

CHEMICAL COMPOSITION OF ATMOSPHERIC AEROSOL (PM₁₀) AT A SEMI-ARID URBAN SITE: INFLUENCE OF TERRESTRIAL SOURCES

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Abstract. Atmospheric aerosol (PM₁₀) measurements were made at a regional representative semi-arid urban site, Tirupati, India over one-year period i.e. from October, 2001 to September, 2002. The samples were collected on polyflex filters, and analyzed for the major water-soluble ions – F, Cl, NO₃, SO₄, Na, NH₄, K, Ca and Mg, employing ion chromatograph. The average mass of PM₁₀ is found to be 32.75 μg/m³ with a total water-soluble aerosol load (total anion + total cation) of 13.56 μg/m³. Composition of aerosol showed higher concentration of SO₄ followed by Na, Ca and NO₃. Very good correlation is observed between crustal ions Ca and Mg ($r = 0.82$) as well as between crustal and acidic ions; Ca and SO₄ ($r = 0.75$) and NO₃ ($r = 0.67$) and Mg and NO₃ ($r = 0.78$) and SO₄ ($r = 0.73$), suggest that the ionic composition was influenced by local terrestrial sources. The presence of SO₄ and NO₃ may be due to re-suspension of soil particles (formation by heterogeneous oxidation). Ca, Mg and Na are mainly soil derived ones. Correlation matrix with meteorological factors, as well as seasonal distribution of PM₁₀ and its ionic components present a clear trend of higher concentrations during summer due to greater particle release and lowering atmospheric levels during the rainy season due to washout effect. ANOVA results showed the significant variation of composition from season to season. Paired comparisons (DMRT) revealed the occurrence of significant difference in pairs of mean concentration from season to season except within monsoon i.e. between S-W and N-E monsoon.

Keywords: aerosol, India, major inorganic ions, meteorological factors, sources

1. Introduction

Today, air pollution acquires a great concern world wide and led to numerous studies on the chemical composition of ambient aerosols and determination of pollution sources. Atmospheric aerosols are solid or liquid particles dissolved in air, which originate from wide variety of natural and anthropogenic processes that exhibit a large spatial and temporal variability as was reviewed by Andreae (1995). These can vary markedly in size, chemical composition, amount distribution in space and time, and how long they survive in the atmosphere. Atmospheric aerosols influence many atmospheric processes including cloud formation; visibility variation and solar radiation transfer (Shaw, 1987) and also contribute to acidification of clouds, rain and fog. Aerosol particles greatly influence the chemistry and kinetics of the

processes occurring in the atmosphere and can act as reservoir species for many pollutants (Liu and Dasgupta, 1999). The chemical composition of aerosols play a major role with respect to their effect and alters fast and frequently due to new particle formation, scavenging of particles, chemical reactions and/or removal by deposition (Seinfeld, 1998). The particles having the size, $<10 \mu\text{m}$ in diameter (PM_{10}) are enough to be inhaled into the lungs where the chemicals ranging from metal compounds to acid droplets lead to severe health risk (Neas *et al.*, 1995). The health effects of this type have become an increasing concern in recent years and many studies have shown the association between particle concentration and rates of illness or even the mortality (Schwartz and Dockery, 1992). While, the particulate in accumulation mode (0.1 to $2.5 \mu\text{m}$) are most critical with respect to human health, visibility and adverse effects caused by acid precipitation (Pope *et al.*, 1995). Temporal variation represents the influence of meteorological conditions, which are known to affect the particle release and dispersion, and – as a result-human exposure (Ye *et al.*, 2003). For example, the concentrations of aerosol and its chemical components were higher during summer compared with monsoon (Kocak *et al.*, 2004). Therefore, to best document the actual environmental exposure of aerosols, the seasonal and temporal effects on the distribution should be considered.

