

Study of precipitation chemistry over an industrial city

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ABSTRACT: The growing trend of industrialization has marked impact on the atmospheric chemistry around the globe. Such impacts are also predicted for developing countries like India. Acid precipitation due to secondary pollutants is one of them. The present investigation was conducted with an aim of studying the precipitation chemistry over Asansol city, India during the monsoon season. The rain water samples, collected on event basis during the period of June to September 2006, were analyzed for HCO_3^- , Cl^- , SO_4^{2-} , NO_3^- , NH_4^+ , K^+ , Na^+ , Mg^{2+} , Ca^{2+} and pH using standard procedures. The study revealed that the precipitation was alkaline in nature with mean pH value of 6.08. Dominant anion in rain water samples was HCO_3^- (36 %), whereas Ca^{2+} was highest among the cations (53 %). The observed acidic events, based on pH value, were only 17 %, indicating the alkaline components effectively neutralize rain water over the area. This was also supported by the total alkaline to acid ion ratio of 1.05. The ratio of $(\text{NO}_3^- + \text{Cl}^-)$ and SO_4^{2-} was >1 , indicating that acidic properties were influenced by HNO_3 and HCl . In the ratio between $(\text{Ca}^{2+} + \text{NH}_4^+)$ and $(\text{NO}_3^- + \text{SO}_4^{2-})$, values >1 were obtained, indicating significant alkaline influence, which effectively prevented acid depositions in the area.

Key words: Rain water, ionic character, acid rain, air pollution, urban area

INTRODUCTION

The clean air mass of the troposphere, the lower most layer of the atmosphere, collects both natural and anthropogenic products as it moves across the surface of the earth. While natural products include emissions from volcanic activity, sea salt sprays, dust storms and forest fires, the man made emission sources are much varied and more detrimental. In an urban area these manmade sources include automobile emission, industrial operations, fuel combustion, open burning, bacterial decomposing of nitrogenous materials, municipal waste disposal, etc. Pollutants that enter directly in to the atmosphere are termed primary pollutants, where as those formed by reaction of the primary pollutants with each other or other components of the air are called secondary pollutants. The latter products then travel in the direction of wind flow and return to earth as particles, droplets or chemicals dissolved in precipitation, among which rain water is most common. The chief contaminants of rain water, which has a natural pH of 5.6, include SO_2 , SO_3 and NO_x among others. They dissolve as strong mineral acids like sulphuric acid and nitric acid in the rain water

and bring down the pH to less than 5.6, causing severe impact on the natural and urban environment. The concentrations of these acids are too weak to cause direct burn, but they produce other effects and cause serious ecological instability depending on a variety of biological, chemical and meteorological factors in soil, water bodies, vegetation, animals and buildings and other man made structures. It causes not only serious health effects, but also economic losses and disrupt ecosystem functions.

In India the total Sulphur emissions are expected to rise from 4,400 kilotons in 1990 to 10,900 kilotons in 2010 and 18,500 kilotons in 2020. Instances of acid rain are increasing and mostly being reported from metropolitan area (Anjaneyulu, 2002). Acid Rain studies have been carried out by Indian Institute of Tropical Meteorology for the last three decades. In order to study the spread of acid rain over India, the pH values and precipitation chemistry have been studied by various authors over the years. In India works that can be cited include those by Khemani *et al.*, 1985, 1989 and 1994, working in areas of various factors that affect the precipitation chemistry as well as its spread over the India subcontinent. Work done by Satsangi *et al.*,

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1998; Tiwari *et al.*, 2004, included analysis of rain water specifically over rural and semi-arid locations in India and thus identifying factors that are exclusive to rural India influencing precipitation chemistry. Saxena *et al.*, 1996, Srinivas *et al.*, 1999; Singh *et al.*, 2007 carried out similar investigations in urban cities of India, namely Agra, Dhanbad, Hyderabad-Secundrabad respectively. Kulshrestha *et al.*, 1995 and 1996 worked along similar areas mostly in the urban New Delhi locations and demonstrated high influence of anthropogenic factors on the precipitation chemistry with lowering of pH values to uncomfortable ranges. Kulshrestha *et al.*, 2003 also carried out similar studies in cities of south-central India and identified similar factors that lead to acid rainfall in these locations. Mahadevan *et al.*, 1984 and 1989 carried out chemical analysis of precipitation in both industrial-urban areas as well as in rural-arid areas of India. Naik *et al.*, 1985, 1988 and 2002 analysed precipitation chemistry at representation and diverse locations including Hill station, Rural India and Industrial-urban area. The studies identified Mumbai-industrial region as having significant acid precipitation and low pH range in rainfall samples. Mukhopadhyay *et al.*, 1992; Parashar *et al.*, 2001 analysed rainfall samples across the Indian subcontinent and identified hotspots of acid rainfall. The associated areas were significantly of Industrial-urban locations. Other similar works carried out by in the Indian subcontinent include those by Rao *et al.*, 1995; Kumar *et al.*, 2002; Pillai *et al.*, 2001. Their study area included sensitive silent valley forested areas, sub-urban arid regions and urban-industrial town of Pune.

