

Principal component analysis of rainwater composition at BAPMoN stations in India

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संक्षेप—भारतीय बैपमन स्टेशनों में लंबी अवधि (1976-87) के दौरान एकत्रित किए गए वर्षा जल के नमूनों में पाए गए खनिज और गैसीय स्रोतों से प्राप्त आयनों के कालिक तथा स्थानिक अभिव्यक्तियों का मुख्य अवयव विश्लेषण (पीसीए) के उपयोग द्वारा मॉडेल के तौर पर परीक्षण करके भारतीय बैपमन स्टेशनों के आंकड़ों का विश्लेषण किया गया है। परिणामों से संकेत मिलता है कि वर्षा जल के पीएच को SO_4^{2-} , NO_3^- , Cl^- , Ca^{+2} और Na^{+1} आयनों की सांद्रता के संदर्भ में सामान्य तौर पर स्पष्ट किया जा सकता है। तथापि अन्य कालिक विभिन्न, परस्पर क्रिया की समग्र, प्रकृति को निर्धारित कर सकनी है। मुख्य अवयव विश्लेषणों द्वारा ये क्रिया विधि सुस्पष्ट हो गई है।

ABSTRACT. Data from Indian BAPMoN stations were analyzed using the Principal Component Analysis (PCA) by examining broadly the temporal and spatial distribution characteristics of the ions, from mineral and gaseous sources, observed in rainwater samples collected over the Indian BAPMoN stations over a long period (1976-87). The results show that the pH of rainwater can be generally explained in terms of the concentration of SO_4^{2-} , NO_3^- , Cl^- , Ca^{+2} and Na^{+1} ions. However, other mechanisms could determine the overall nature of the interactions. These mechanisms have become more clear by performing principal component analysis.

Key words — Principal component analysis, Spatial and temporal homogeneity, pH, EOF, BAPMoN.

1. Introduction

The chemical composition of rainwater gives an indication of the gaseous and particulate composition of a large depth of atmosphere that is traversed by raindrops falling from clouds in the atmosphere. The composition of rainwater is affected by both, cloud micro-physical processes and the sub-cloud layer pollutant composition. Thus rainwater samples from different shower events, exhibit variable compositions. But when monthly mixed samples of rainwater are chemically analyzed and the net depositions are accounted for over a substantial length of time, it is mainly the ambient atmospheric burden of pollution that gets reflected while the weather related factors only cause the variations about the mean values.

The India Meteorological Department operate ten station Background Air Pollution Monitoring Network (BAPMoN) (Table 4), that forms a part of Global Air Pollution Observational Network wherein precipitation samples are specially collected under a 'wet only' sampling programme (*i.e.*, avoidance of any dry deposition in the collector (Krishnanand 1984, Mukherjee *et al.* 1986). The monthly mixed samples are analyzed for determining the concentration of the following constituents H^{+1} , Na^{+1} , K^{+1} , Ca^{+2} , Mg^{+2} , NH_4^{+1} , SO_4^{2-} , NO_3^- , Cl^- . Among these ions the dominant species are Ca^{+2} , Na^{+1} , Cl^- , SO_4^{2-} , and NO_3^- (Table 4), which form

the main objective of our study. Hence the above mentioned five ions have been used to explain the pH as well as test the spatial and temporal homogeneity of the network.

Inter-relationship between the observed ions is a key to identification of different sources of these ions, and also the determination of hydrogen ions in rainwater, that controls acidity.

Earlier attempts at developing linear regression models of pH of rainwater with the precursors (SO_4^{2-} , NO_3^-) have not explained the observed variances of pH satisfactorily. The unexplained proportion arises due to the limited resolution in sampling which is unable to differentiate between SO_4^{2-} and NO_3^- coming from chemically contrasting sources (Vermeulen 1980, Fay *et al.* 1980) which could push the ionic balance either in favour of H^{+1} or away. Thus, the chemically identifiable ionic species do not form a set of independent factors by themselves. The search for linearly independent set of precursors for acidity of rainwater would naturally lead one to consider the application of the Principal Component Analysis (PCA) technique to the observed data. In the present paper, the 1976-87 data series for the precipitation components mentioned earlier has been analysed using PCA technique.

