

Variation in the Chemistry of Aerosols in two Different Winter Seasons at Pune and Sinhagad, India

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Abstract

Samples of Total Suspended Particulates (TSP) were collected at Pashan, Pune (semi-urban) and Sinhagad (rural) locations in India, during the winter season of 2003/4, using a high volume air sampler and were analyzed for major water-soluble chemical components using Ion Chromatograph and Atomic Absorption Spectrophotometer. Results of the study were compared with those reported for the same locations in winter season, about two decades back. It was observed that TSP and all of its chemical components showed marked increase in concentrations after the period of two decades at both Pune and Sinhagad. Especially, for NH_4^+ , SO_4^{2-} , NO_3^- and Ca^{2+} , the concentrations increased by about 9, 7.7 and 4 times at Pune and about 23, 10, 10 and 6 times at Sinhagad, respectively. This feature shows the significant impact of anthropogenic activities at both the locations. SO_4^{2-} is still the main acidifying component of aerosols and NH_4^+ dominates over Ca^{2+} in the neutralization of acidity in 2003/4 whereas earlier two decades back, Ca^{2+} was the major neutralizer.

Keywords: TSP, chemical composition, acidity, neutralization potential.

1. Introduction

Aerosols have been a vital link between the source–transport–deposition of various pollutants in the atmosphere. Chemical nature of aerosols is one of the important factors in assessing the impact of aerosols after their deposition on terrestrial as well as aquatic surfaces. Studies on the composition of aerosols, reported from the Indian region are mainly from urban/semi-urban locations (Khemani et al 1982; Kulshrestha et al 1998, Momin et al, 1999; Rao et al, 2001; Ali et al, 2002), whereas, those from the rural or remote regions are very few (Khemani et al, 1985; Mahadevan et al, 1989; Safai et al, 1993 and 2004a). Also the changes in the aerosol composition, after a considerable period of time

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are not sufficiently documented mainly due to the scarcity of past data. Studies were carried out on Total Suspended Particulates (TSP) at Pashan, Pune and at Sinhagad during the winter season of 2003/4. Results obtained from this study have been compared with those reported for winter season at the same locations, more than twenty years back i.e. in 1978 for Pune and in 1982 for Sinhagad (Khemani, 1989). The main objective of the present study is to investigate the variation in concentrations of major acidifying (SO_4^{2-} and NO_3^-) and alkaline (Ca^{2+} and NH_4^+) constituents of aerosols and their effect on the acidification/neutralization potential of aerosols. The results presented here are confined to the winter season only and are not representative of the annual cycle. Due to the frequent inversion conditions and low ventilation coefficients prevailing over this region the in winter (Devara et al, 2001), the pollution potential is rather more in this season and and it is of interest to study the chemistry of aerosols in view of the assessment of atmospheric composition over this region particularly in winter.

2. Sampling Locations and Methodology

TSP samples were collected at Pune ($18^{\circ}32'N$, $73^{\circ}51'E$, 559 m asl), during 29 Dec. 2003 to 9 Jan. 2004 and those at Sinhagad ($18^{\circ}21'N$, $73^{\circ}45'E$, 1400 m asl) were collected during 27 Jan. to 1 Feb. 2004. Pune is about 100 km inland on west coast of India, on the leeward side of Western Ghats. Sampling was carried out on the terrace of the building at about 10 m above the surface, at the Indian Institute of Tropical Meteorology, Pashan, Pune. Pashan is a semi-urban site, located about 10 kms from the center of Pune city. The site is surrounded on all sides by hillocks of variable heights (up to 200 m), forming a valley like appearance. Brick kilns are situated at a distance of about 1 km to the west of observational site. The major urban activity goes on to the east of the observational site. Pune receives marine as well as continental airmasses during different seasons. During June to September, southwesterly and westerly winds, rich with marine airmasses from the Indian Ocean and Arabian Sea, prevail over this region. Whereas, from December through February, during the winter season, easterlies coming over from the land and rich with the continental airmasses, dominate over this region. The industrial sources have been located to the north, north-west of the city. Also, there are more than 13,00,000 vehicles of which, about 10,00,000 are two-wheelers. The population of the city is nearly 25,00,000. Figure 1 shows the growth in population as well as industries in Pune district. Though this figure depicts values for the whole district, it would certainly give some idea about the rapid increase from 1960's up to 1990's in both industries as well as population in this region. However, after 1990, these growth rates have been stabilised (District Social and Economic Summary, Pune District, 1999-2000). The construction activity is still on the rise in all the surroundings of the city especially around the present site.