These studies revealed that the pH values were higher (pH>7.0) in the north and north-west parts of India while they were slightly lower (6.0 d" pH d"7.0) in the northeastern and southern parts of India. Work done by Balachandran *et al.* (2001) in Delhi during the monsoon period, reported pH values higher than 5.6. Work done by Granat *et al.* (2001) in the north east India showed that weighted mean concentrations of H⁺ and HCO⁻ were almost equal. Shrestha *et al.*, 2002 analysed the precipitation chemistry including aerosol characteristics over the isolated areas of Himalayan mountain range at Nepal. Comparable research investigations have also be carried out else where by Clarke *et al.*, 1987, Masamichi *et al.*, 2005; Mphepya *et al.*, 2004, Primerano *et al.*, 1998; Williams *et al.*, 1998 investigated the industrial dust and particulate factors that influence the characteristics of urban areas.

Asansol geographically located between latitude: 23°40'48" to 23°40'68" North and longitude: 86°58'48" to 86°40'98" East is situated in Burdwan district of West Bengal state. It is urban agglomerate with an area of 127 Km² and a population density of about 3737 individuals per Km². The city falls in the eastern, industrial belt of India. Industries present in and around the city include iron and steel, cement, distillery, silicate, soap and detergent, ceramics, engineering, paint, food processing, flour mills, etc. The city forms a part of the Raniganj-Coal field area. The industrial and mining operations contribute considerably to the atmospheric pollution along with vehicular emission and domestic burnings. The National Highway and The Grand Trunk Road passing through the city, transport massive traffic and consequently account for heavy discharge to the atmosphere. Emission from the vehicular movement and the Iron and Steel works are the chief polluting sources for this area (Banerjee *et al.*, 2005).

The climatic pattern is of typical tropical monsoon type with the year subdivided into three major seasons, namely summer, south-west monsoon and winter. During summers the mean max. temperature is 38.2 °C where as the mean min. temperature is 24.3 °C. The monsoon season starts during end of June with temperature ranging from 29.1 °C to 34.4 °C. By October, the mean temperature falls gradually marking the onset of the winter season. The maximum humidity, observed during the monsoon season, has mean value of 85%. The minimum humidity in Pre-monsoon (April-June) is 27 %. Annual rainfall obtained from the nearest meteorological office of IMD (Indian Meteorological Department) over a period of 10 years indicates average annual rainfall to be about 1500 mm. Nearly 80 % of the rainfall is received during monsoon season. The monsoon starts in end of June and continues till September.

The maximum amount of rainfall is received in July and the maximum rainy days occur in July. Wind is normally light to moderate in the area in the pre-monsoon season. During early monsoon wind speed is very high. The predominant wind direction during the rainy season (monsoon) is South West followed by West. The major objectives of the study was to appraise the chemical composition of rain water during the monsoon period and observe if acid rain fall events, if any, are occurring due to anthropogenic or natural processes.

