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# Chemical Composition of Precipitation in the Coastal Environment of India

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**Abstract** The present study investigated the chemical composition of precipitation at Comba, Madgaon, South Goa during southwest monsoon. The rainwater samples were collected on event basis during June–September 2008 and were analyzed for pH, major anions F, Cl, $NO_3$, $SO_4$) and cations (Ca, Mg, Na, K, $NH_4$). The pH value varied from 5.36 to 6.91 (6.25 ± 0.28) indicating alkaline nature of rainwater and dominance of Cl and Na in precipitation. The Neutralization factors (NF) was found to be NFCa = 1.22, NFMg = 0.42, $NFNH_4$ = 0.37 and NFK = 0.14 indicating below cloud process in which crustal components are responsible for neutralization of anions.

**Keywords** Precipitation chemistry · Statistical correlation

Precipitation is wet removal pathway through which aerosol particles and gases are removed from the atmosphere and are the most effective scavenging of pollutants from atmosphere. Scavenging generally occurs via two processes (in-cloud and below cloud scavenging). The in-cloud scavenging process is predominant in Europe and North America due to high emissions of $SO_2$ and $NO_X$ and low soil-derived particulate matter in the atmosphere. The below-cloud scavenging process was more predominant in India and China, due to high concentration of soil-derived particulate matter. Nowadays, acid rain is recognized as one of the most serious threat to global environment. The pH < 5.6 (being reference level) is considered acidic which may cause damages to trees, plants and crops, acidification of lakes and rivers, respiratory disorders in human beings, poisoning in wildlife and damage of the most treasured monuments (Parashar et al. 2001; Lin et al. 2009).

Goa State lies in western coast of India. Comba suburbs is about 8–10 km from Arabian sea, of South Goa, India The climatic condition is typically humid due to its proximity to the sea. The weather is mostly foggy during dawn and at dusk during the rainy season. Goa receives rainfall from South-West monsoon between the months of June to September. The average rainfall varies from 3,000 to 3,500 mm.

To investigate the precipitation chemistry in coastal environment, sampling was carried out at Comba during monsoon season with an aim to identify and characterize ionic species, factors responsible for controlling the acidity and relative contributions of different sources of chemical species in precipitation to better understand the major processes and sources that control precipitation chemistry.

## Materials and Methods

Precipitation samples were collected at Comba (15°16′49″N, 73°57′37″) on event basis during monsoon period viz. June to September (2008) using one litre polyethylene bottle kept under funnels (diameter 14 cm). The glassware was thoroughly washed in deionized water to prevent contamination. They were placed at a height of 1-m to counter the contamination by droplet splashes. These polyethylene bottles and funnels were deployed for sampling immediately after rain began and retrieved soon after the rain stopped. A total of 51 samples were collected and had sufficient volume for

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chemical analysis, which represented most of the rain events during monsoon season.

Immediately after collection, the pH of samples was measured. Samples were analyzed for cations and anions by ion chromatography (ICS 3000). The limits of detection for cations and anions were 1 ppb in Dionex ion chromatograph. The analytical precision was maintained through running of known standards (Salve et al. 2006, 2008). $HCO_3$ exists in precipitation through dissolution of atmospheric $CO_2$ and other particulate carbonate species. $HCO_3$ concentration would be significant at the alkaline pH values encountered in most of the rain samples. As $HCO_3$ was seldom determined experimentally, it was calculated using a theoretical relation between the pH and $HCO_3$.