2. Method of analysis

The principal component analysis provides a simple means of transforming linear functions of p original variables, e.g., x_m , $m=1, \dots, p$ to another set of variables, e.g., X_i . The function X_i may be written as :

$$X_i = \sum_{m=1}^p a_{im} x_m, \quad i = 1, \dots, N, \quad m = 1, \dots, p$$

where, a_{im} values are constants.

As a_{im} varies, one gets different linear functions and one can calculate the variance of any such linear function. The first Principal Component (PC) is that linear function which has the maximum possible variance; the second PC is the linear function with maximum possible variance subject to being uncorrelated with the first PC and so on. Thus, it is easy to construct ' p ' principal components providing optimal m -dimensional representation of the data for each $m=1, 2, \dots, p$ for various different definitions of optimality. In particular at each stage the sum of the variance of the PCs is as large as possible. In other words, with PCA we get for each $m=1, 2, \dots, p$ the ' m ' linear functions of x_1, x_2, \dots, x_p which account for the maximum possible proportion of the original variations. Here a_{im} ($m=1$ to p) are vectors consisting of the weights of different variables in the i th PC. We compute eigen vectors of the ($p \times p$) co-variance (Covar) matrix between all x_m . The orthogonality condition on X_i implies that covar matrix of x_m has off diagonal terms as (zero while that of x_m has the diagonal and off diagonal terms also). The transformation from x - X therefore can be achieved by only diagonalising the covariance matrix.

Thus the first eigen vector is the set of coefficient a_{im} , $m=1, \dots, p$ appearing in the first PC. Similarly, subsequent eigen vectors consist of coefficients of X_2, X_3, \dots, X_p in each successive PC. The first eigen value is the variance of the first PC and so on. To define the principal components uniquely, the normalisation condition is imposed. Method of normalisation used in this paper is given by:

$$\sum_{i=1}^{10} a_{mi}^2 = \frac{1}{\lambda_i}$$

where, $\text{Var}(X_i) = 1$ for $m = 1, \dots, p$ being the corresponding eigen value.

As an example, we may consider subscripts ' i ' to define the time and ' m ' to define space domains. The primary advantage of the principal component solution is its ability to compress almost the total variability of the original data set into relatively a few temporally uncorrelated components. However, the spatial orthogonality of the eigen vectors is a strong and often undesirable constraint imposed on the principal component solution. While the first principal component and its eigen vectors are not influenced by this constraint, the remaining eigen vectors often bear predictable geometric relationships to the first eigen vector,

TABLE 1 (a)

Factors (eigen vectors) for chemical compositions of rainwater

Factor	SO ₄ ⁻²	NO ₃ ⁻¹	Cl ⁻¹	Ca ⁺²	Na ⁺¹	Percentage covariance explained
First	.54	.24	.41	.43	.55	51.88
Second	.21	.62	-.57	.37	-.33	30.61
Third	.08	.64	.26	-.73	.02	11.07
Fourth	-.81	.39	.18	.31	.25	5.41
Fifth	-.04	-.06	-.64	-.24	.72	1.03

In the present study the PCA has been performed with the following three objectives :

(i) Explanation of pH variations at each station

Here Ca⁺², Na⁺¹, SO₄⁻², NO₃⁻¹, and Cl⁻¹ concentrations form the five dimensional X_m . Each year represents one value of i . Hence there is non-zero variance of each x_m considering the yearly averages. In addition to this all X_{ms} are not independent since each ion may have multiple sources. Thus a covariance also exists between pairs like (x_1, x_2), (x_1, x_3) and so on. Now if the variable x_m is transformed on to a new set X_m with the constraint that only year-to-year variance of X_i exists, but no covariance between pairs like (X_1, X_2) and so on, then effectively we are left with five sets of linear combinations of Ca⁺², Na⁺¹ etc each of which are orthogonal. These five orthogonal vectors (or principal components) now may be used in a regression relationship to explain the dependable variable pH.