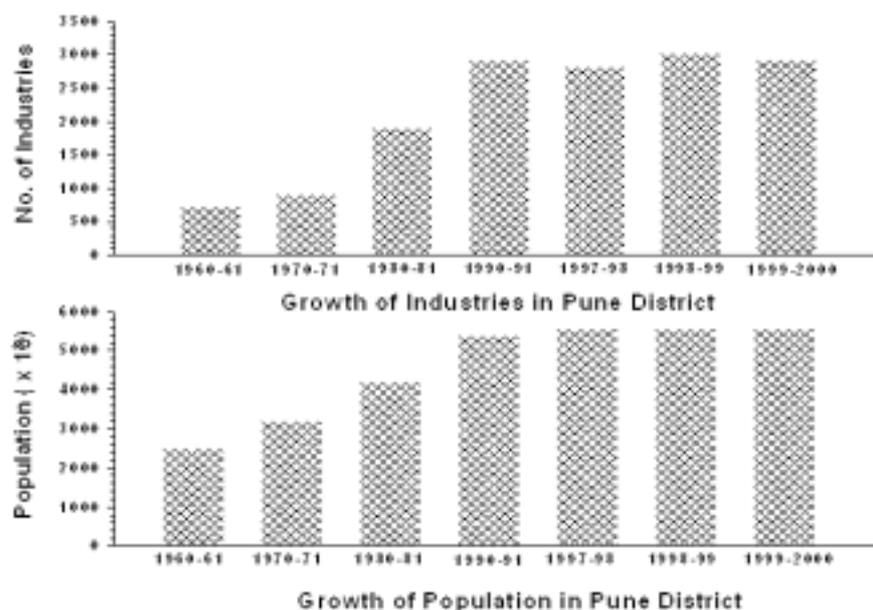


Figure 1. The growth in population as well as industries in Pune district during 1960-2000.

Sinhagad is a historical fort situated on the hill at a mountain top in the Western Ghat region. It is located about 40 km by road, to the southwest of Pune. The top of the hill is flat terrain with an area of about 0.5 sq. km and is surrounded by other mountain peaks of comparable heights. The area is covered with vegetation during monsoon season (June to September). Local inhabitants on the fort are very few. The only noticeable local source of pollution is wood burning, mainly for cooking. A few vehicles, mainly cars, jeeps and two-wheelers, enter the mountain but they have to stop at the entrance of the fort only. The sampling of TSP was carried out in the complex of a Micro wave tower building owned by Bharat Sanchar Nigam Limited (BSNL), Government of India. It is a protected site and trespassers or tourists are not allowed to enter. Thus, it can be considered to be a rural, least polluted site.

A high volume sampler (Anderson Inc., USA, Model 2200) was used for the collection of TSP samples on Whatman No. 41 filter papers of size 8 inch by 10 inch. The average flow rate of sampling was about 1.2 m³/min and the sampling duration for each sample was about 6 hours. The filter papers were equilibrated for humidity before and after the weighing. After the sampling, the filter papers were extracted for water soluble chemical components (Cl⁻, SO₄²⁻, NO₃⁻, NH₄⁺, Na⁺, K⁺, Ca²⁺ and Mg²⁺). Chemical analysis of anions (Cl⁻, SO₄²⁻ and NO₃⁻) was carried out with the help of Ion Chromatograph (Dionex DX-100) using analytical columns Ion Pac-AS4A- SC 4mm, anion micro membrane suppressor ASRS -1, 1.8mM Sodium Carbonate/1.7mM Bicarbonate as eluent and triple distilled water as regenerant. Atomic Absorption Spectrophotometer (Perkin Elmer 373) was used for the analysis of cations (Na⁺, K⁺, Ca²⁺ and Mg²⁺) and NH₄⁺ was analyzed using spectrophotometric method (Weatherburn, 1967). The detection limit for ion chromatographic analysis was about 0.01 ppm whereas, that for the atomic absorption spectrophotometric analysis varied from 0.002 to 0.02 ppm. Field Blanks were taken and analyzed, using procedures similar to

