out at Marylebone Road, UK (Airbone Particle Expert Group 1999)

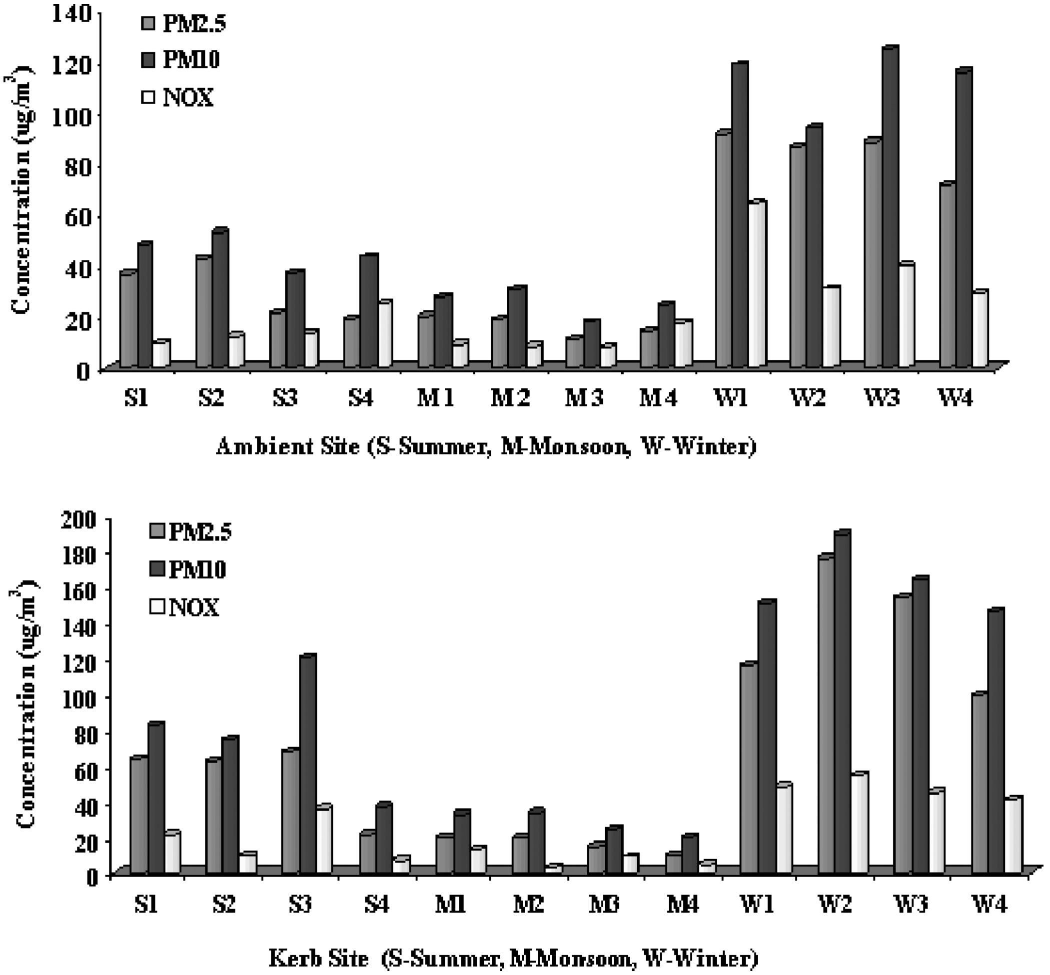

FIG. 3. Levels of $PM_{2.5}$, PM_{10} and NOx at Ambient and Kerb Sites During Different Seasons.

for PM_{10}, $PM_{2.5}$, and NOx. The study showed good correlation between PM_{10} and NOx ($R = 0.88$), $PM_{2.5}$ ($R = 0.79$). Good daily mean enhancement of $PM_{2.5}$ and NOx indicated common source of emission from vehicles.

The average concentration of anions and cations and percentage ionic composition in $PM_{2.5}$ is given in the Figure 4. The ionic composition accounted for about 50 and 49% in ambient and kerbsite respectively. The standard error of the mean for ±1 standard deviation at ambient site and kerbsite for $PM_{2.5}$ mass was 9.20 and 16.36 respectively. The standard errors for mean at ambient site were F^- (0.07), Cl^- (0.09), $NO_3{}^-$ (0.34), SO^{2-}_4 (3.49), Na^+ (0.62), NH_4^+ (0.93), K^+ (0.59), Mg^{2+} (0.2), Ca^{2+} (0.44) whereas at kerbsite these were F^- (0.09), Cl^- (1.28), $NO_3{}^-$ (0.56), SO_4^{2-} (3.11), Na^+ (0.69), NH_4^+ (0.88), K^+ (1.00), Mg^{2+} (0.29), Ca^{2+} (0.59).