We therefore, find five eigen vectors each of whose coefficients are nothing but weightages given to each ion in each of the PCs. Thus, say for the first PC, each weightage multiplied by the annual mean concentration of the corresponding ion and summed for all ions forms the index for that years pH. Similarly five such indices can be formed for different PCs. Hence $\text{pH}_i = A_1 I_1 + A_2 I_2 + \dots + A_5 I_5$ becomes the new regression equation for pH which is obviously different from a relation like:

$$\text{pH} = A_1(\text{Ca}^{+2}) + A_2(\text{Na}^{+1}) + A_3(\text{SO}_4^{-2}) + A_4(\text{NO}_3^{-1}) + A_5(\text{Cl}^{-1})$$

where, A_1, A_2, \dots, A_5 are regression coefficients.

(ii) Spatial homogeneity of each ion

In this case PCs are worked out for each ion separately. The m values of say SO₄⁻² (or any ion for that matter) correspond to m different stations and the different values of ' i ' correspond to the annual concentrations for the i th year.

TABLE 1 (b)
Multiple correlation with pH of rainfall

Station	Observed pH	1st index *		1st & 2nd index†	
		Expected	Error	Expected	Error
Allahabad	7.2300	6.7827	0.4473	7.0736	0.1564
Jodhpur	7.5700	7.2690	0.3010	7.3691	0.2009
Kodaikanal	6.4800	6.4327	0.0473	6.3950	0.0850
Minicoy	6.5800	6.8406	-0.2606	6.3585	0.2215
Mohanbari	6.2800	6.4019	-0.1219	6.4615	-0.1815
Nagpur	6.4900	6.5756	-0.0856	6.5983	-0.2083
Port Blair	6.2900	6.7765	-0.4865	6.3143	-0.0243
Pune	6.8000	6.6824	0.1176	6.7864	0.0316
Srinagar	7.4100	6.8222	0.5878	7.0960	0.3140
Visakhapatnam	6.5900	7.1364	-0.5464	7.1673	-0.5773

*—Simple cc=0.50, †—Multiple cc=0.82

TABLE 2
Factors associated with spatial distribution of ionic concentration

Factor	Percentage covariance	Allahabad	Jodhpur	Kodai-kanal	Minicoy	Mohanbari	Nagpur	Port Blair	Pune	Srinagar	Visakhapatnam
Sulphate 1st	62.2	0.37	0.28	0.35	0.17	0.31	0.34	0.34	0.34	0.31	0.30
Nitrate 1st	58.2	0.35	0.31	0.21	0.29	0.28	0.38	0.35	0.33	0.32	0.32
Chloride 1st	39.6	0.40	0.46	0.41	-0.17	-0.13	0.30	-0.28	0.19	0.40	0.25
2nd	22.4	-0.12	-0.09	-0.06	-0.37	-0.43	0.42	-0.32	-0.55	-0.28	0.02
Calcium 1st	37.2	0.01	0.00	0.50	-0.06	-0.12	0.50	0.48	0.08	-0.08	0.48
2nd	27.4	-0.45	0.31	0.08	0.56	0.22	0.05	-0.12	0.11	0.51	0.20

TABLE 3
Temporal variations of ionic concentrations associated with first factor (eigen vector)

Ions	Year												Percentage of covariance explained
	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	
Sulphate	0.35	0.33	0.30	0.36	0.35	0.32	0.25	0.36	0.22	0.07	0.25	0.13	52.6
Nitrate	0.14	0.33	0.36	0.34	0.09	0.33	0.32	0.34	0.28	0.18	0.27	0.32	54.3
Chloride	—	—	—	0.36	0.30	0.31	0.33	0.37	0.37	0.31	0.35	0.29	75.7
Calcium	—	—	—	0.35	0.34	0.36	0.32	0.32	0.37	0.37	0.20	0.36	82.5

TABLE 4

Mean and standard deviations (1976-1987) of annual precipitation volume weighted concentrations (Unit : mg/lit)