those adopted for samples and necessary correction factors were taken into consideration while computation of ionic concentrations. Detailed procedures of sampling and analysis techniques have been mentioned elsewhere (Safai, 1999).

The earlier observational site at Sinhagad (during 1982) was about 200 meters away from the present site indicating negligible spatial difference however, the surroundings have changed considerably and the human activities (especially tourist related) have increased significantly in the last 20 years. At Pune, the observational site was near city center (about 3 kms) in 1978 whereas the present site is seven km further away from city center. But as far as the human activities (like vehicular and constructional) are concerned, situation at the site in 1978 more or less resembles the present site as the city has expanded vastly in last two decades. Regarding sampling and analysis part, filter media used in the earlier observations was Whatman 41 only, but with circular shape of 10 um dia and average flow rate was about 0.7 m³/min. The duration of each sample was about 3 hrs. in 1978-82 period. A handy high volume sampler (GMW Inc., USA, Model 2000) was used for TSP collection. Analysis of cations (NH₄⁺, Na⁺, K⁺, Ca²⁺ and Mg²⁺) was carried out using same techniques as those in present study. However, anions (Cl⁻, SO₄²⁻ and NO₃⁻) were analysed by spectrophotometric method for which detection limit varied from 0.01 to 0.1 ppm (Safai, 1999). The compatibility of this method with the ion chromatographic technique, was assessed and found to be statistically satisfactory.

3. Results and Discussions

3.1. TSP and its chemical components

TSP concentrations are generally found to be high in the Indian region as compared to those in North America and Western Europe. Especially in the north western India, values ranging from 100 to 500 µg/m³ have been reported which was attributed to the dust from adjoining arid regions (Khemani, 1993). Rao et al, 2001 have reported 108 µg/m³ average concentration of TSP at Pune in the winter season of 1998. In the present study, concentrations of TSP were more at Pune than those at Sinhagad during the winter season of 2003/4 (Figure 2). This could be obviously due to the more human activities at Pashan, Pune, being a semi-urban location. Also, the concentrations of TSP go on decreasing as the altitude of the location increases (Junge, 1963). Mahadevan et al (1989) have reported average TSP concentration of 22 µg/m³ at Mallikadevi, a site in the foothills of Himalayas at an altitude of about 2200 m asl. All the chemical constituents of TSP were more at Pune (except sea salt i e., Na⁺ and Cl⁻). Especially, concentrations of NO₃⁻ and Ca²⁺ showed much high values at Pune, mainly due to high density of vehicular population and also due to the rapid construction activity going on, in and around the city. Whereas, at Sinhagad, these activities are comparatively less and also the extent of vegetation cover is more in the surrounding areas which leaves less open land that generally acts as a source for Ca²⁺ aerosols. As seen from Figure 3, concentrations of TSP and its

chemical constituents especially, NO_3^- , NH_4^+ and Ca^{2+} were more at Sinhagad on holidays (Saturdays and Sundays) than those on non-holidays (Monday to Friday). Concentration of F^- was about $0.04 \mu\text{g}/\text{m}^3$ on holidays whereas on non-holidays, it was below the detection limits. Baum et al (1972) have reported burning of coal as one of the sources for F^- . This feature indicates towards the impact of increased fuel/biomass burning activities at the hill station, generally observed on holidays.