Charge balance of the observed aerosol ionic components offers insights into whether the major ionic species comprising the aerosol particles have been identified and quantified. Further in the case of departure from neutrality, we may estimate the possible identities of the missing ionic species viz. carbonate and bicarbonates and their contributions. In addition, measurement reliability can also be evaluated. Ionic difference percentage was 23 and 35% for ambient and kerbsite respectively. The ratio of $\sum$Cations/$\sum$Anions neq m^{-3} was 1.59 and 2.303 at ambient and kerbsite respectively. The correlation between $\sum$Cations neq m^{-3} and $\sum$Anions neq m^{-3} at ambient site showed a very good correlation ($r^2 = 0.96$). At kerbsite the correlation coefficient of ($r^2 = 0.65$) reflected the fairly scatter of the data. The unidentified fraction may include organic carbon and elemental carbon, trace metals and crustal elements. The minimum, maximum and standard deviation values are presented in Table 1.

Among the anionic components, sulfate was the major and fluoride was the minor component at both the sampling locations. The possible sources of sulfate include fuel combustion in vehicles and industries. Sea spray is also source for sulfate. The average percentage of SO_4^{2-} in $PM_{2.5}$ was estimated as 16 and 13% at both ambient and kerbsite, respectively.

The ionic constituents such as SO_4^{2-}, K^+, Ca^{2+}, and Mg^{2+} are derived from either marine or non marine origins, such as anthropogenic and soil emissions. Hence it is necessary to discriminate