Ion balance is an important parameter for data quality assessment. Data quality of each precipitation events was checked by ionic balance. The data was rejected if it did not meet the quality criteria, which allowed for 15% deviation of ion balance ratio (sum of cations/sum of anions). Based on the principle of electro-neutrality, the quality of chemical analysis, to some extent, may be assessed by the ionic balance considering all the major ions in a single sample. The percent error (E%) was calculated by using equation reported elsewhere (Singh et al. 2007) and the % error was obtained as 2.02 which was found to be within ±5%. The statistical analysis, Oneway ANOVA was applied to estimate the measurement uncertainty in analysis. The analytical variance for the data should comprise not more than, say, 20% of the total variance. The statistics were performed within and between batches using SPSS software and *F* test results are presented in Table 1. The *F* test statistic was applied to test the null hypothesis. The calculated $F_{value}$ were observed in the range of 0.07–13.9 of which Na, Cl, and F is less than $F_{critical}$ of 3.08 for a stated level of confidence (typically 95%) means that the difference being tested is statistically significant at 95% confidence level. To reiterate the interpretation of ANOVA results, a calculated *F* test that is greater than $F_{critical}$ for a stated level of confidence (typically 95%) means that the difference being tested is statistically significant at that level for remaining ionic species. In this application, the measurement variance contributed 5.51% of the total variance, which is well within 20% target (Ramsey et al. 1992; Satapathy et al. 2009).

**Table 1** Significance of *F*-test in replicate analysis of ionic species in precipitation

| Ions | Variation | Sum of squares | *df* | Mean square | *F* |
|---|---|---|---|---|---|
| F | Between groups | $3.5 \times 10^{-2}$ | 1 | $3.5 \times 10^{-2}$ | 0.071 |
| | Within groups | 49.566 | 100 | 0.496 | |
| | Total | 49.601 | | | |
| Cl | Between groups | 47.4 | 1 | 47.437 | 0.131 |
| | Within groups | 36,081.1 | 100 | 360.841 | |
| | Total | 36,131.5 | | | |
| $SO_4$ | Between groups | 259.5 | 1 | 259.58 | 6.63 |
| | Within groups | 3,910.6 | 100 | 39.1 | |
| | Total | 4,170.2 | | | |
| $NO_3$ | Between groups | 15.042 | 1 | 15.042 | 7.234 |
| | Within groups | 205.392 | 100 | 2.054 | |
| | Total | 220.234 | | | |
| Na | Between groups | 4.23 | 1 | 4.237 | 0.021 |
| | Within groups | 20,317.7 | 100 | 203.178 | |
| | Total | 20,321.9 | | | |
| K | Between groups | 8.54 | 1 | 8.54 | 12.97 |
| | Within groups | 65.84 | 100 | 0.658 | |
| | Total | 74.38 | | | |
| Ca | Between groups | 329.72 | 1 | 329.72 | 4.87 |
| | Within groups | 6,758.9 | 100 | 67.59 | |
| | Total | 7,088.67 | | | |
| Mg | Between groups | 98.00 | 1 | 98.00 | 13.94 |
| | Within groups | 702.71 | 100 | 7.027 | |
| | Total | 800.71 | | | |
| $NH_4$ | Between groups | 14.95 | 1 | 14.95 | 3.80 |
| | Within groups | 393.29 | 100 | 3.933 | |
| | Total | 408.24 | | | |

## Results and Discussion

The average and standard deviation of ionic composition was found to be 321.2 ± 38 μeq/L. The statistical summary of volume weighted mean (vwm) concentration of ionic species, together with pH in precipitation is presented in Table 2. The ionic species in precipitation showed the general trend $Cl > Na > Ca > SO_4 > Mg > NH_4 > NO_3 > HCO_3 > F > H$. Due to close proximity to sea, Cl seems to be dominating species followed by Na in the coastal environment. The high concentrations of sea salt (C1 and Na) may be due to the monsoon currents which carry these particles from the sea. The excess Cl may be due to the contribution of gaseous chloride from manifestation of Cl displacement from sea-salts (NaCl) by anthropogenic acids. This could be possible frequently during more precipitation events. Under such meteorological conditions, contribution of gaseous chloride could have dominant influence on the precipitation composition (Rastogi and Sarin 2005). The total anions and cations contribute 48.99% and 51.01%, respectively, indicating that all the ionic species are measured. The skewness and kurtosis statistics provides information about the central tendency and variability of major ions. The skewness analysis indicates the distribution shape and variation of major ions based on the mean and the median values. If the