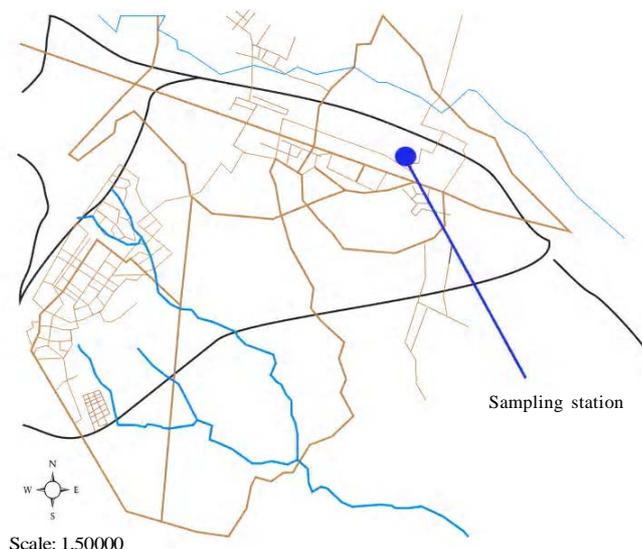


Fig. 1: Map showing study area and sampling station

MATERIALS AND METHODS

Samples of rainwater were collected on the open roof (about 10 m from the ground) during the monsoon season, from 2nd June 2006 to 29 September 2006 on an event basis. The location of study area is shown in Fig. 1. The sampling station was located in B.B.College, Asansol in the Burdwan District, West Bengal State of India. The site is about 200 meters from the busy and Grand Truck Road is located in a residential zone. Industries and mining activities are located at distances of 1.5 - 2 km away, which contributes heavily to the atmospheric load of pollutants in this city. Collected samples were analysed in the Department of Environment and Water Management, B.B.College, Asansol. Sampling was conducted as per guidelines of Tiwari *et al.* (2004) and analysis of rain water was as per APHA (APHA, 1998). Rain gauge, previously washed with deionized water, was used to collect the sample on 24 h. basis. Rain gauge and collecting system were washed during morning and evening throughout the study period to avoid dry deposition, although contamination due to dust fall had to be taken into account. All the samples were filtered through Whatman-41 filter paper before analysis. Samples were removed at 10:30 h. for night time rainfall events and 17:00 hours for day time rainfall. The concentrations of cations, namely, Ca^{2+} and Mg^{2+} were estimated by Titrimetric method; Na^+ and K^+ by Flame photometric method and NH_4^+ by Spectrophotometric method.

Chloride (Cl^-) was estimated using Titrimetric method; SO_4^{2-} and NO_3^- by Spectrophotometric method. pH values were determined with digital pH meter. All instruments/systems were properly standardized and calibrated ahead of every estimation. HCO_3^- was calculated from alkalinity values of rain water samples. All reagents, standard solutions were prepared from AR grade chemicals and de-ionised distilled water. The meteorological data and previous climatic norms of the study region were obtained from IMD, Pune.

RESULTS AND DISCUSSION

The mean ionic composition and pH values, along with the minimum and maximum value of the rain water samples, collected and analyzed during the June 2006 – September 2006 South-West Monsoon period at Asansol are given in Table 1. Results based on similar studies in India are given in Table 4 for comparison. Fig. 2 shows the frequency distribution of pH values of rain water samples during the study period. The relative percentage contribution of different ions to the total ionic content is Fig. 3. The scatter diagrams showing association between $(\text{NO}_3^- + \text{Cl}^-)$ vs SO_4^{2-} and $(\text{NH}_4^+ + \text{Ca}^{2+})$ vs $(\text{NO}_3^- + \text{SO}_4^{2-})$ is shown in Fig. 4. The correlation matrix for different ions in rain water samples is given in Table 2 and ratios of selected species with sodium is given in Table 3. The pH of natural precipitation is 5.65, a value created by equilibration of atmosphere CO_2 with precipitation (Naik *et al.*, 2002).

In the present study the pH varied between 5.02 and 6.93 (mean 6.08 ± 0.45), demonstrating acidic to alkaline range of the rainwater samples. The pH profile showed that out of the total rainfall event 17 % were acidic (<5.65), whereas 83 % accounted for alkaline rainwater. The pH frequency distribution for all samples were estimated by assigning them into nine categories (Naik *et al.*, 2002) ranging between 4.0 to 8.0 with 0.5 increment. The pH data was plotted for the different classes and shown as Fig. 2. Samples with low pH i.e. < 5.0 were not observed, indicating absence of very acidic rainwater. Similarly samples with pH values higher than 7.0 were also not recorded. The pH ranged in the mid values with 10 % samples in the pH range of 5.5; 25 % in the 6.0 and 7.0 and highest 40% in the 6.5 group. This is close to the Indian average rain water pH of 6.5. The results demonstrated that strong influence of strong acids was lacking in the rain water samples, where as alkaline components over the area played vital part in keeping the precipitation in the neutral to alkaline category in most cases. As shown in Table 3, the pH values had significant positive correlation with cationic components like Na^+ , Mg^{2+} and NH_4^+ . The chief acidic components were Cl^- and NO_3^- , while the presence of SO_4^{2-} was less (Fig. 4b). From Table 1 and Fig. 3 it was observed that the dominant ion species in