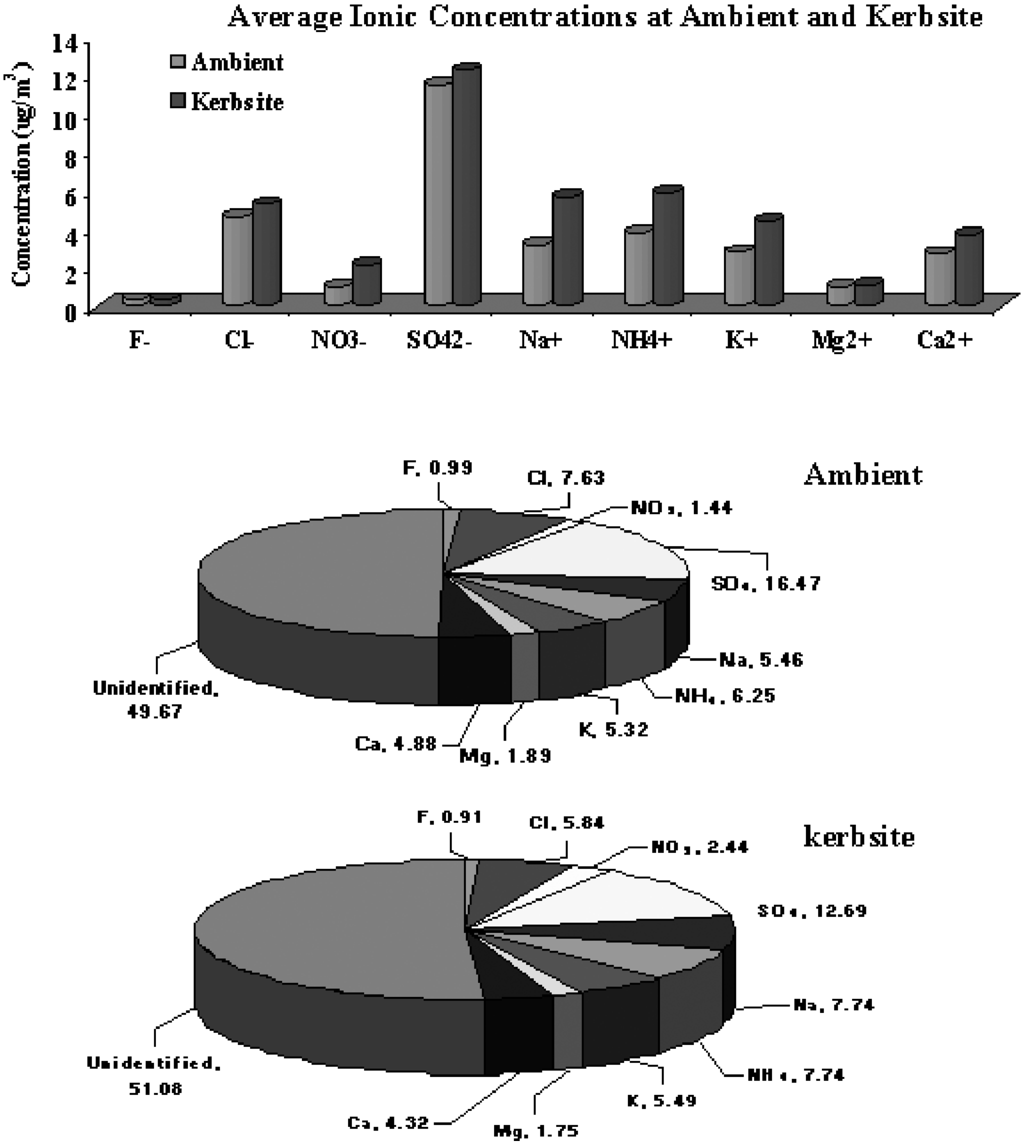

FIG. 4. Average Ionic Concentrations and their Percentage Contribution to $PM_{2.5}$.

sea salt (ss), $SO_4{}^{2-}$, K^+, Na^+, and Ca^{2+} from non-sea-salt (nss) contribution for a city like Mumbai which has three side marine environment. The non marine component was evaluated from the Na^+ concentration on an assumption that all the sodium ions originated from sea. The marine sea salt (ss) and non marine non sea salt (nss) contribution and their percentage to the total ionic content in $PM_{2.5}$ were calculated using ratio like SO_4^{2-}/Na^+, K^+/Na^+, and Ca^{2+}/Na^+ in bulk sea water using the formulae (Keene et al. 1987).

$$\text{nssX} = \text{Total X}_{\text{Measured}} - \text{ssX}$$

where, $\text{ssX} = [\text{Na}]_{\text{Measured}} * [\text{X/Na}]_{\text{Sea}}$

X is an ionic component in aerosol and X/Na is the standard sea water ratio. The nss contribution to fine fraction was over 80% suggesting an anthropogenic origin or natural origin like soil as shown in Table 2.

Potassium ions are commonly found in plants as a constituent of chlorophyll. nss-K^+ contributed 95% of the total K^+ indicating biomass burning at both the sites. Equivalent ratio of Mg^{2+}/Na^+ ratio was found to be in 0.7 and 0.36 at ambient and kerbsite, which is greater than the ratio found in sea water indicating additional source as soil.

Chloride Depletion

The next highest concentration amongst anions was of Cl^- which is of mainly marine origin. Chloride depletion reactions in sea salt aerosols have been observed in many field measurements (Zhuang et al. 1999). The substitution of the sea salt chloride

TABLE 1
Ionic concentration at ambient and kerbsite (μ gm^{-3}) (2003–2004)

	F^-	Cl^-	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}
Ambient									
Average	0.30	4.62	0.97	11.44	3.15	3.78	2.85	0.97	2.73
Minimum	0.10	1.11	0.19	2.10	1.21	1.40	1.51	0.50	1.28
Maximum	0.56	6.86	2.52	20.86	4.61	7.08	5.04	1.64	3.92
Stdev	±0.18	±2.24	±0.84	±8.56	±1.53	±2.30	±1.45	±0.52	±1.07
N	6	6	6	6	6	6	6	6	6
Kerbsite									
Average	0.37	5.30	2.12	12.23	5.67	5.84	4.44	1.08	3.66
Minimum	0.14	1.80	0.89	3.20	2.80	2.30	0.89	0.30	1.19
Maximum	0.76	10.51	4.76	23.35	7.28	8.52	6.60	1.95	5.49
Stdev	±0.24	±3.15	±1.39	±7.62	±1.68	±2.16	±2.47	±0.71	±1.44
N	6	6	6	6	6	6	6	6	6

by nitrate and sulfate changes the optical properties and thus radiation transfer characteristics of the atmospheric aerosols (Ten Brink 1997).

The mechanisms of the Cl^- loss reaction have been widely studied (Cheng et al. 2000). The most commonly discussed reaction is the reaction with acids, followed by the evaporation of HCl to the gas phase. Much attention has been given to formation of NO_3^- and nss- SO_4^{2-}, and more recently organic anions (Pakkanen 1996; Zhuang et al. 1999).