Chemical characterization of aerosols in ambient air is more important in making clear the sources, transportation and formation mechanism of atmospheric aerosols. In order to understand the sources, behavior and mechanism of formation of the particles and to assess the environmental status of the area, several investigators have reported on the atmospheric particle size distribution and chemical composition for continental, marine background and urban influenced aerosols from various locations throughout the world (Adams, 1983; Ohta and Okita, 1990; Karakas and Tuncel, 1997; Zhuang *et al.*, 1999; Chan *et al.*, 2000). But in Indian context, little has been done to characterize the aerosols and most of the reported results were focussed on major cities (Khemani *et al.*, 1982; Negi *et al.*, 1987; Sharma and Patil, 1992; Safai *et al.*, 1993; Kulshrestha *et al.*, 1995; Parmar *et al.*, 2001).

In the present paper, we report the ionic composition of PM_{10} and the influence of meteorological factors as well as the sources on composition studied over one year period. A comparison has been made with total aerosol measurements from our earlier report (Chandra Mouli *et al.*, 2003), and by measurements reported at Agra (Kulshrestha *et al.*, 1995).

2. Materials and Methods

2.1. SITE DESCRIPTION

Tirupati, a holy pilgrimage town for devotees of Lord Sri Venkateswara is situated in Chittoor district of southern Andhra Pradesh state in India at an altitude of 182.9 m (13.05° N latitude; 79.05° E longitude). Tirupati, as a sampling site, represents

a semi-arid urban area surrounded by major industrial and agricultural activities, and the demographic details of the study area are shown in Figure 1. The town owes its existence to the sacred temple of Lord Sri Venkateswara situated on the seven hills (Tirumala) adjoining it. The town is considered to be high profile center for education, tourism (mainly due to pilgrimage) and business with a population of 3,09,000 (2001 census). Also the town is affected by a floating population of 60,000 per day (average) in the form of pilgrims. The sampling site is located in the center of the study area encompassed with educational institutions surrounded by areas, exclusively devoted to residential, agricultural, industrial activities and forest wealth. Industrial zones are located in a radius of about 15 km on east, west and southern side of the town (Figure 1). Traffic density is minimal with maximum traffic during the day, in the immediate vicinity of the site. However, there is dense vehicular traffic on National Highway situated about 1 km north of the site.

2.2. CLIMATOLOGICAL CONDITIONS

Continental type of climate with three distinct seasons: winter, summer and monsoon are prevailing in the region. The winter months are from January to February, summer season is from March to May and monsoon months are from June to December (S-W monsoon from June to September and N-E monsoon from October to December). Since our measurements were made in University campus besides the meteorological centre, we used its meteorological data. It is clear from the data that the sampling site is mostly wet during the monsoon period and dry in the summer months. The local ambient temperature (which is affected by solar radiation) starts to increase in February and reaches its maximum in April–May (approx. 40 °C) after which decreases to a minimum of 11 °C during January. The basic climatological conditions during the study period are summarized in Table I. During summer the climate is characterized by high daytime temperature ranging between 16.1 and 41.2 °C and low relative humidity 19–72% while during the monsoon period, the temperature ranges between 13.5 and 37 °C and relative humidity varies between 27–98% respectively. The winter months are little bit cool with a temperature ranging between 11 and 33 °C while the relative humidity is 23–95%. The average wind speed in the region is 4.18 mile/h during the study period.

2.3. SAMPLING AND ANALYSIS

Sampling was performed on the roof of Physical Sciences building in University campus at a height of about 12 m above ground level using High Volume Sampler (PM₁₀) (Ruprecht & Patashnik, USA; Model: 2100). Aerosol samples were collected on polyflex filter paper for 24 h monitoring in three different intervals; 07.00–14.00, 15.00–22.00 and 23.00–06.00 h. After sampling, the filters were removed from the filter cartridges and placed in pre-cleaned air-tight glass bottles which were kept at room temperature. Due to hygroscopic nature of the filter, which results in