the rainwater sample was Calcium (28 %), followed by Carbonate (17 %) and Chloride (16 %). The ionic abundance in the rain water was seen to follow the trend: $\text{Ca}^{2+} > \text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{Mg}^{2+} > \text{NH}_4^+ > \text{Na}^+ > \text{NO}_3^- > \text{K}^+$. The Alkaline components (Cation), namely, Ca^{2+} , Mg^{2+} , Na^+ , K^+ and NH_4^+ contributes about 51.2% of the total ion strength, where as acidic components (Anion) comprising of SO_4^{2-} , Cl^- , NO_3^- , HCO_3^- was 48.7%.

The precipitation chemistry is dependent on its acidic and alkaline ions composition and the acidic effect neutralization capacity on the alkaline species like Ca^{2+} , Mg^{2+} , Na^+ , K^+ and NH_4^+ (Singh *et al.*, 2007). The anionic and cationic composition of the rain water is given in Fig. 4. Among the anions the dominant was HCO_3^- , contributing 36 % to this group and 17% to the total ions composition. This was followed by Cl^- (33%), SO_4^{2-} (23%) and NO_3^- (8 %). In the cation category Ca^{2+} was dominant, contributing 53 % in the group and 28 % to total ions chemistry. This was followed by Mg^{2+} (18 %), NH_4^+ (17 %), Na^+ (10 %) and K^+ (2 %). These components are generally derived from particulates washed down by the precipitation mainly in the monsoon period. After the removal of such species, the rain water turns more acidic during the late monsoon phase with the acidic components dominating.

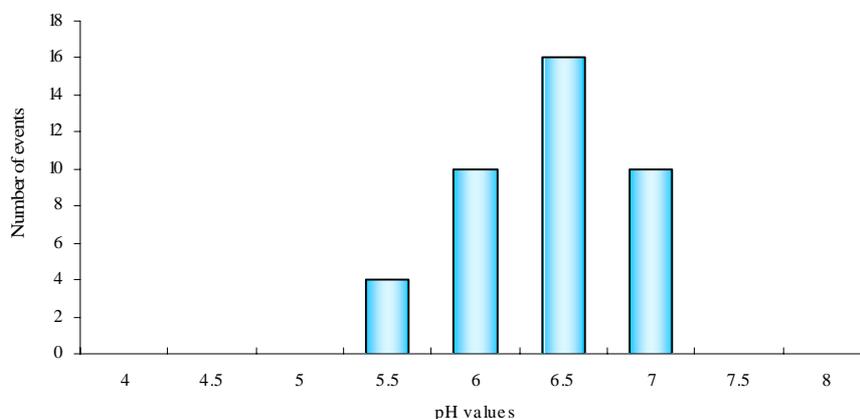


Fig. 2: Frequency distribution of pH values of rain water samples over Asansol during 2006 monsoon

Table 1: Statistical analysis of chemical composition of rainwater over Asansol during 2006 monsoon

	pH	K	Na	Ca	Mg	Cl	SO ₄	NO ₃	HCO ₃	NH ₄
S.D.	0.45	1.71	8.21	25.26	5.11	25.04	22.54	3.71	10.45	6.61
Min.	5.02	1.14	7.36	6.58	23.49	11.16	9.04	9.00	37.37	21.25
Mean	6.08	4.18	20.09	106.86	36.72	63.07	44.22	16.12	68.38	33.48
Max.	6.93	8.81	33.84	141.68	44.44	88.11	84.40	25.07	84.61	46.02

All units in $\mu\text{eq/L}$, except pH

The higher concentration of Ca^{2+} , an important acid neutralizing species, could be contributed in the form of Calcite (CaCO_3 , Dolomite ($\text{CaCO}_3, \text{MgCO}_3$), etc, in the soil blown up by wind. Other major sources of Ca, i.e. Cement and Iron and Steel industries are present within and around the study area in good number. High concentration of NH_4^+ may be attributed to nearby agricultural and local 'Khataals' (diary) inside the city, where bacterial decomposition of urine and 'cow dung' occur openly. The presence of higher concentration of these species in the rainwater neutralizes the acidic components and prevents acid deposition in the area. This was also proved by the observed pH value range of 5.02 – 6.93 (mean 6.08 ± 0.45), with majority of the samples having pH near the 6.5 mark. Among the anions HCO_3^- was highest, followed by Cl^- (33 %).