NO_3^- ions present in the air originate from the reaction between NO_2 and O_3, and from hydrolysis of N_2O_5. NO_3^- in the fine mode can be mainly generated by the reaction between HNO_3 and NH_3 while, in the coarse mode, by the reactions between HNO_3 and/or NO_2, and sea salt in a marine environment (Gao et al. 1996; Pakkanen 1996). Chloride depletion in sea salt particles has been explained by the reaction of gaseous or aqueous HNO_3 or H_2SO_4 with NaCl in sea salt particles or by the gaseous SO_2 by sea salt droplet later oxidized to sulphuric acid (Harrison and Pio 1983; Wall et al. 1988; Mamane and Gottlieb 1992).
The sea salt concentrations is defined as (Holland 1984)

$$[Cl^-\ ss] = 1.174\,[Na^+\ measured]$$

where = [Na^+ measured] is the Na^+ equivalent concentrations. The percentage of chloride depletion of sea salt aerosols can be estimated as

$$[Cl^-\ Depletion]\% = \{([Cl^-ss]-[Cl^-measured])/[Cl^-ss]\}*100$$

where = [Cl^- Measured] is the chloride equivalent concentration.

The measured ratio of Cl^-/Na^+ at both the site was less than that of sea water. The percentage chloride depletion was higher at kerbsite than at ambient site as is evident from Table 3.

In order to examine the role of NO_3^- and nss-SO_4^{2-} in depletion, a correlation was sought of NO_3^- and nss-SO_4^{2-} with Cl^-. A good correlation between NO_3^- and Cl^- at ambient site ($r^2 = 0.75$) and at kerbsite ($r^2 = 0.95$) was observed. The correlation between nss-SO_4^{2-} and Cl^- was good with ($r^2 = 0.93$ and 0.96) at ambient and kerbsites, respectively. This concludes that Cl^- loss was related to NO_3^- and nss-SO_4^{2-} formation on sea salt particles. Fine nitrate association with Cl^- leading to depletion is corroborated with no correlation of NO_3^- and nss-SO_4^{2-} with Na^+ at Kerbsite. Good correlation of Na^+ with NO_3^- and nss-SO_4^{2-} ($r^2 = 0.75$ and 0.92) at ambient site could be due to association with sea salt particles. Better correlation between NO_3^- and Cl^- at kerbsite than ambient site showed that in a more polluted place like traffic junction, depletion of Cl^- due to NO_3^- is more visible. "Higher Cl^- depletion can also be contributed to the low Ca^{2+}/Na^+ ratio (competition between Cl^- and Na^+ for acidic gaseous)" (Zhuang et al. 1999).

TABLE 2
Concentration of nss (non sea salt) and their percent contribution to $PM_{2.5}$

	nss-SO_4^{2-} (μg m^{-3})	nss-SO_4^{2-} %	nss-Ca^{2+} (μg m^{-3})	nss-Ca^{2+} %	nss-K^+ (μg m^{-3})	nss-K^+ %
Ambient	11	91	3	96	3	95
Kerbsite	11	85	3	94	4	94

TABLE 3
Equivalent ratios of Cl^- and Ca^{2+} to Na^+ and percentage chloride depletion

	Cl^-/Na^+	Ca^{2+}/Na^+	Cl^- Depletion
Ambient	0.95	1.1	19%
Kerbsite	0.61	0.74	49%

Reaction of NO_3^- and nss-SO_4^{2-} with Soil Particles

HNO_3, H_2SO_4 and SO_2 can also react with carbonates such as $CaCO_3$ and $MgCO_3$ on soil particles to form coarse nitrate and sulfate. The NO_3^- which replaced Cl^- in sea salt particles was estimated on an equivalent basis and subtracted from total measured NO_3^- to give nitrate available for association with soil particles (Venkataraman et al. 1999).

$$[NO_3^-]_{soil} = [NO_3^-]_{Total} - (1.174 * Na^+Cl^-)$$

No nitrate was available for reaction with soil. A good correlation was obtained between nss-SO_4^{2-} and Ca^{2+} ($r^2 = 0.98$ and 0.90) at ambient and kerbsite respectively indicating a nss-SO_4^{2-} association with soil particles containing Ca^{2+}. Calcium is generally believed to be derived from soil and crustal material. There is also a possibility that construction activities in urban areas may play a role in aerosol Ca^{2+} content.