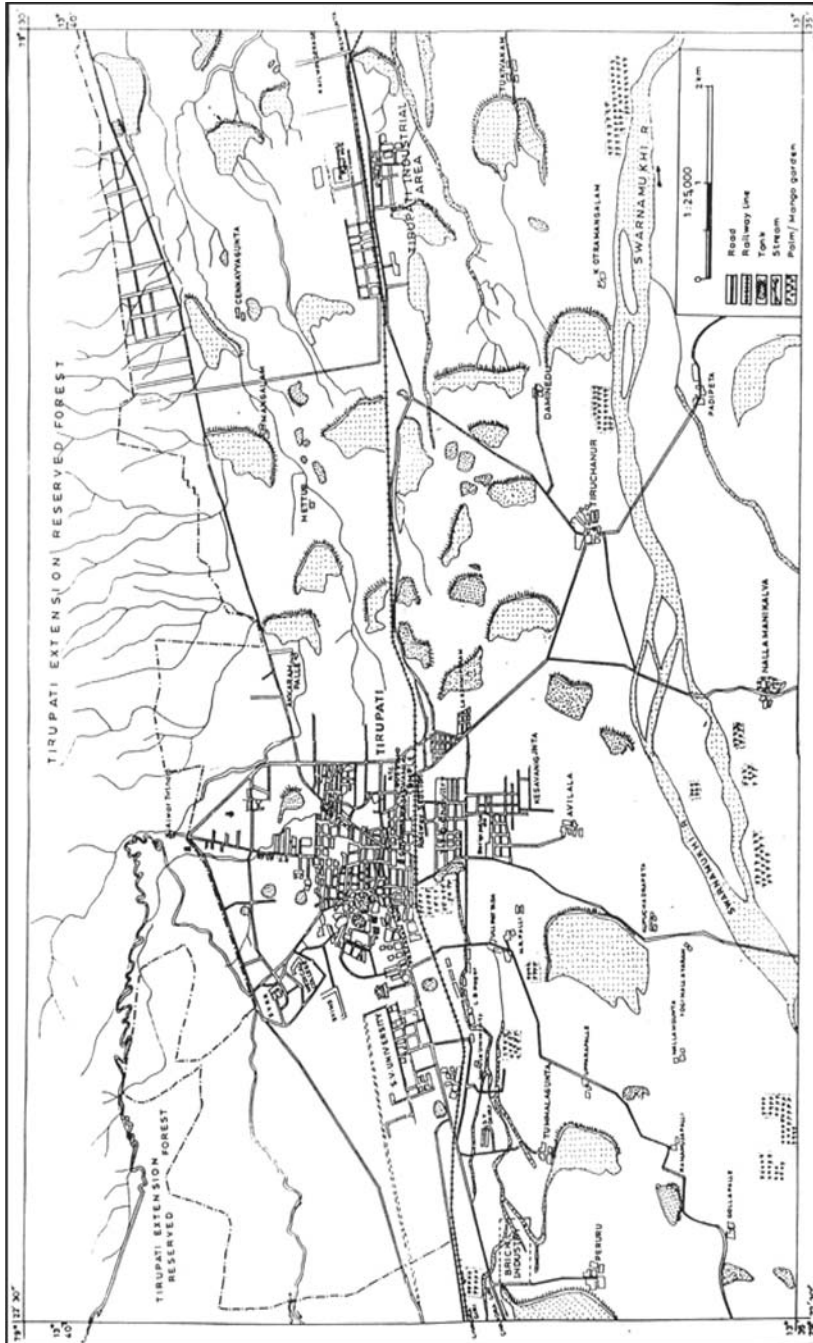


Figure 1. Map showing the demographical details of the study area, Tirupati region.

TABLE I
Monthly mean of basic climatological conditions during the study period at Tirupati

Period	Temperature (°C)		Relative humidity (%)		Wind speed (mile/h)	Rain (mm)
	Max	Min	RHI	RH II		
October	32.31	23.06	79.39	53.23	2.78	5.63
November	29.28	20.92	84.66	64.50	2.38	1.50
December	27.69	17.77	83.00	50.23	3.20	3.40
January	28.76	18.59	82.52	52.74	3.00	1.00
February	30.87	16.66	84.14	43.11	3.66	0.00
March	35.73	20.92	73.45	28.45	3.38	0.00
April	37.88	24.93	66.80	29.77	3.51	0.00
May	38.69	27.61	59.94	30.55	6.31	2.83
June	35.53	26.16	70.13	44.40	5.25	5.25
July	35.77	27.37	57.26	36.03	7.21	0.44
August	34.37	25.20	66.97	43.68	5.59	5.83
September	34.48	24.39	61.13	46.37	3.85	5.04

the change of weight as a function of humidity, filters were carefully equilibrated for 2 minute, in dessicator both before and after sampling to eliminate the effect of humidity and also to obtain accurate particulate measurement. Mass of the aerosol particles collected on each filter was determined by the difference in weights before and after sampling, using electronic micro balance (Mettler AJ100L, Switzerland) with 100 μ g precision. Sampling was carried out with a frequency, once in a week for a period of one year i.e. from October 2001 to September 2002.