**Table 2** Summary of ionic species in precipitation

| Major ions | Min | Max | Average | SD | Skewness | Kurtosis | % |
|---|---|---|---|---|---|---|---|
| N = 51 | | | | | | | |
| pH | 5.6 | 6.91 | 6.25 | 0.28 | 3.10 | −1.33 | |
| F | 1.50 | 4.82 | 2.18 | 0.69 | 4.75 | 2.07 | 0.68 |
| Cl | 66.24 | 152.14 | 109.80 | 18.81 | −0.65 | 0.01 | 34.10 |
| $SO_4$ | 16.58 | 33.89 | 23.35 | 4.02 | −0.24 | 0.43 | 7.25 |
| $NO_3$ | 10.04 | 14.57 | 12.15 | 1.21 | −0.84 | 0.16 | 3.77 |
| Na | 60.14 | 122.40 | 86.81 | 12.54 | 0.38 | 0.53 | 26.96 |
| Ca | 28.53 | 60.28 | 43.47 | 7.92 | −0.61 | 0.26 | 13.50 |
| K | 3.13 | 5.87 | 4.82 | 0.67 | −0.14 | −0.64 | 1.50 |
| Mg | 8.45 | 20.45 | 15.18 | 2.54 | 0.19 | 0.08 | 4.72 |
| $NH_4$ | 9.42 | 18.42 | 13.21 | 2.17 | −0.56 | 0.49 | 4.10 |
| H | 0.12 | 4.36 | 0.73 | 0.81 | 11.72 | 3.40 | 0.23 |
| $HCO_3$ | 1.17 | 19.62 | 10.24 | 4.58 | −0.28 | 0.10 | 3.18 |

All values of major ions in μeq/L except pH

mean is less than the median, the distribution would be skewed to the left and few points with low values would disproportionately affect the mean; and vice versa. The kurtosis parameter characterizes the relative peak-holding or flatness of a distribution compared with the normal distribution. Positive kurtosis implies a relatively peaky distribution; however, negative kurtosis corresponds to a relatively flat curve (Mouli et al. 2005). It was noticed from the table that the skewness is positive for pH, F, Na, Mg, and H, indicating that the distribution has a long tail extending to the right and negative for Cl, $SO_4$, K $NH_4$, $NO_3$, Ca, and $HCO_3$; while the kurtosis values were negative values for K and pH whereas positive for remaining ionic species.

The pH was found to be in the range of 5.36–6.91 (6.25 ± 0.28). In this study, out of 51 rain events, in 94% cases the pH was higher than 5.6 was due to high loading of particulate matter in the atmosphere, which is mostly found in Indian climatic conditions (Charlson and Rodhe 1982). The difference in free acidity between the lowest sample (pH 5.36, [H] 4.36 μeq/L) and natural precipitation in equilibrium with atmospheric $CO_2$ (pH 5.6, [H] 2.51 μeq/L) is only 1.84 μeq/L. This suggested that most of the $SO_4$ and $NO_3$ are present as neutral salts ($(NH_4)_2SO_4$, $NH_4NO_3$) and not as dissociated acids.

In order to estimate the sea salt and non-sea salt contributions to precipitation, equivalent ratios were calculated using Na as the reference element and assuming that all Na is of marine origin. The enrichment factor of Cl, Mg, Ca, Mg, and $SO_4$ was higher than one indicating significance influence of local and coastal sources (Table 3). The observed precipitation ratio of Cl/Na (1.19) is closer to that of seawater ratio (1.16) indicates resuspension of