Khemani (1989) has earlier reported studies on TSP and its composition at these two locations and the present results have been compared with those reported by him (Figure 4). Similar feature with more concentration of TSP and its chemical components was observed. However, at both the locations, there is a significant increase in TSP and constituent ionic components. The average concentration of TSP at Pune ($170.8 \mu\text{g}/\text{m}^3$) increased by about 1.7 times in 2004 as compared to that in 1978 ($100 \mu\text{g}/\text{m}^3$). Whereas, at Sinhagad the average concentration of TSP ($112 \mu\text{g}/\text{m}^3$) increased by about 2.5 times in 2004 as compared to that ($45 \mu\text{g}/\text{m}^3$) in 1982. There was about 7 times rise in the concentrations of both SO_4^{2-} and NO_3^- at Pune whereas at Sinhagad, concentrations of both these constituents increased by about 10 times, after the period of two decades. Also, concentrations of Ca^{2+} increased by about 4 times at Pune and that at Sinhagad by about 6 times. But the most significant increase was in the concentration of NH_4^+ , which was more than 23 times at Sinhagad and about 9 times at Pune. Concentration of F^- was found to be very low (average value was $0.01 \mu\text{g}/\text{m}^3$) at Sinhagad during 2003/4 whereas at Pune, the average concentration was about $0.1 \mu\text{g}/\text{m}^3$. Concentration of F^- was not carried out in the earlier study (Khemani, 1989) due to non-availability of Ion Chromatograph. Fluoride aerosols are generally originated from various anthropogenic sources like solid waste disposal, emissions from aluminium and steel industries, use of phosphate fertilizers, etc. (Rose and Marier, 1977). The one order more concentration of F^- at Pune indicates the effect of urbanization and emissions from surrounding industrial areas.

Numerous settlements have come up on the Sinhagad Fort during the past few years as the tourist activities have increased. Consequently, there is an increase in the burning of biomass, as the cheapest and most affordable means for cooking. In addition to it, due to the increasing tourist activities, vehicular emissions of precursor gases for sulfur and nitrogen species have added subsequently to the production of sulfurous and nitrogenous aerosols (Crutzen and Andreae, 1990). Apart from fertilizer use, animal and human excretions are reported to be the major sources for NH_4^+ in aerosols (Galbally and Gillett, 1988). Increase in human population and activities related to animal husbandry were one of the main reasons for the increase in NH_4^+ at Sinhagad and Pune. Vegetative emissions could be an important source for K^+ aerosols at Sinhagad. Similar results have been earlier reported at Silent Valley forest in south India (Safai et al, 1993). In the Indian region, crustal source has been widely referred for Ca^{2+} , Mg^{2+} and K^+ and marine source for Na^+ and Cl^- (Khemani, 1989). Sea salt fractions were calculated to assess the marine contribution towards chemical composition of TSP, using the standard sea water ratios for different components with reference to Na^+ (Keene et al, 1986). Sea salt contribution was computed for Ca^{2+} , K^+ , Mg^{2+} and SO_4^{2-} , because Cl^- is mainly from sea and the marine contribution for nitrogenous components like NO_3^- and NH_4^+ is very meager (as seen from the

bulk sea water composition reported by Horn and Adams, 1966). Table 2 shows the non-seasalt (Nss) contribution for SO_4^{2-} , Ca^{2+} , K^+ and Mg^{2+} . A decrease in marine contribution for all of these components was observed after the period of two decades, at both Pune and Sinhgad.