For the analysis of water-soluble fraction, one-half part of the particulate loaded paliflex filters were extracted by ultrasonic agitation in 50 ml nanopure water for a period of 1 hour. These extracts were filtered through a filter paper (Whatman No. 41) into pre-cleaned polypropylene bottles. These were refrigerated at 4 °C and used for the analysis of major inorganic ions. The solutions obtained were analyzed for major inorganic ions: F, Cl, NO₃, SO₄, Na, NH₄, K, Ca and Mg by ion chromatograph (IC). Anion analysis was performed by Metrohm Modulor IC: consisting of 709 IC pump, 733 Separation center, 732 Conductivity detector, 753 Suppressor module and 714 IC Metro data, while the cations as well as NH₄ were analyzed by Metrohm 761 Compact IC, using 1.3 mM Na₂CO₃/2 mM NaHCO₃ and 4 mM Tartaric acid/0.75 mM Pyridine-2,6-dicarboxylic acid as eluent respectively. In both cases, the samples were filtered through 0.2 μ in-line filtration cartridge. In order to check the instrumental errors and for procedural contamination, quality control (QC) standards prepared from commercially available (E-Merck) 1000 ppm stock standards of all the ions were used. Each calibration curve was evaluated by analysis of these standards ranged between 0.02 to 10 ppm in triplicates, i.e. before, during and after the analysis of a set of samples. Analysis was repeated if QC fell

outside $\pm 7\%$. The column and other operating conditions employed for analysis of both the ions as well as the detection limits, precision and accuracy of the method are same as reported in our previous work (Chandra Mouli *et al.*, 2003, 2005). Filter blanks were also prepared in the same manner and analyzed for both the inorganic species. Blank values were found to be below detection limits. To test, for complete extraction, filters, which had been extracted in the above manner, were subjected to the same procedures. The values of both the ions were below detection limits in the second extract.

2.4. STATISTICAL ANALYSIS

The statistical analysis of the obtained data was performed using SPSS (SPSS Inc., Chicago, IL, USA) software. Summary of statistics and correlation analysis has been carried out in order to characterize variation in individual measurements as well as to estimate the relation between the considered parameters. The single factor analysis of variance (ANOVA) followed by Duncan multiple range test (DMRT) were used to determine the significance of variations between the seasons.

3. Results and Discussion

3.1. MASS CONCENTRATION AND IONIC COMPOSITION

The average mass of PM_{10} along with concentrations of water-soluble major inorganic ions (F, Cl, NO_3 , SO_4 , Na, NH_4 , K, Ca, and Mg) as well as their statistical results are presented in Table II. The average mass of PM_{10} was $32.75 \mu g/m^3$ which is higher than the value observed at Silent Valley ($30 \mu g/m^3$) (Safai *et al.*, 1993)

TABLE II
Average concentration of atmospheric aerosol (PM_{10}) and its ionic components ($\mu g/m^{-3}$) along with statistical results

Component	Mean	SD	CV	Skewness	Kurtosis	Minimum	Maximum
F	0.08	0.02	29.81	0.27	-0.57	0.04	0.14
Cl	1.06	0.36	33.92	0.42	0.01	0.22	1.85
NO_3	1.05	0.42	39.53	1.03	-0.29	0.59	1.97
SO_4	5.72	1.19	20.77	0.87	1.13	3.87	9.43
Na	3.37	0.74	21.97	0.98	0.20	2.32	4.98
NH_4	0.19	0.04	22.24	-0.14	-1.06	0.11	0.28
K	0.41	0.08	20.12	0.93	0.33	0.31	0.62
Ca	1.47	0.52	35.33	0.78	0.12	0.58	2.80
Mg	0.19	0.07	35.74	0.15	-0.49	0.06	0.35
PM_{10}	32.75	6.11	18.66	1.49	2.97	25.27	54.71