The ration of $(\text{NO}_3+\text{Cl})/(\text{SO}_4)$ and $(\text{NH}_4+\text{Ca})/(\text{NO}_3+\text{SO}_4)$ was estimated, since these ions determine the acid producing and acid neutralization capacity of the precipitation to a huge extent. The balance among these species dictates the acidic or basic nature of the rainwater. The scatter plots among these species are given in Fig. 4. The ratio of $(\text{NH}_4+\text{Ca}) : (\text{NO}_3+\text{SO}_4)$ varied between 0.88 to 9.13 (mean 2.76 ± 1.52) and the scatter plot in Fig. 4a shows that all points except one fell above the equiline, demonstrating that both NH_4 and

Ca^{2+} appreciably neutralized the anions in the analyzed rain water samples. The ration of $(\text{NO}_3+\text{Cl}) : (\text{SO}_4)$, which indicates whether the acidity in rain water is contributed by HNO_3/HCl or H_2SO_4 ranges from 0.64 to 8.08 with mean of 2.28 ± 1.50 (Singh *et al.*, 2007). The bivariate plot in Fig. 4b, representing this association, showed that nearly all points are above the equiline, indicating that the acidity in the analyzed rain water samples is significantly influenced by HNO_3 and HCl in the area. The total contribution of NO_3+Cl ions is 41 % and supports the ratio results. The high Cl^- in the rain water samples, ranging from 6.58 to 141.68 $\mu\text{eq/L}$ ($106.86 \pm 25.26 \mu\text{eq/L}$) suggested significant contribution from seawater and also to some extent on industrial activities like chemical, silicate and refractory plants present in and around the study area. The NO_3^- contribution was typically from road vehicles inside the city, which has evidenced an alarmingly high increase in vehicular population due to rapid development and urbanization in the area within a short period.

To collect information about association among the ionic species present in the rain water samples and to evaluate their likely sources a correlation matrix was prepared and the results are given in Table 3. Significant positive correlation was observed between pH and Cl^- , NH_4^+ , NO_3^- , Na^+ , SO_4^{2-} , HCO_3^- and Mg^{2+} , showing their

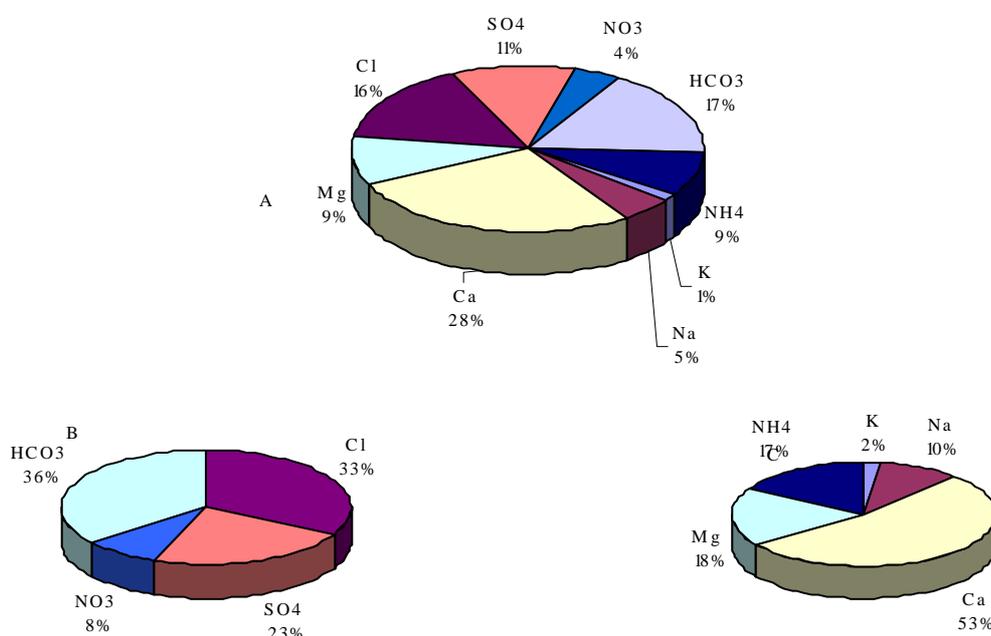


Fig. 3: Chemical composition of monsoon rainfall over Asansol city
 A) All ionic Composition B) Anionic contribution C) Cation contribution