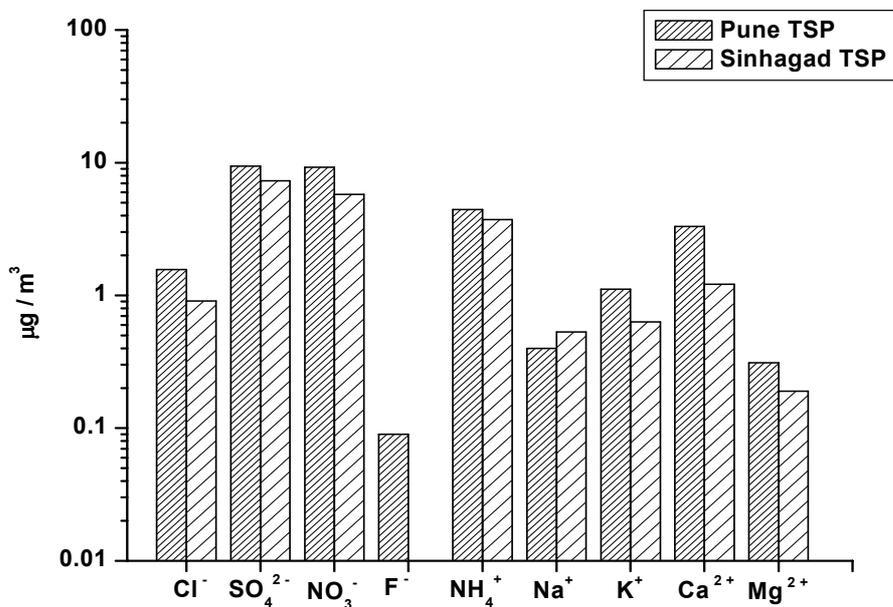


Figure 2. Ionic composition of TSP at Pune and Sinhgad in the winter season of 2004.

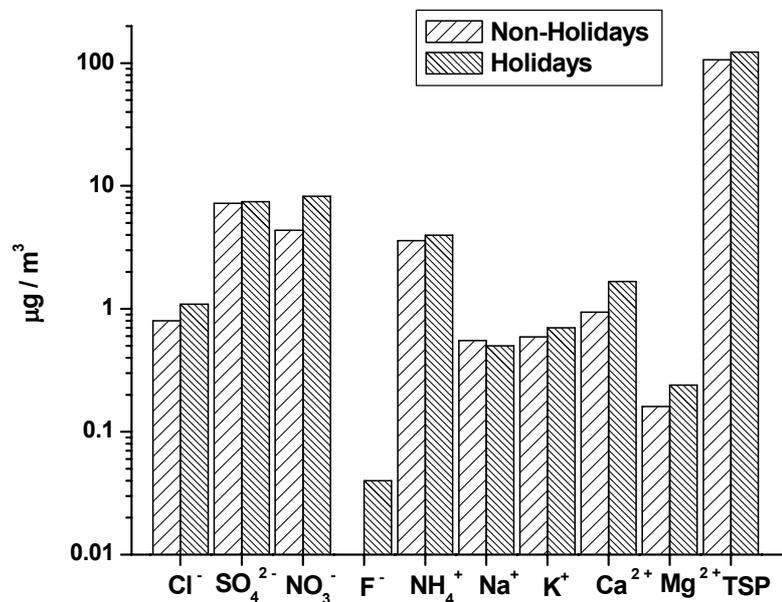


Figure 3. Ionic composition of TSP at Sinhgad on holidays and non-holidays.