Volatilized Nitrate and Non-Volatilized Nitrate

"Denuder difference method for fine particulate nitrate is employed in addition to collection of nitrate aerosol on Teflon filter. In the denuder difference method, a nylon filter located down stream of a Teflon coated cyclone separator is used to collect fine NO_3^- and HNO_3 vapour, while a second nylon filter located below MgO coated diffusion denuder collects only fine particle nitrate" (Christoforou et al. 2000). Denuder difference for Nitrate was employed only at ambient site.

By denuder difference method percentage of volatilized nitrate was found to be in the range of 16–55% with average percentage loss of 40 as illustrated in the Figure 5. "NO_3^- loss may be due to partial vaporization of NH_4NO_3 aerosol. The nylon filters, if used correctly are effectively a perfect sink for the nitrate content of NH_4NO_3. The difference in nitrate content between the Teflon and Nylon filter samples is a measure of the amount of NH_4NO_3 volatilized from the Teflon filter during the sampling" (Christoforou et al. 2000).

Relation of NH_4^+ with NO_3^- and nss-SO_4^{2-}

Ammonia emitted by decay and decomposition reacts with acidic gases like sulphuric and nitric acid in the air forming ammonium salts. Ammonia is not transported very long distances as it is rapidly converted to NH_4^+ aerosols at a rate of 30% h^{-1} (Asman and Van Jaarsveld 1991). Many studies performed at different places in the world have pointed out that this cation accumulates in the fine particle ranges and correlates well with sulfate, suggesting that ammonium sulfate must be the major ammonium salt (Khemani et al. 1982; Lewin et al. 1986).

The average equivalent ratio of NH_4^+ to nss-SO_4^{2-} was 1.24 and 1.82 for ambient and kerbsite, respectively. The average equivalent ratio of NH_4^+ to the sum of nss-SO_4^{2-} and NO_3^- at ambient and kerbsite was 1.15 and 1.52, respectively. It suggests high source strength of NH_3 to neutralize not only sulphuric acid completely but also HNO_3 almost completely. From these results, NH_4^+ containing particles at Worli site could exists $(NH_4)_2SO_4$ and $(NH_4)NO_3$. Ammonia balances between measured NH_4^+ and calculated NH_4^+ for both forms of sulfate. It is very well correlated as given in the Figure 6. Ammonium balance ($\mu g\ m^{-3}$) was computed based on:

$$NH_4NO_3 \text{ and } NH_4HSO_4 = 0.29(NO_3^-) + 0.192(SO_4^{2-})$$
$$NH_4NO_3 \text{ and } (NH_4)_2SO_4 = 0.29(NO_3^-) + 0.38(SO_4^{2-})$$

Ammonium results were higher in Worli as sewage treatment plant is in close vicinity of sampling station. Higher ammonium levels at Kerbsite can be attributed to unhygienic conditions prevailing in the adjoining areas.

COMPARATIVE STUDY

The values obtained in present study corroborate the data obtained in limited studies carried out earlier in Mumbai. An average ratio $PM_{2.5}/PM_{10}$ of about 0.64 was observed at an ambient site during 2001 (Kumar et al. 2003). The percentage of SO_4^{2-} in $PM_{2.5}$ was reported as 14 and 12% in summer and

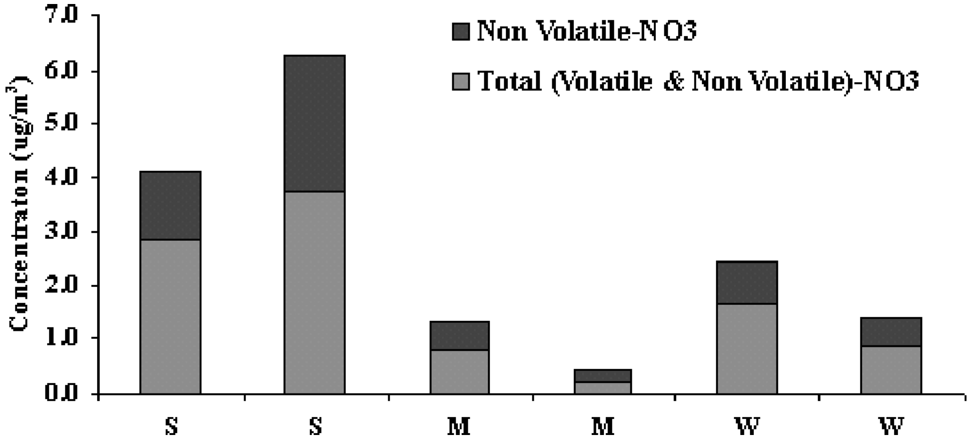

FIG. 5. Ambient Nitrate Levels ($\mu g\ m^{-3}$).