TABLE III
Average concentrations of different components in $\mu\text{g}/\text{m}^3$

Species	Present study	Previous report ^a	Agra ^b
F	0.08	0.02	0.89
Cl	1.06	0.23	6.78
NO ₃	1.05	0.84	8.37
SO ₄	5.72	2.48	14.67
Na	3.37	0.75	2.97
NH ₄	0.19	0.77	6.52
K	0.42	0.22	2.50
Ca	1.47	0.38	3.02
Mg	0.19	0.05	1.22
Aerosol mass	32.75	54.75	190

^aChandra Mouli *et al.* (2003).

^bKulshrestha *et al.* (1995).

where as lower than the value observed at Pune ($73.5 \mu\text{g}/\text{m}^3$) (Khemani *et al.*, 1982). Table III compares the average concentration of the major inorganic ions of aerosol in the present study with the previous study at the same site (Chandra Mouli *et al.*, 2003) as well as for other semi-arid site, Agra (Kulshrestha *et al.*, 1995). It is evident from the Table III that concentrations of all the ions are greater than the previous study except for NH₄. Lower concentrations in the previous study are due to the frequent rain showers, washout effects and also low emission from soil, while the higher concentration of NH₄ is due to intense gas phase reactions. Where as the concentration of all the ions are too lower than the concentrations observed at Agra during winter (Kulshrestha *et al.*, 1995).

The average total water-soluble aerosol load (total anion + total cation) was $13.56 \mu\text{g}/\text{m}^3$. The percentage distribution of each ion to the total ionic mass showed a trend $\text{SO}_4 > \text{Cl} \cong \text{NO}_3 > \text{F}$ for anions and $\text{Na} > \text{Ca} > \text{NH}_4 > \text{Mg} > \text{K}$ for cations. From the results, it is evident that SO₄ contributed maximum to the water-soluble aerosol mass (42%) followed by Na (25%). All the cations combinely accounted about 41% to the total water-soluble aerosol mass. The lower contributions of Ca and Mg suggest that they occur mainly as insoluble carbonates, bicarbonates and silicates. Positive kurtosis values have been obtained for Cl, SO₄, Na, K, and Ca that indicate a relatively peaked distribution. This signifies that there are sporadic high emissions of these elements from certain emission sources causing a peaked distribution. Skewness calculations in a data set characterize the degree of asymmetry of a distribution around its mean. In our data set only NH₄ has shown a negative skewness, which could be the result of probable losses of the component due to chemical reactions of NH₃ with acidic gases and the remaining all ionic components showed a distribution with an asymmetric tail extending toward values

The average concentration of SO₄ is 5.72 μg/m⁻³ (Table II). Primary emission, gas-phase nucleation or condensation cannot explain the occurrence of SO₄ particles in fine mode. A possible mechanism could be aqueous-phase oxidation of SO₂ in cloud droplets (Seigneur and Saxena, 1988). Contribution of sea-salt sulfate will be insignificant, because this sampling site is an inland site away from the coastal areas. This is further corroborated by high SO₄/Na ratio (1.70). Hence in this area, sulfate may be soil derived or formed by reactions of gas-phase SO₂ on the wet surface of basic soil particles. The average concentration of NO₃ is found to be 1.05 μg/m⁻³ (Table II). Fine particles of NO₃ may be formed by the homogeneous gas-phase transformation of NO_x to HNO₃, followed by the reaction with NH₃ to form NH₄NO₃, which is highly volatile and can remain stable in the particulate phase, if the product of gas-phase concentration of ammonia and nitric acid [NH₃][HNO₃] in the air exceeds the equilibrium product (Seinfeld and Pandis, 1998). The Dairy Farm existed near by the site, with a livestock of around 500 herds of cattle might have act as a major source of NH₃ gas and the suitable humidity and temperature conditions would be favored for the formation of NH₄NO₃.