**Table 3** Enrichment factor (EF), sea salt (ss) and non sea salt (nss) fractions of ionic species in precipitation

| Major ions | Cl/Na | K/Na | Mg/Na | Ca/Na | $SO_4$/Na |
|---|---|---|---|---|---|
| Sea water | 1.16 | 0.037 | 0.038 | 0.12 | 0.25 |
| Rainwater | 1.19 | 0.056 | 0.175 | 0.501 | 0.269 |
| SS (%) | 91.36 | 33.36 | 21.73 | 23.96 | 92.94 |
| NSS (%) | 8.64 | 66.64 | 78.27 | 76.04 | 7.06 |
| EF | 1.027 | 1.501 | 4.602 | 4.174 | 1.076 |

previously deposited sea salt onto the soil. The non sea salt (nss) contributions of ionic constituent, viz. nss-Ca was observed as 76% suggesting their crustal origin whereas nss-Mg and nss-K showed their contribution as 78.2% and 66.6% indicating influence of soil sources. The nss-$SO_4$ contribution was very less 7.06% indicating remaining major contribution was from sea.

In order to determine the dominance of species in atmosphere, a linear regression model was applied to ionic concentrations of potential acidic species ($SO_4$ and $NO_3$) as dependent variables and the major cations ($NH_4$) as independent variables due to the highest mutual correlation ($r = 0.93$) (Table 4). Study of the table indicates that $NH_4$ was associated with $SO_4$ followed by $NO_3$ in the reaction of $HNO_3$ and $H_2SO_4$ with atmospheric $NH_3$ originating from fertilizer industries located nearer to sampling site. A linear regression of [$NH_4$ vs. $SO_4 + NO_3$], [Ca + Mg + $NH_4$ vs. $NO_3 + SO_4 + Cl$], [Ca + Mg + $NH_4$ + K vs. $NO_3 + SO_4 + Cl$], [Ca + $NH_4$ vs. $SO_4 + NO_3 + Cl$] and [Ca vs. $SO_4 + NO_3 + Cl$] was 0.93, 0.90, 0.90, 0.89, and 0.75 indicating that acidity was neutralized by ammonium, calcium and magnesium. A regression of [K + $NH_4$ vs. $NO_3 + SO_4$] was 0.88 indicating possible species that exist in the atmosphere are $(NH_4)_2SO_4$, $NH_4NO_3$, $K_2NO_3$, and $K_2SO_4$. The ratio of $H^+$ / ($NO_3 + SO_4$) is also an indicator to determine the extend of neutralization process in precipitation. The ratio close to zero indicates extensive neutralization whereas values close to one indicate lack of

**Table 4** Linear regression of ionic species in precipitation

| | $NO_3$ | $SO_4$ | $NO_3 + SO_4$ | $(NO_3 + SO_4 + Cl)$ |
|---|---|---|---|---|
| Ca | −0.28 | −0.26 | −0.27 | **0.75** |
| $NH_4$ | **0.90** | **0.93** | **0.93** | −0.06 |
| Ca + $NH_4$ | −0.04 | −0.01 | −0.01 | **0.89** |
| Ca + Mg | −0.20 | −0.18 | −0.19 | **0.88** |
| Ca + Mg + K | −0.20 | −0.17 | −0.18 | **0.88** |
| Ca + Mg + K + $NH_4$ | 0.04 | 0.07 | 0.06 | **0.90** |
| K + $NH_4$ | **0.85** | **0.89** | **0.88** | −0.04 |
| Ca + Mg + $NH_4$ | 0.04 | 0.06 | 0.06 | **0.90** |

$r$ = >0.70 is considered as significant

neutralization in precipitation. The ratio was 0.0205 which is close to zero indicate 99.99% of acidity was neutralized in precipitation (Seinfield 1986).

Neutralization factors (NF) were used to evaluate the neutralization of ionic species by crustal components and ammonia. The NF species was found to be $NF_{Ca} = 1.22$, $NF_{Mg} = 0.42$, $NF_{NH_4} = 0.37$, and $NF_K = 0.14$, respectively, revealing below cloud process in which crustal components are responsible for neutralization of anions (Table 5). The % contribution of Ca, Mg, $NH_4$, and K are 56.6, 19.7, 17.23, and 6.28, respectively. Similar study reported at Bhubaneswar, Ahmedabad and Goa (Das et al. 2005; Rastogi and Sarin 2005; Parashar et al. 2001) showed dominance of calcium in precipitation.