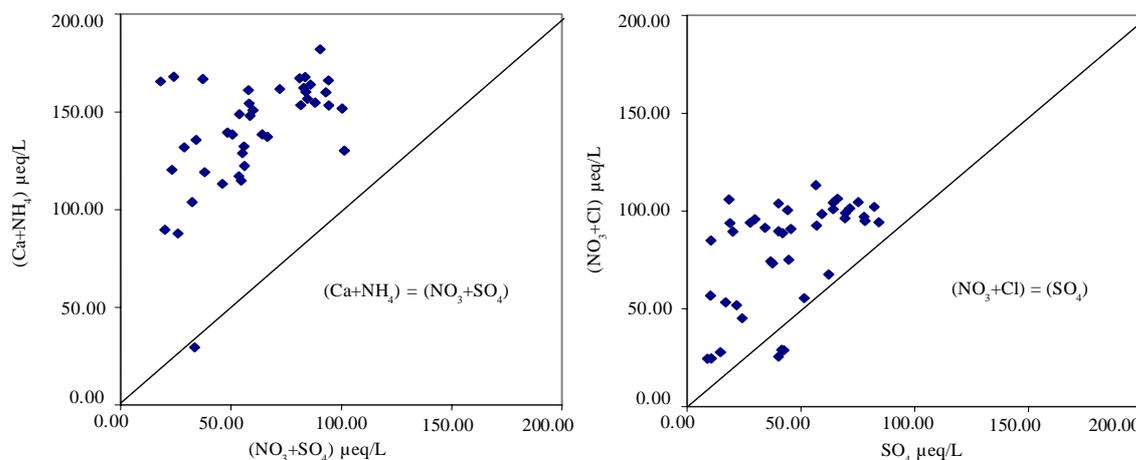


Fig. 4: Scatter plot diagram between A) $(\text{NH}_4 + \text{Ca})$ vs $(\text{NO}_3 + \text{SO}_4)$ and B) $(\text{NO}_3 + \text{Cl})$ vs SO_4 based on analysis of rain water sample over Asansol

influence on the acid-base nature on the rain water samples. Strong positive correlation was recorded amongst NH_4^+ with most of the other ions except K^+ and Ca^{2+} . The highest correlation ($r = 0.762$) was observed with NO_3^- , which suggested the significant presence of NH_4NO_3 in the area. The weak to moderate correlation among Ca^{2+} , Mg^{2+} , K^- and Na^+ suggests that probably they have originated from different sources. The moderately strong correlation of NO_3^- with SO_4^{2-} suggests that probably they are mostly due to man made contribution and this is supported by the fact that the region is an industrial area with significantly high atmospheric pollutant load (Banerjee *et al.*, 2005). Significant positive correlation is observed between Na and Cl^- (0.745), NO_3^- (0.700) and HCO_3^- (0.529). The influence of the south-west monsoon on the rain chemistry was assessed by estimating the ratios of Na with other ions and the results are shown in Table 3. The Na/Cl ratio of 3.140 is many times higher than the seawater, indicating strong influence of the south-west wind brought over the Bay of Bengal. Some portion of the contribution may be through man made sources like industrial activity. The ratios of other ions to Na showed similar trends, i.e. much higher than the seawater and it can be said the chemical makeup of rain water over Asansol was markedly influenced by the sea fraction.

The relative chemical analysis records of rain water studies done elsewhere in India with the present investigation in Asansol (Table 4) showed that pH value of rain water over Asansol was lower than the Indian average of 6.5 (Singh *et al.*, 2007). The mean pH was lower than other areas like Gopalpura, Hyderabad,

Colaba, Alibag and Ballia, but was higher than that in Kalyan and Dhanbad. The concentration of Na and Cl were significantly lower than those recorded in the coastal belts of western India, namely Kalyan, Colaba and Alibagh.