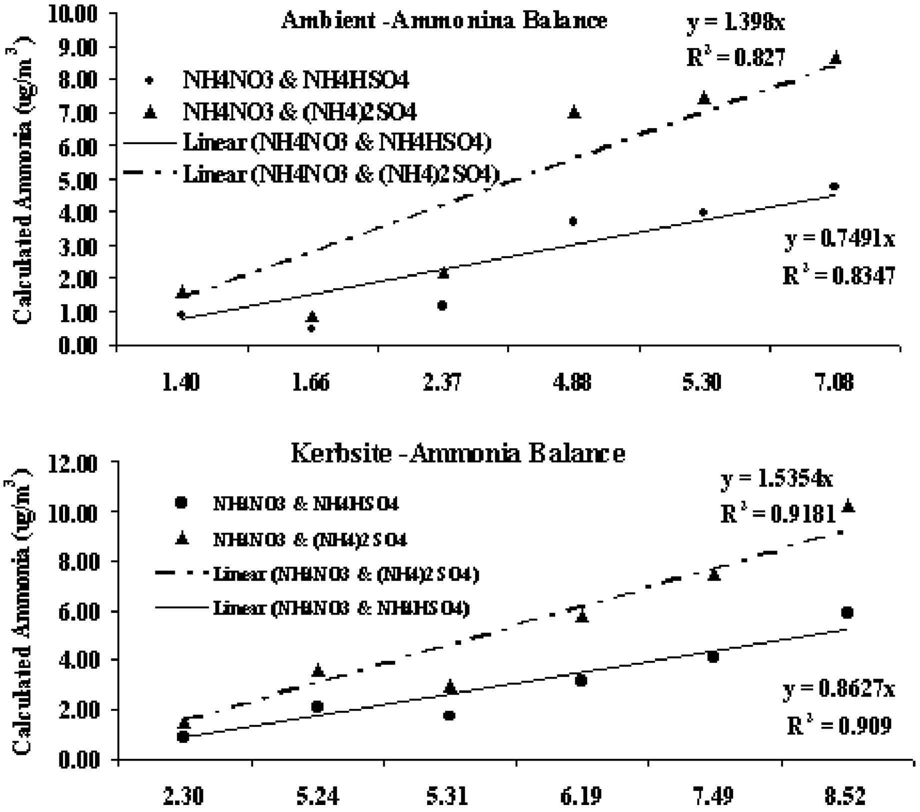

FIG. 6. Ammonia balance between measured NH^{+4} and Calculated NH^{4+}.

winter in Mumbai City (Chowdhury 2004). The average percentage of SO_4^{2-} and Cl^- in PM_{10} at Kalbadevi a commercial site in Mumbai city was reported 30 and 11% respectively (Abba et al. 2002). Predominance of K^+ in the fine mode is attributed to emissions from vegetations and from burning of plant material. It was reported that domestic refuse burning in Mumbai contribute about 23% of the PM_{10} (World Bank 1996), which could be a possible source at the present location. Crematoria and bakeries located within 1 km radius of the sampling locations could be the possible sources as these units use wood as a primary fuel for burning.

The $PM_{2.5}$ levels reported in India are already much higher than the levels reported elsewhere (see Table 4). Fine fraction ($PM_{2.5}$) comprises a large fraction of PM_{10} in all selected cities of India. Though $PM_{2.5}/PM_{10}$ mass ratios in India are relatively similar when compared with other studies in Asia, Europe, and the United States, but the $PM_{2.5}$ and PM_{10} mass concentrations in urban areas are high in India in comparison with other parts of the world.

Total ionic compositions at ambient and kerbsite accounted about 50% of mass. Sulfate, nitrate ammonium ions are major contributors to the total $PM_{2.5}$. During the entire sampling period, these three secondary aerosols accounted for about 24% at both the sties.

The ionic composition in terms of sulfate, nitrate ammonium was reported as 23–33% at background sites and 16–38% in China (Wu et al. 2003), 42% in Taiwan (Chen et al. 1999), and 37% in Hong Kong during winter season (Ho et al. 2003).