A correlation matrix between ionic species is presented in Table 6. A significant correlation was seen between the acidic ions $SO_4$ and $NO_3$ ($r = 0.95$) indicating their origin from similar sources, because of the similarity in their behavior in precipitation and the co-emissions of their precursors $SO_2$ and $NO_X$ (Kumar et al. 2002). Highly significant correlation for $NH_4$ and $SO_4$ ($r = 0.92$), followed by $NH_4$ and $NO_3$ ($r = 0.90$) implies that they have occurred in the form of $(NH_4)_2SO_4$ and $NH_4NO_3$ (Seinfield 1986). The relationship between Na and Cl ($r = 0.91$) indicated predominantly associated sea salt ions. The principal component analysis (PCA) with Varimax rotation was applied to the set of original data (concentration of major ions) in order to determine the influence of anthropogenic and natural sources in precipitation (Table 7). Three factors were considered with Eigens values higher than unity. Totally, 72.16% of the variance explained by the three factors was sufficient to describe most of the variances. The PCA1 explained 25.24% of variance with high loading of $NH_4$, $SO_4$, and $NO_3$ which is attributed to anthropogenic sources (combustion, traffic for $NO_X$ and $SO_2$, fertilizers factories and agriculture activity for $NH_3$). Sulfates could also have a marine origin, on the seaside (Jeanton et al. 2009). The $NH_4$ ion could be linked to co-concurrence of $NH_4$ with $NO_3$ which may be caused by dissolution of aerosol/secondary pollutants in precipitation containing $NH_4NO_3$ (Khare et al. 2004). The PCA2 explained 24.36% of variance with high loading of Na and

**Table 5** Neutralization factor for ionic species in precipitation

| Locations | Ca | Mg | $NH_4$ | K | References |
|---|---|---|---|---|---|
| Comba | 1.22 | 0.42 | 0.37 | 0.14 | Present study |
| Ahmedabad | 1.77 | 0.11 | 0.73 | – | Rastogi and Sarin (2005) |
| Bhubaneswar | 0.70 | 0.17 | 0.64 | 0.06 | Das et al. (2005) |
| Goa | 1.26 | 0.12 | 0.27 | – | Parashar et al. (2001) |

**Table 7** Multivariate analysis of ionic species in precipitation

| Major ions | PCA1 | PCA2 | PCA3 |
|---|---|---|---|
| Ca | **−0.254** | 0.799 | 0.212 |
| Cl | −0.228 | **0.856** | 0.127 |
| F | −0.430 | −0.491 | 0.257 |
| $HCO_3$ | 0.07 | 0.173 | **0.860** |
| K | 0.371 | 0.513 | −0.357 |
| H | 0.06 | 0.03 | **−0.89** |
| Mg | **0.392** | 0.197 | 0.126 |
| Na | −0.136 | **0.907** | 0.125 |
| $NH_4$ | **0.883** | −0.246 | 0.028 |
| $NO_3$ | **0.891** | −0.229 | 0.014 |
| $SO_4$ | **0.913** | −0.203 | −0.03 |
| % of variance | 25.24 | 24.36 | 22.55 |
| Eigens value | 3.81 | 2.51 | 2.33 |