The comparison of the chemical analysis of the present study with that of Dhanbad, situated about 65 km away, showed that although Na^+ , NO_3^- and NH_4^+ values are more or less closely related but other parameters varies significantly. The pH value of 5.3 in Dhanbad area is much lower than the 6.08 value recorded over Asansol. This may be because although both the areas have similar features, with Dhanbad being more industrialized, the anthropogenic contribution of the acid producing ions are less over Asansol. The mean value of SO_4^{2-} and NO_3^- was comparable to most of the other areas. The values were less in comparison to those found in Hyderabad, Kolaba, Kalyan and Alibagh, since the congestion factor and heavy vehicular movement are extremely high in these cities.

The present investigation was conducted to evaluate the precipitation chemistry over Asansol during 2006 monsoon season. The rain water samples showed neutral to alkaline nature with mean pH value much higher than the reference value of 5.65. Frequency distribution of pH values established that 40% of rainfall events were in the 6.5 category, which is in accordance to the average Indian value. The major ion chemistry showed alkaline components influences were greater than the acidic types. The mean alkaline to acidic ratio was 1.05. The dominant cation was Ca , followed by Mg and NH_4^+ . Among the anions, HCO_3^-

Table 2: Correlation between the major ions present in rain water samples in Asansol

	pH	K	Na	Ca	Mg	Cl	SO ₄	NO ₃	HCO ₃	NH ₄
pH	1.000	0.237	0.658	0.228	0.550	0.699	0.564	0.672	0.554	0.680
K		1.000	0.151	0.226	0.378	0.150	0.274	0.358	0.392	0.473
Na			1.000	0.352	0.391	0.745	0.476	0.700	0.529	0.590
Ca				1.000	0.627	0.354	0.387	0.413	0.059	0.394
Mg					1.000	0.460	0.638	0.628	0.286	0.655
Cl						1.000	0.511	0.611	0.477	0.587
SO ₄							1.000	0.520	0.355	0.619
NO ₃								1.000	0.461	0.762
HCO ₃									1.000	0.653
NH ₄										1.000

Table 3: Ratios of various ions with respect to Sodium in rain water samples

	Cl:Na	Mg:Na	K:Na	Ca:Na	SO ₄ :Na
Rainwater (present study)	3.140	1.828	0.208	5.320	2.201
Seawater	1.160	0.227	0.022	0.044	0.125
Tiwari et al	1.320	0.810	0.560	6.410	5.950

Table 4: Average Ionic concentrations (µeq/L) of rainwater at different geographical locations in India

Location	Reference	Study period	pH	K	Na	Ca	Mg	Cl	SO ₄	NO ₃	HCO ₃	NH ₄
Gopalpura	Satsangi <i>et al.</i>	1996	6.1-7.4	3	19	134	78	31	15	43	-	43
Hyderabad	Srinivas <i>et al.</i>	1999	6.34	8	38	41	20	73	30	29	-	-
Kalyan	Naik <i>et al.</i>	1994-95	5.28	6	147	130	48	134	110	66	-	14
Colaba	Naik <i>et al.</i>	1994-95	6.38	6	179	155	48	171	52	34	-	12
Alibag	Naik <i>et al.</i>	1994-95	6.74	5	220	133	64	236	36	9	-	8
Dhandad	Singh <i>et al.</i>	2003 - 2005	5.30	13.1	17.8	70.0	15.6	27.4	62.8	10.2	-	35.3
Ballia	Tiwari <i>et al.</i>	2001	6.31	3	6	40	5	8	37	16	18	58
Asansol	(Present Study)	2006	6.08	4	20	107	37	63	44	16	68	33

was highest, followed by Cl. Among the major acidic components SO₄ was higher than NO₃ by a factor of about 3. In the total contribution Ca was the major ion, generated from dust derived fractions in the region. The dominant alkaline components effectively prevent acid precipitation in the area, by neutralizing the acids. The bivariate plots and ratios between (NO₃+Cl) and (SO₄) showed that the acidic factor is significantly influenced by HNO₃ and HCL, rather than H₂SO₄. Whereas similar studies between (NH₄+Ca) and (NO₃+SO₄), showed that NH₄ and Ca were dominating factors in prevention of acid rain over Asansol. The very high NH₄ concentration can be attributed to bacterial decaying of organic matter like cow dung, etc. It can be said the alkaline components, influenced mostly by the natural process are neutralizing the acidic components, generated from manmade sources, effectively.

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