Station	Elevation (m)	Location	Latitude (°N)	Longitude (°E)	SO ₄ ⁻²	NO ₃ ⁻¹	Cl ⁻¹	Ca ⁺²	Na ⁺¹
Allahabad	98	Continental alluvial	25°27'	81°44'	0.86 (0.72)	2.99 (2.63)	0.71 (0.38)	2.87 (1.35)	1.55 (0.62)
Jodhpur	217	Continental arid	26°18'	73°01'	1.85 (1.80)	1.84 (1.67)	1.97 (0.62)	6.22 (1.70)	3.13 (1.30)
Kodaikanal	2343	Tropical high altitude	10°14'	77°28'	0.57 (0.48)	0.52 (0.50)	0.45 (0.22)	0.55 (0.47)	0.52 (0.33)
Minicoy	2	Arabian Sea marine	08°18'	73°00'	0.55 (0.32)	0.61 (0.90)	3.21 (0.75)	1.19 (0.27)	2.53 (0.63)
Mohanbari	111	Humid alluvial	27°29'	95°01'	0.91 (0.69)	1.75 (1.19)	0.53 (0.51)	0.57 (0.19)	0.59 (0.40)
Nagpur	310	Continental	21°06'	79°03'	0.84 (0.72)	1.65 (1.14)	0.57 (0.23)	1.19 (0.40)	0.75 (0.40)
Port Blair	79	Bay of Bengal marine	11°40'	92°43'	0.84 (0.72)	1.01 (1.16)	2.53 (0.59)	0.62 (0.24)	2.33 (0.51)
Pune	659	Deccan traps semi-arid	18°32'	73°51'	1.21 (1.00)	1.71 (0.72)	0.99 (0.31)	1.29 (0.35)	0.87 (0.28)
Srinagar	1587	Extra-tropical elevated valley	34°05'	74°50'	1.11 (0.99)	2.33 (2.61)	1.07 (0.39)	4.11 (2.27)	1.40 (0.24)
Visakhapatnam	72	Coastal	17°41'	83°18'	1.83 (1.22)	3.32 (1.72)	2.61 (1.17)	1.39 (0.40)	2.70 (1.25)

N.B. — The standard deviations are given in brackets.

If the weightages appearing in each PC are similar in value then the SO₄⁻² data is spatially homogeneous for each PC. A different PC in this case leads to a different mechanism or a different source for SO₄⁻².

(iii) Temporal homogeneity

The m and i can be interchanged in the above analysis and in a similar way of interpretation, the temporal homogeneity can be studied.

3. Results

The ionic components of rainwater namely SO₄⁻², NO₃⁻¹, Cl⁻¹, Ca⁺², Na⁺¹ which account for more than 90% of total dissolved salts are subjected to PCA. Long term annual means for all the BAPMoN stations were worked out for this purpose. The five eigen vectors (PCs) which are identified as five independent factors, along with the variances explained by each of them, are given in Table 1 (a). The first two PCs account for 82.5% of total co-variations with the first one accounting for 51.9%. Considering the multiplicity of sources of

these ions normally encountered in rain chemistry, these factors may be expected to represent dominant source mechanisms rather than refer to the identity of individual sources. The five indices are formed for each station by multiplying the respective weights taken from each of these factors to mean values of the corresponding ionic concentrations and then adding the products. These indices, therefore, relate to the relative significance of each of these mechanisms for every station. Linear multiple correlations were subsequently worked out to explain the observed values of pH with one and more, indices taken at a time. It was noticed that the first and the second index combined accounts for 82.5% of total variance of pH. Simple linear correlation coefficient (cc) with the first index, and multiple cc with the first as well as second, are given in Table 1 (b). It may be noted that a group of stations (*viz.*, Mohanbari, Nagpur, Port Blair and Visakhapatnam) have persistently lower pH values observed as compared to those explained.

In the second PCA, the spatial distribution of SO₄⁻², NO₃⁻¹, Cl⁻¹, Ca⁺², ions are studied with the long term average values for the ten stations (Table 2). The variance

observed for SO_4^{-2} , NO_3^{-1} , Cl^{-1} and Ca^{+2} is given below :

(i) Sulphate

The first two factors account for 76.9% of total variance with the first one explaining 62.2%. As the contribution of the second factor is rather small it appears adequate to consider only the first one for meaningful interpretation.