The average concentrations of Na and Cl are found to be 3.37 and 1.059 μg/m⁻³ respectively (Table II). The ratio of Cl/Na in aerosols (0.32) is less than the sea-water ratio (1.8) indicating a deficiency of chloride relative to the Na concentration. Since the site lies about 150 km away from the sea, loss of Cl would have occurred as the air masses traverse such long distances over continental regions from the sea. Loss of Cl from particulate may be ascribed to reaction of H₂SO₄ and HNO₃ with NaCl to produce HCl (Chandra Mouli *et al.*, 2003). Several investigators have examined the variation of Cl/Na ratio with particle size. Cadle and co-workers (Cadle and Robins, 1960) observed that NaCl aerosol in the presence of 0.1–100 ppm NO₂ at relative humidities of 50–100% lost Cl from the particulate phase. Average concentration of the fluoride is seen to be 0.08 μg/m⁻³ (Table I). Fluoride may be contributed by emissions from industrial activities like brick kilns and lime pulverization units situated in and around Tirupati.

The average concentration of NH₄ is seen to be 0.19 μg/m⁻³ (Table II). Fine particulate ammonium may originate by the reaction of NH₃ vapors with acidic gases such as H₂SO₄, HNO₃ and HCl or ammonia vapor may react or condense on an acidic particle surface of anthropogenic origin. The average concentration of K is found to be 0.41 μg/m⁻³ (Table I). Normally, soil is considered to be the main source of K. The fine particles of K may be released into the atmosphere by burning of plant material (Cooper, 1980); however, vegetation might have act as another source of K particles through guttation (Kleinman *et al.*, 1979), which takes place under hot and humid conditions and is likely to be favored at this site also. The average concentrations of Mg and Ca are found to be 0.19 and 1.47 μg/m⁻³ (Table II). Generally soil is considered to be the main source of both Mg and Ca. Fine particles may be of anthropogenic origin. Higher concentrations of Ca reveal that, it may be contributed by the emissions of industrial activities like ferrous and non-ferrous foundries.

Thus it is surmised that the aerosol at Tirupati region is predominantly influenced by the local terrestrial sources. Since the study area is with dense of forest and agricultural activity, lead to the contribution of basic cations, and K through the respiration of vegetation (Kleinman *et al.*, 1979). SO₄ may originate from soil particles and the possible mechanism could be the aqueous-phase oxidation of SO₂ in cloud droplets (Hegg and Hobbs, 1982; Pandis, 1992), while the NO₃ may be formed by homogeneous gas-phase transformation of NO_x to HNO₃, followed by the reaction with NH₃ to form NH₄NO₃. Apart from these the industrial and urban activities may also play a little role in the contribution of acidic ions.

3.3. SEASONAL VARIATION

The meteorological factors like temperature, relative humidity, wind velocity and rainfall can influence the atmospheric levels of aerosol and its components (Marcazzan *et al.*, 2002). Table V presents the Spearman correlation coefficient matrices between the concentrations of PM₁₀ as well as its ionic components and the meteorological factors. The correlation between the measured parameters and temperature and wind velocity are positive except in the case of PM₁₀ and Na with wind velocity. But the correlations are low and negligible in most of the cases (Table V). Significant correlations were observed between crustal ions (Ca and Mg) as well as acidic ions (SO₄ and NO₃) with the meteorological factors, which suggest that higher wind speed and temperature may cause greater release of dust particles due to erosion and re-suspension (Jones and Harrison, 2004). The increased concentration of acidic ions during summer months may be attributed to an enhanced formation of a secondary aerosol due to higher temperature and humidity. Both RH and rainfall showed a negative correlation with measured parameters (Table IV). During the high humidity episodes, the particle hygroscopic growth and condensation may result in an increase of the coarse particle (Mirme and Ruuskanen, 1996) and also may reduce the particle release from the surface (Jones and Harrison, 2004).