$r$ = >0.80 is considered as significant

**Table 6** Correlation matrix of ionic species in precipitation

| | Ca | Cl | F | H | $HCO_3$ | K | Mg | Na | $NH_4$ | $NO_3$ | $SO_4$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Ca | 1.0 | | | | | | | | | | |
| Cl | **0.91** | 1.0 | | | | | | | | | |
| F | 0.01 | −0.01 | 1.0 | | | | | | | | |
| H | −0.15 | −0.05 | −0.06 | 1.0 | | | | | | | |
| $HCO_3$ | 0.20 | 0.16 | 0.01 | **−0.67** | 1.0 | | | | | | |
| K | −0.01 | 0.04 | **−0.94** | 0.12 | −0.11 | 1.0 | | | | | |
| Mg | 0.02 | 0.13 | −0.17 | 0.01 | 0.17 | 0.10 | 1.0 | | | | |
| Na | **0.83** | **0.91** | −0.17 | −0.09 | 0.23 | 0.20 | 0.01 | 1.0 | | | |
| $NH_4$ | −0.30 | −0.31 | −0.09 | 0.11 | −0.05 | 0.06 | 0.14 | −0.25 | 1.0 | | |
| $NO_3$ | −0.32 | −0.27 | −0.05 | 0.13 | −0.04 | 0.01 | 0.22 | −0.22 | **0.90** | 1.0 | |
| $SO_4$ | −0.28 | −0.28 | −0.11 | −0.15 | −0.02 | 0.06 | 0.21 | −0.23 | **0.92** | **0.95** | 1.0 |

$r$ = >0.80 is considered as significant

**Table 8** Comparison of ionic species in different parts of India (μeq/L)

| Site | Cl | $NO_3$ | $SO_4$ | Na | Ca | K | Mg | $NH_4$ | References |
|---|---|---|---|---|---|---|---|---|---|
| Cl dominance sites | | | | | | | | | |
| Comba | 109.8 | 12.1 | 23.3 | 86.8 | 43.4 | 4.8 | 15.1 | 13.2 | Present study |
| Alibagh | 236 | 9 | 36 | 220 | 46 | 5 | 64 | 8 | Naik et al. (2002) |
| Goa | 113.4 | 5.5 | 27.4 | 97.2 | 41.5 | 2.5 | 24.5 | 5.5 | Parashar et al. (2001) |
| Bombay | 138 | – | 10 | 115 | 36 | 3.6 | 24 | – | Seqeuira (1976) |
| Na dominance sites | | | | | | | | | |
| Colaba | 171 | 34 | 52 | 179 | 133 | 6 | 59 | 12 | Naik et al. (2002) |
| Silent Valley | 43.0 | 21.0 | 20.0 | 46.0 | 43.0 | 4.0 | 14.0 | 3.0 | Rao et al. (1995) |
| Chembur | 164.5 | 29.5 | 70.4 | 168.2 | 89.5 | 6.9 | 36.5 | 41.1 | Khemani et al. (1994) |
| Bhubaneswar | 18 | 10 | 19.1 | 15 | 20.2 | 1.8 | 5.2 | 18.7 | Das et al. (2005) |

Cl suggesting influence of long range transport of sea winds or sea salt on the observed concentrations and low K concentration could either be due to terrestrial potassium or anthropogenic aerosols (biomass burning, waste burning) and Ca is brought together in the region by marine wind from the west coast (Budhavant et al. 2009). The PCA3 explained 22.5% of variance with high loading of $HCO_3$ and negative loading of $H^+$ indicating only for acidity due to lower presence of ions $H^+$ in the samples of precipitation.

The dominance of ionic species in precipitation was geographically differs from locations and regions are presented in Table 8. In Alibagh [Cl > Na > Mg > Ca > $SO_4$ > $NO_3$ > $NH_4$ > K], Goa [Cl > Na > Ca > Mg > $SO_4$ > $NO_3$ > K], and Bombay [Cl > Na > Ca > Mg > $SO_4$ > $NO_3$ > K] showed dominance of Cl (Naik et al. 2002; Parashar et al. 2001; Seqeuira 1976) whereas at Colaba [Na > Cl > Ca > Mg > $SO_4$ > $NO_3$ > $NH_4$ > K], Silent Valley [Na > Ca > Cl > $NO_3$ > $SO_4$ > Mg > K>$NH_4$], Chembur [Na > Cl > Ca > $NO_3$ > $NH_4$ > Mg > $SO_4$ > K], there is a dominance of Na as these sites are nearer to sea coast (Naik et al. 2002; Rao et al. 1995; Khemani et al. 1994).

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