(ii) Nitrate

For this ion the first factor explains 58.2% variance and the second 16.45%. Thereafter, the contributions gradually taper off.

(iii) Chloride

The first factor explains only 39.6% variance and the second one an additional 22.4%.

(iv) Calcium

The first factor explains 37.2% variance and second one an additional 27.4%.

It may be noticed that in the case of chloride and calcium ions, the first factor is not enough to account for a significant proportion of variance and hence consideration of the second factors becomes necessary. Stating differently, a more complex nature of interactions with rainwater is envisaged, in the case of chloride and calcium. The latter two ions come from solid-aerosol phases and hence intensity and spacing of rain events become important for their washout. On the other hand SO_4^{-2} and NO_3^{-1} ions are substantially contributed by different gases. The distribution of gases is far more homogeneous than aerosols. Relative strengths of emissions from the region, therefore, could control their distributions to a greater extent.

Another analysis was carried out to test the temporal homogeneity of the data for all the stations combined as a whole and look for trends if any. Table 3 gives the factors whose components are same in number, as the number of years of data. It may be noted that the homogeneity in time is somewhat better than that in space and unlike the earlier case, with the first factor explaining a relatively lower proportion of covariances, for SO_4^{-2} and NO_3^{-1} even though individually they are still substantial.

4. Discussion

4.1. The emphasis on rain chemistry stems from the interest in determining the acidity of rainwater. But the net H^+ concentrations are highly variable on account of uptake of a wide variety of natural as well as anthropogenic mineral salts and gases within the cloud and sub-cloud environments. The total cation concentration of rainwater being less in the extra-tropics (WMO, BAP-MoN) compared to what is encountered in India, the gases play an important role in producing H^+ ions on dissolution. There have been attempts in European region (Likens and Bormann 1974, Vermeulen 1980, Fay *et al.* 1980) and for Indian region (Varma 1980) to explain the acidity of rainwater as a function of gaseous oxides. Recent studies in the India Meteorological Department indicate that at none of the background stations in India, strong acid radicals like SO_4^{-2} , NO_3^{-1}

and Cl^{-1} have any significant control over the pH of rainwater (Mukhopadhyay *et al.* 1992). This could possibly be due to the fact that the total cation concentration of rain over India is very high and plenty to replace whatever H^+ ions that may form in the solution.

Table 1 (a) shows that five independent mechanisms lead to different combination of weights to different ions. The NO_3^{-1} ions have a relatively low covariance (with other ions), as is seen from the first PC. This must be the only ion that originates to a large extent in the gaseous phase and also from soil. It is scavenged by precipitation with much lower efficiency. The source of NO_3^{-1} ions are likely to be emissions of NO_x gases from soil and internal combustion engines. The first PC which explains about 52% of total covariance must then represent the relative selectivity in scavenging by rain. The second factor has negative weightages for the ions from marine sources (*viz.*, Cl^{-1} and Na^{+1}) and therefore must represent a process of accumulation of pollutants in the atmosphere in the non-rainy days. A high weightage in NO_3^{-1} indicates that anthropogenic emission of NO_x is the principal component in this factor, closely followed by calcium which comes from soil dust under similar dry conditions. It is interesting to note that this factor accounts for only 30.6% of covariances answering why attempts to co-relate industrial emissions with rainwater acidity have not been successful for background areas in India. Other factors beyond the second are not significant. When the factors mentioned above are called upon to explain the pH of rainwater [see Table 1(b)], it is noticed that for Allahabad, Jodhpur, and Srinagar the first two indices computed from the first two factors under estimate the pH slightly. This is not surprising since these stations are known to be rich in alkaline dust (Krishnanand 1984). Also some of the components have not been included in this analysis (*e.g.*, Mg, K). For Mohanbari, Nagpur and Visakhapatnam, the pH is over estimated, because of additional acidic components present in the atmosphere in these regions, which have not been incorporated in the present study for example organic acids (Mukhopadhyay *et al.* 1992). The pH variations in Kodaikanal, Port Blair and Pune are seen to be reasonably well explained by this model.