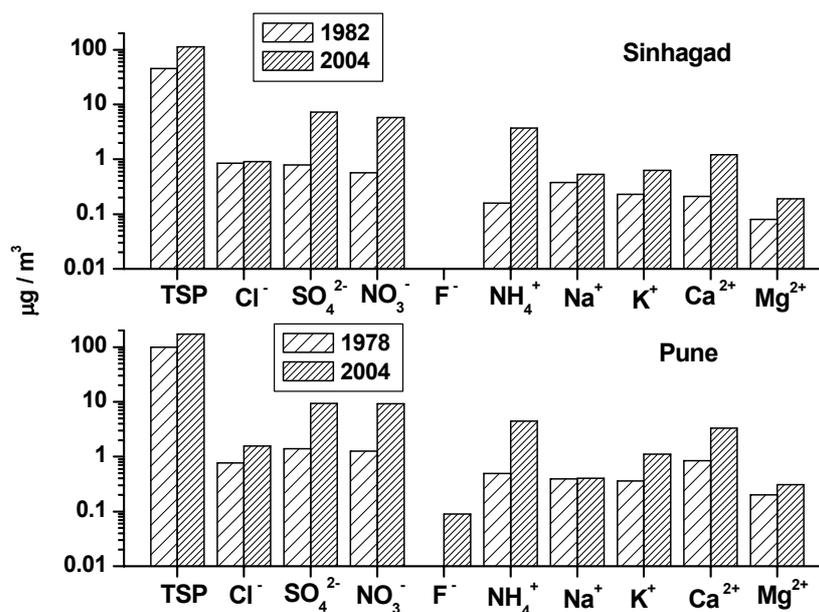


Figure 4. Variation in the concentrations of TSP and its ionic constituents at Pune and Sinhadgad after a period of two decades.

Table 1. Percentage contributions of ionic constituents to the total composition of TSP during the winter season at Pune and Sinhadgad.

Ionic Constituent	Pune % Contribution to TSP		Sinhadgad % Contribution to TSP	
	1978	2004	1982	2004
Cl ⁻	12	5	24	4
SO ₄ ²⁻	16	23	17	26
NO ₃ ⁻	11	17	9	16
NH ₄ ⁺	15	28	9	35
Na ⁺	9	2	17	4
K ⁺	5	3	6	3
Ca ²⁺	23	19	11	10.5
Mg ²⁺	9	3	7	2.5

Table 2. Marine contribution towards SO_4^{2-} , Ca^{2+} , K^+ and Mg^{2+} in TSP at Pune and Sinhagad.

Location	Year	Percentage contribution of marine source			
		SO_4^{2-}	Ca^{2+}	K^+	Mg^{2+}
Pune	1978	7.0	2.0	4.0	23.0
	2004	1.0	0.5	1.0	15.0
Sinhagad	1982	13.0	7.0	6.0	56.0
	2004	2.0	1.0	3.0	33.0

Table 1 shows the percentage contributions of different ionic components to the total composition of TSP during the winter season of 2003/4 at Pune and Sinhagad along with those observed for the same locations, from the data reported by Khemani (1989). It can be seen that the contributions of SO_4^{2-} , NO_3^- and NH_4^+ have increased at both the locations. The increase in the contribution of NH_4^+ is highly significant (two times more at Pune and about four times more at Sinhagad). However, there is a reduction in the percentage contribution of all the other ionic components. Thus, the impact of anthropogenically produced (mainly biomass and fuel burning) aerosols has been increasing than those by natural sources like soil (Ca^{2+} , K^+ , Mg^{2+}) and sea (Na^+ and Cl^-) at Pune and Sinhagad.

3.2. Acidic components of TSP

Out of the two major acidifying constituents i.e. Nss SO_4^{2-} and NO_3^- , contribution of Nss SO_4^{2-} was found to be more towards acidification of aerosols at Pune as well as at Sinhagad. This could be seen from the ratio, $[\text{NO}_3^-] / [\text{Nss SO}_4^{2-}] + [\text{NO}_3^-]$ which shows the relative contribution of NO_3^- to the total acidity (assuming that the total acidity of aerosol is from Nss SO_4^{2-} and NO_3^-). At Pune, this ratio was 0.43 in 1978, indicating about 57 % contribution of Nss SO_4^{2-} to total acidity; similar ratio was observed in 2004, indicating the same contribution. At Sinhagad, this ratio was 0.39 in 1982 and 0.38 in 2004, indicating ≥ 60 % contribution from Nss SO_4^{2-} to the total acidity of aerosols. Thus, the overall contribution of Nss SO_4^{2-} to the acidity was earlier about 60 % and has still remained almost unchanged. Rao et al (2004) have reported results of aerosol studies at Sinhagad for 2002-03, using a Filter Pack method. Acidic fine aerosols and alkaline coarse aerosols were observed and this feature was attributed to the dominance of SO_4^{2-} in fine and that of Ca^{2+} in coarse size particles. Also, they have reported high concentrations of SO_4^{2-} and NH_4^+ in aerosols during winter.