Although air-borne concentrations depend on many processes, including upwind chemical processes, removal by wet and dry deposition and variability in source regions, the wet deposition is an effective mechanism for the removal of aerosols, especially in the size range of 0.1–10 μm (McGann and Jennings, 1991). It is

TABLE V
Correlation matrix between meteorological factors and PM₁₀ as well as its ionic components

	PM ₁₀	F	Cl	NO ₃	SO ₄	Na	NH ₄	K	Ca	Mg
Temp	0.23	-0.02	0.21	0.32	0.26	0.03	0.00	0.10	0.50	0.57
RH	-0.42	-0.33	-0.45	-0.59	-0.46	-0.36	-0.25	-0.21	-0.73	-0.76
Wind vel.	-0.05	0.00	0.12	0.15	0.16	-0.08	0.07	0.03	0.49	0.42
Rain	-0.41	-0.52	-0.38	-0.37	-0.41	-0.48	-0.51	-0.28	-0.43	-0.29

therefore expected that low concentrations of the atmospheric constituents will occur during and following precipitation events. Mean concentration of each water-soluble inorganic ion components as well as PM₁₀ during each season along with ANOVA was depicted in Figure 2. All the ions along with PM₁₀, in general, showed

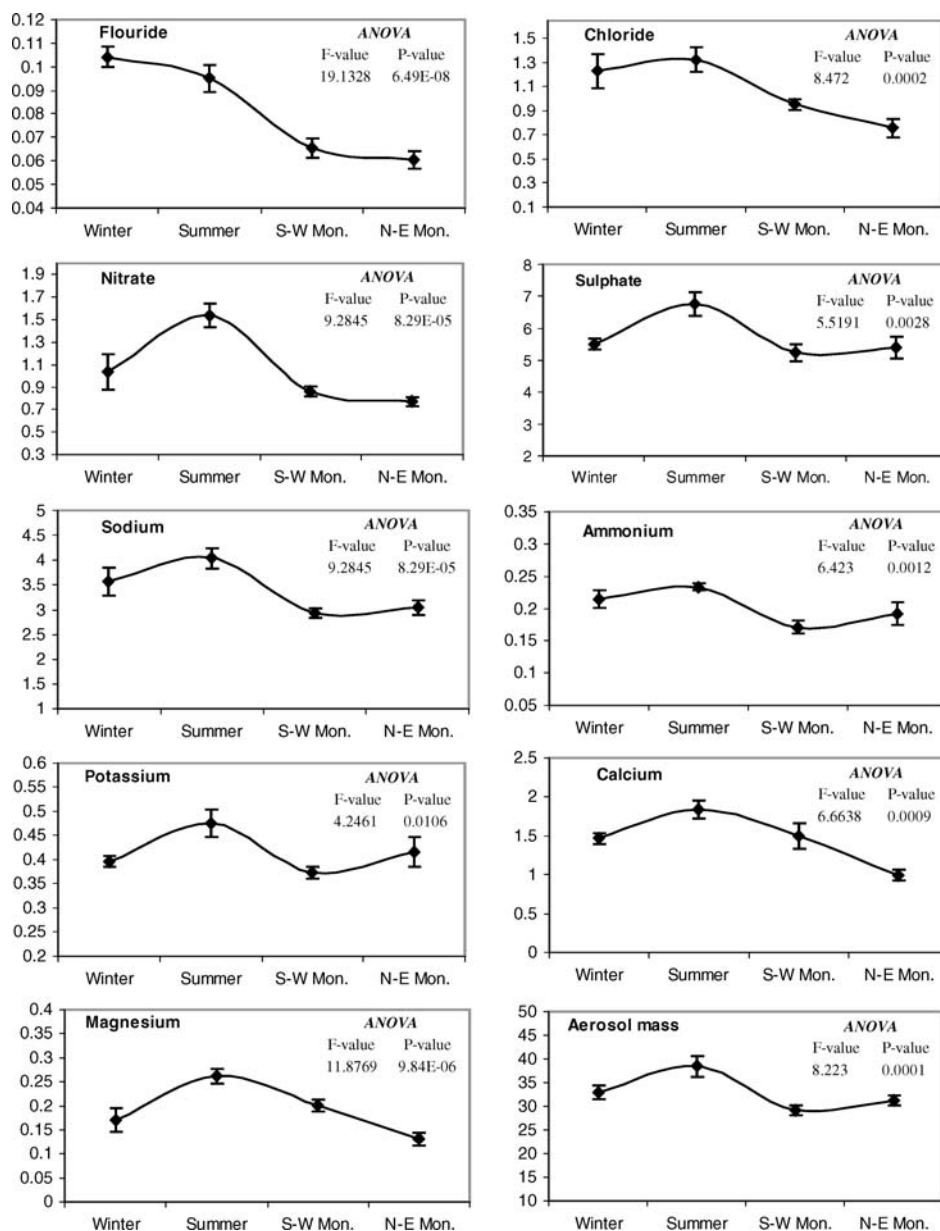


Figure 2. Seasonal variations of mean concentration of aerosol and its ionic components (bars indicate the standard deviation).