The ionic composition observed at various locations in Southern California ranged between 32–45% (Bong Mann et al. 2000), Spain (20–40%) (Artinano 2004), Mexico (30%) (Chow et al. 2002), Milan (9–48%) (Giovanni et al. 2005), and Switzerland (30–48%) (Gehrig 2001). Thus, it is concluded that, the ionic composition measured during present study period at the two sites in Mumbai in terms of sulfate, nitrate and ammonium is less as compared to other countries.

Na^+ and Cl^- are also major component of airborne particulate in coastal areas. As Mumbai is city of islands and thus influenced by sea salt, Na^+ and Cl^- contribute to about 13% in present study. During the study period the percentage of sulfate and ammonium at ambient and kerb site was 23 and 20%, which is less than that reported in Hong Kong (40%), Cheju Island (43%), Hanoi (25%), and Sado (39%) (Cohen et al. 2002). "Composition and association of size differentiated aerosols in a suburban area in an another city away from coast like Agra, 58% of sulfate was found in the fine size fraction (diameter $<2\ \mu$) and 42% in a coarse size fraction" (Kulshreshtha et al. 1998).

The average equivalent ratio of NH_4^+ to nss-SO_4^{2-} and NO_3^- was over 1 in the present study indicating higher source strength of NH_3. This is corroborated with the study in Agra (Parmar et al. 2001). "The Agra study showed that the ratio for the coarse

TABLE 4
The annual concentrations and mass ratios of $PM_{2.5}$ and PM_{10} at different sites around the world ($\mu g\ m^{-3}$)

		Mass ($\mu g\ m^{-3}$)			
Location	Site	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	Reference
Europe					
Switzerland					
Bern	Kerbsite	24.6	40.2	0.61	Hueglin et al. (2005)
Zürich Kasene	City Centre, Courtyard	20	24.4	0.81	
Basel	Suburban	18.9	24.8	0.76	
Chaumont	Rural	7.7	10.8	0.71	
Spain	Madrid Network	34	47	0.72	Artinano et al. (2004)
United States					
United States	Urban/Semi Rural Areas	—	—	0.3–0.27	EPA (2001)
Mexico, South America					
Tlalnepantla (TLA)	Commercial/Industrial	33.39	59.16	0.56	Chow (2002)
Netzahualcoyotl (NET)	Commercial/Residential	55.36	111.50	0.49	
Pedregal, PED	Suburban/Residential	24.56	43.31	0.56	
Asian Region					
Manila, Philippines	Urban	45	—	0.45	David (2002)
Hong Kong Island	Elevated site	31	—		
Cheju Island, S. Korea	Island	18	—		
Hanoi, Vietnam	Urban/Industrial	42	—		
Sado Island, Japan	Island	8.9			
Sihwa, South Korea	Industrial	—	—	0.38–0.89	Seung et al. (2001)
Macao, China[b]					
Avenida da Amizade Road	Kerbsite	28.4	32.3	—	Ye (2003)
Rua de Ferreira do Amaral Road	Kerbsite	39.8	29.2	—	
Rua do Camp Road	Kerbsite	NA	42.3	—	
Rua da Ribeira do Patane Road	Kerbsite	58.4	41.8	—	
Cidade Taipa	Background	32.3	14.1	—	
Taipa Grande	Background	25.1	15.1	—	
Hong Kong[c]					
Hung Hom (PolyU)	Kerbsite	50.92	83.52	0.61	Ho et al. (2003)
Kwun Tong (KT)	Industrial	57.38	73.11	0.78	
Hok Tsni (HT)	Background	42.37	80.01	0.53	
Taichung, Taiwan	Urban-Commercial	40	61	0.65	Chen et al. (1999)
India					
Punjab					
Morinda	Non Industrial	24.0–90.0	—	0.42	Nautiyal (2000)
Mandi-Gobindgarh	Industrial	32.0–100.0	—	0.42	
Delhi[d]					
Pachkuain Road	Kerbsite	94.0–100.0	—	0.35–0.40	Agarwal et al. (2002)
Okhla	Industrial	90.0–102.0	—	0.56–0.66	
Green Park		78.0–109.0	—		
Munirka	Kerbsite	78.0	—		
Nagpur[d]	Several locations	45.0–74.0	—	0.3–0.68	Agarwal et al. (2002)
Chennai	Background	35.0	—	0.50	Ramakrishna et al. (2003)
	Mixed/Traffic/	46.0	—		
	Industrial	54.0	—		
Mumbai					
IIT, Mumbai	Background Urban Site–Period 1		182		Venkataraman (2002)
	Background Urban Site–Period 2		128		
Worli, Mumbai	Residential/Commercial	52	83	0.62	Rakesh Kumar et al. (2003)
Worli, Mumbai	Ambient site	43	63	0.68	Present Study
	Kerbsite	69	98	0.70	

[a] South California $PM_{2.5}$ and PM_{10} values reported as monthly average (24 hour basis).