3.3. Neutralization potential of TSP

Considering SO_4^{2-} and NO_3^- as the main acidifying components and Ca^{2+} and NH_4^+ as main neutralizing components, the ratio NP/AP i.e. ratio of Neutralization Potential to Acidic Potential was computed as follows,

$$\text{NP/AP} = ([\text{Nss Ca}^{2+}] + [\text{NH}_4^+]) / ([\text{Nss SO}_4^{2-}] + [\text{NO}_3^-]).$$

The NP/AP value was 1.45 in 1978 at Pune, which decreased up to 1.20 in 2003/4, indicating reduction in the neutralization potential of aerosols. Similar feature has been observed for rain water at Pune at the same location, during the course of two decades, i.e. 1984 to 2002 (Safai et al, 2004b). At Sinhagad, this ratio was about 0.79 in 1982 and 1.10 in 2004, indicating increase in the neutralization potential of aerosols, in spite of increase in the concentrations of SO_4^{2-} and NO_3^- . This phenomenon is mainly due to the substantial rise in the concentrations of NH_4^+ aerosols. The neutralization factors for all the major cations were computed (Parashar et al, 1996). As seen from Figure. 5, Ca^{2+} was the main neutralizing constituent of aerosols at both Pune and Sinhagad. It contributed about 52 % to the total neutralisation potential in 1982 at Sinhagad and about 60% in 1978 at Pune. However, in 2003/4, NH_4^+ contributed more towards neutralization of acidity at Sinhagad (~ 79%) and at Pune (~ about 60%).

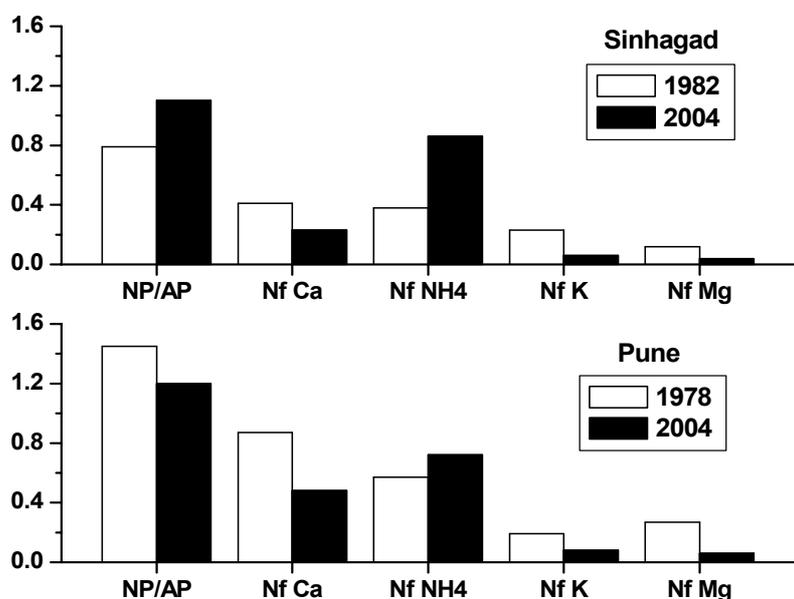


Figure 5. Neutralisation Potential / Acidic Potential of TSP and Neutralisation Factors for different cations of TSP at Pune and Sinhagad

4. Conclusions

Concentrations of TSP and its chemical constituents, mainly SO_4^{2-} , NO_3^- , NH_4^+ and Ca^{2+} have increased significantly at Pune as well as at Sinhagad after a period of about two decades (1980's to 2003/4) indicating the increasing impact of anthropogenic activities. SO_4^{2-} was the major contributor for acidity whereas, NH_4^+ showed more neutralization potential than Ca^{2+} (which was earlier the dominant neutralizing cation).

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