similar seasonal variations with a greater variation from season to season. ANOVA resulted that the means are likely different and are more significant ($P < 0.05$) in the case of Cl, SO₄, NH₄, K, Ca and aerosol mass. DMRT resulted the pair-wise comparisons of the measured parameters i.e. the difference in mean of each parameter from season to season. Results showed the significant difference between almost all pairs of means ($P < 0.05$) from season to season, except in few cases such as; between winter and summer for F ($P = 0.213$), Cl ($P = 0.476$), NH₄ (0.282); winter and S-W monsoon for NO₃ ($P = 0.186$), SO₄ ($P = 0.566$), K ($P = 0.491$), Ca ($P = 0.884$), Mg ($P = 0.175$) and PM₁₀ ($P = 0.478$); winter and N-E monsoon for SO₄ ($P = 0.840$), NH₄ ($P = 0.235$), K ($P = 0.573$) and Mg ($P = 0.126$); and S-W and N-E monsoon for F (0.438), Cl ($P = 0.105$), NO₃ ($P = 0.448$), SO₄ ($P = 0.702$), Na ($P = 0.643$), NH₄ ($P = 0.176$), K ($P = 0.167$) and PM₁₀ ($P = 0.310$) suggest that there is no more significant variation in PM₁₀ and its components within the monsoon i.e. between S-W and N-E monsoon. The concentrations of all the ions except fluoride, relatively low during winter season subsequently raised to a maximum during summer followed by the decrease during monsoon season (Figure 2). Figure 3 shows the seasonal distribution of aerosol mass and its ionic components during the study period, which presents a clear trend of lowering atmospheric levels during monsoon season due to washout effect and higher levels during summer due to high temperature and wind velocity and the low RH, which increases the particle release from the surface (Jones and Harrison, 2004). From the pattern of profiles, it can be clearly visualized that the concomitant increase and decrease in aerosol mass is directly related to the ionic concentration and the aerosol mass in the atmosphere is also dependent on the rainfall and on other climatological conditions subsequently.

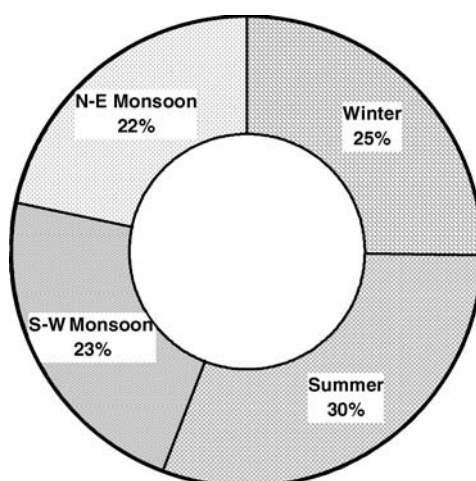


Figure 3. Seasonal distribution of ionic components at Tirupati.

4. Conclusions

This study reveals that the ionic concentrations of total aerosol are significantly lower than that of concentrations in respirable particles (PM₁₀). As the site is away from the sea, influence of marine sources is almost negligible on aerosol contribution. However, it was found that the crustal ions as well as with acidic ions are well correlated. Hence, it may be inferred that a major part of the atmospheric aerosol is contributed from terrestrial sources, mainly soil. The levels of aerosol were highly varied from season to season and are significantly higher during summer and lower during monsoon. Correlation with meteorological factors, seasonal variation of means of atmospheric components and their pair-wise comparison are statistically sound and suggested the considerable impact of climatology. Skewness values indicate asymmetric distribution of the components signifying the pronounced effects of climatological conditions and source strength effects.

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