[b] Monitoring of $PM_{2.5}$ and PM_{10} in China sampling on different days.

[c] Levels reported at Hong Kong studies is for winter season.

[d] Delhi and Nagpur $PM_{2.5}$ values reported as monthly average (24 hour basis).

size fraction varied between 0.7 and 1.4 while that for the fine size fraction ranged between 1.4 and 1.9. These ratios show that the aerosols were mainly alkaline in nature. The alkalinity of coarse mode due was to the higher concentrations of soil derived alkaline component while for the fine mode it was due to acidity neutralization by ammonia."

The Cl^- depletion percentages in the present study were 19–49% at ambient and kerbsite, is comparatively less than other Asian cities like Hong Kong (89%), Cheju Island (96%), Hanoi (83), and Manila (83%) (Cohen et al. 2002). Percentage of Chloride depletion in PM_{10} reported during INDOEX -IFP study in Mumbai was 78% and 40% (Venkataraman 1999).

CONCLUSIONS

$PM_{2.5}$, PM_{10}, and NO_2 measurements were carried out for one year at two sites in Mumbai city. Results indicated that on an average, ratio of $PM_{2.5}$ to PM_{10} was 0.68 and 0.70 at ambient and kerbsite, respectively. $PM_{2.5}$ fractions at ambient as well as kerbsite indicate that fine particles is the major component of atmospheric aerosol in Mumbai City. $PM_{2.5}$ levels in Mumbai exceeded USEPA standard of 65 μg m^{-3} (24 hourly) during winter season. As most of the city area has very high density of traffic for major parts of the day and night, the ambient sites are also highly impacted by vehicular emission, as witnessed from strong correlation of NO_2 and $PM_{2.5}$. Strong correlation of PM_{10} and $PM_{2.5}$ indicates that major part of the PM_{10} particles are fine particles being emitted by fossil fuel combustion. Most of the air quality monitoring data for Indian cities gives only Total Suspended Particulate Matter (TSP) and PM_{10} concentration in ambient air. Currently $PM_{2.5}$ is not monitored; however its presence is responsible for high air quality related morbidity. Therefore, there is an urgent need to monitor $PM_{2.5}$ levels, which would lead to development of air quality guidelines and adoption of appropriate control strategies.

Characterization of $PM_{2.5}$ for anions and cations revealed that the water soluble inorganic ionic composition of atmospheric aerosols accounts for about 50% at both the ambient and kerbsite locations, respectively. Among the detected anions, SO_4^{2-} is the largest chemical component at both the sites and the percentage contribution in $PM_{2.5}$ was 16 and 13% at ambient and kerbsite, respectively. The average equivalent ratio of NH_4^+ to nss-SO_4^{2-} and NO_3^- was over 1, indicating higher source strength of NH_3. Chloride depletion from nitrate and sulfate reaction was estimated during the study and it was corroborated well with low Ca^{2+} and Na^+ ratios. The nss contribution of SO_4^{2-}, Ca^{2+}, and K^+ was over 80% indicating that sources other than sea salts are also present. Biomass burning contributes to K^+ emissions. It is desirable, therefore, to monitor PM_{10} and $PM_{2.5}$ and sulfate aerosol concentration along with $PM_{2.5}$ since it provides complimentary information on air borne particulate composition. This may also have important benefits in understanding any adverse health consequences associated with exposure to $PM_{2.5}$. In addition, concurrent measurement of both PM_{10} and $PM_{2.5}$ fraction will provide further information since wind blown inorganic material is likely to be dominant in the coarse fraction of PM_{10} whereas secondary aerosol is predominant in the fine fraction. Source apportionment studies needs to be taken up for $PM_{2.5}$ pollution in Mumbai city along with rural background for a longer duration with a view to asses the quality of $PM_{2.5}$ and understand the seasonal impacts on the quality and quantity of particulates.

Levels reported in India are already higher than reported elsewhere indicating that more efforts should be taken towards the reduction and establishment of $PM_{2.5}$ standards but before that data base of $PM_{2.5}$ level should be created to know the existing $PM_{2.5}$ levels in different parts of India. There is a need to also know characteristics of particulate matter in background areas along with polluted regions for better understanding of sources.

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