4.2. Temporal and spatial homogeneity of BAPMoN rain chemistry data

Principal component analyses over space and time domains bring out the character of non-conformity through the comparison of the corresponding weights in the analysed factors. These abbreviations could be a result of several independent causes, each represented by one of the factors.

Chemical characteristics for non-congruous station of years could be attributed to several causes ranging from geochemical to meteorological. It is possible to broadly identify specific regions and epochs.

4.2.1. Spatial homogeneity

Table 2 shows that the weights for sulphates and nitrates are evenly distributed between the stations, with the exception for 'sulphate' at Minicoy. It was earlier reported (Mukherjee *et al.* 1986) and more vividly identified from the BAPMoN data. Mukhopadhyay *et al.*

(1992) found that sulphate depositions in Minicoy were the lowest in the network. This is because Minicoy is a coral island and lacks in mineral diversity compared to the mainland. Much of sulphates at other stations are attributed to the soil and the nitrates from the biogenic sources.

The uniformity in distributions of sulphates and nitrates in rainwater suggests that the network is homogeneous with respect to these parameters.

The first factor for chloride indicates that Minicoy and Port Blair and to a lesser extent Mohanbari, fall in a different category. The influence of the sea on the island stations and strong transport of marine air over the Assam valley during the southwest monsoon are possibly the cause for this pattern of distribution of chloride.

The second factor for chloride also puts Minicoy, Port Blair, Mohanbari and additionally Pune and Srinagar into a group since variations in chloride at these stations occur in a manner different from that for other stations. These variations are caused due to the remnant marine aerosol during seasons other than the monsoon. Proximity to sea in the case of Minicoy and Port Blair and a relatively more significant transport of aerosols with the low level southerly flow from north Bay of Bengal for Mohanbari could make the chloride variations at these stations different from those seen at other stations. Western disturbances that carry marine aerosols from the Mediterranean and Red Sea region and has higher concentrations during a time when the mainland Indian stations get a lesser supply are mainly responsible for chloride in aerosol at Srinagar. The cause for Pune is yet to be explained. Not being due to aerosol phase reaction with strong acids as evidenced in a relatively high Cl/Na ratio (Mukherjee *et al.* 1986), it could ultimately be a reflection of its seasonal pattern of rainfall itself.

Variations in chloride at Nagpur are conspicuous. This could be due to its central location in the country and hence weather systems transporting chlorides from east or west could transport marine aerosols in that region.

Calcium distributions are also quite inhomogeneous. The known strong source regions like Allahabad, Jodhpur and Srinagar do not vary in unison with other stations as is seen from the first factor. Nagpur, Port Blair, Kodaikanal and Visakhapatnam, which probably do not have strong local modulation are better correlated within the network.

Similar to chloride, the second factor is difficult to identify. This could be due to continental dust carried by the westerly wind regime that prevails in the sub-continent during the non-monsoon months. Minicoy, Srinagar and Jodhpur being the western frontiers of the geographic domain capture the highest response and thereby have higher weightage in the second factor.

4.2.2. Temporal homogeneity

BAPMoN rain chemistry has essentially retained the characteristics inter-relationships between the stations over the period of analysis, *i.e.*, 1976-1987. All the

ionic components show remarkable uniformity in temporal behaviour as may be seen from the annual weights in the first factors (Table 3). A large part of the total variances is explained by the first factor. The only significant differences are for sulphate in 1985 and nitrate in 1980. Such variations can be expected due to large fluctuations in spatial distribution of rainfall in these years.

5. Conclusions

Application of principal component analysis has brought out the following results :

(i) More or less uniform spatial pattern of ionic species over the years suggests that industrial pollution has not yet posed a significant problem in background areas in India.

(ii) The pH of rainwater can be very well explained by the interplay between the ion SO_4^{2-} , NO_3^- , Cl^- , Ca^{2+} , Na^+ . However, there are regions where pH is underestimated from these ions and that natural pH is higher. Similarly there are places where opposite results were found.

(iii) Sulphates and nitrates are more homogeneously distributed and may, therefore, have gaseous precursors from the natural emissions.

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