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Rainwater chemistry over Indian sea areas during monsoon season

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सार — मिनिकॉय तथा पोर्ट ब्लेअर में 1979 से 1982 तक प्रतिवर्ष मानसून के मौसम में एकत्रित किए गए वर्षाजल के नमूनों का विभिन्न रासायनिक घटकों के लिए विश्लेषण किया गया है। वर्षाजल की संरचना में विभिन्न अंशदायी स्रोतों के महत्व तथा वर्षाजल के pH को नियंत्रित करने में धनायनों एंव ऋणायनों की भूमिका का अध्ययन करने के लिए आंकड़ों का उपयोग किया है।

ऐसा पाया गया कि दोनों स्थानों पर क्लोरीन तथा सोडियम तत्वों का प्रमुख स्रोत समुद्री हवा है। तथापि, सोडियम के लिए सुदूर स्थित महाद्वीपीय स्रोत भी पर्याप्त महत्वपूर्ण हैं। अन्य धनायनों के लिए महाद्वीपीय स्रोत उत्कृष्ट आपूरक हैं। सल्फेट के लिए समुद्र के अतिरिक्त अन्य स्रोत महत्वपूर्ण नहीं हैं। वर्षाजल तथा समुद्रजल के घटकों के अनुपातों की तुलना से पता चलता है कि मिनिकॉय व पोर्ट ब्लेयर में एकत्रित किए गए वर्षाजल के नमूनों में कैल्सियम प्रचुर मात्रा में था। अध्ययन से पता चलता है कि दोनों ही स्थानों पर क्लोरीन तथा सल्फेट की कमी है। सल्फेट की कमी अनापेक्षित है। pH मान बुनियादी परास में रहे तथा दोनों स्थानों पर धनायनों व ऋणायनों के बीच धनात्मक संतुलन विलयन तथा विनिमयी अभिक्रियाओं के महत्व को बताता है। इनमें विविक्त सामग्री भी शामिल होती है जो H^+ आयनों का उपयोग करती है। नाइट्रेट, सल्फेट तथा H^+ आयनों के बीच महत्वहीन सहसम्बन्ध यह संकेत देता है कि pH नाइट्रिक अम्ल या सल्फ्यूरिक अम्ल द्वार नियन्त्रित नहीं है।

ABSTRACT. Rainwater samples collected at Minicoy and Port Blair during the monsoon season from 1979 to 1982 have been analysed for various chemical constituents. Data have been utilised to study the importance of various contributing sources to the composition of rainwater and the role of cations and anions in controlling the pH of rainwater.

It has been found that maritime air is the major source of Cl and Na at both the stations. However, for Na remotely situated continental sources also appear to be quite important. For other cations, continental sources also appear to be quite important. For other cations, continental sources are the predominant suppliers. For SO_4, sources other than sea are not important. Comparison of ratios between various constituents in rainwater and sea water suggests that rainwater samples at Minicoy and Port Blair get enriched to a large extent in terms of Ca. Studies also show deficiency in Cl aswell as in SO_4 at both the stations. Deficiency in SO_4 is somewhat unexpected. pH values remained in basic range and at both the places the balance between cation and anion is positive which is suggestive of the importance of dissolution and exchange reactions involving particulate material by which $H+$ ions are consumed. Insignificant correlation between NO_3, SO_4 and $H+$ ion indicates that pH is not controlled by HNO_3 and H_2SO_4.

1. Introduction

In the geochemical cycling of various elements, atmosphere plays an important role. Variety of pollutants which are emitted into the atmosphere by human activities enter the natural cycles and rainfall return these pollutants to earth where they can effect biological processes in aquatic and terrestrial ecosystems. Study of chemical composition of rainwater can, thus help in understanding the geochemical cycle of various elements and also in assessing the pollution status of different regions. Since, rainwater can contribute significantly to the nutrient requirement of plants, studies regarding the deposition of various chemical constituents is of importance from agricultural point of view also. In recent years number of studies on rainwater chemistry including Indian area have been undertaken specially over land. However, over oceanic areas rainwater chemistry data are lacking all over the world.. A few measurements, such as over Pacific Ocean (Selezneva 1974) and North Atlantic (Nyberg 1977) were made by collecting the rainwater samples over board ships. However, it is felt that quality of precipitation chemistry data would be better from the regular stations located on small island than a few ship board results. Only report on small island station is by Eriksson (1957) for Hawaii. Infact, even coastal stations are not very much suited for such studies

TABLE 1

Monthly mean values of meteorological parameters at Minicoy during May to September

Month	Mean temp. (°C)	Rainfall (mm)	No. of rainy days	Mean R.H. (%)	Mean surface wind (kmh^{-1})
May	28.8	199.8	9.5	77.0	11.3
Jun	27.6	293.6	17.2	80.0	17.1
Jul	27.3	217.6	14.2	80.5	16.3
Aug	27.3	199.8	12.3	80.5	14.9
Sep	27.3	144.1	10.3	79.0	13.0

TABLE 2

Monthly mean values of meteorological parameters at Port Blair during May to September

Month	Mean temp. (°C)	Rainfall (mm)	No. of rainy days	Mean R.H. (%)	Mean surface wind (kmh^{-1})
May	27.6	362.5	15.1	80.5	12.1
Jun	26.6	589.5	23.5	85.5	19.8
Jul	26.5	435.5	20.5	86.0	19.9
Aug	26.5	435.9	22.5	86.0	18.9
Sep	26.1	516.2	20.4	86.5	15.9

since under certain meteorological conditions the samples may be influenced from the inland sources.

Accordingly, monthly precipitation chemistry data collected from Minicoy and Port Blair which are located in Arabian Sea and Bay of Bengal, respectively for four years (1979-1982) for monsoon season (June to September) have been studied. The above data would give an idea about the background levels of various chemical constituents, their sources and also the pH of rainwater at oceanic locations.

2. Details of sampling and analytical methods

Rainwater collectors at Minicoy and Port Blair are installed in the main meteorological instruments enclosure. At Minicoy, the station is located in the middle of the island and the surrounding country is sandy abounding in coconut trees. At Port Blair the surrounding country (around the station) is undulating and hilly with open sea towards east. Rainwater samples are manually collected with rainwater collector (WMO 1978) and stored in polythylene bottles. Samples are analysed for sulphate (SO_4), chloride (C1), ammonium (NH_4), sodium (Na), potassium (K) calcium (Ca) and magnesium (Mg). pH and conductivity of the samples have been also measured. Analytical methods used are same as those given by Maske and Krishna Nand (1982). The overall accuracy of the analytical results is $\pm$ 10 per cent.

TABLE 3

Monthly mean chemical composition of monsoon rainwater at Minicoy (1979-1982), concentrations are in mg/litres

Month	Rainfall (mm)	pH	Conductivity (μs/cm)	SO_4	Cl	NH_4	Na	Ca	K	Mg
Jun	333	6.75	34.5	0.93	3.73	0.31	3.59	1.30	0.30	0.69
Jul	237	6.66	38.3	0.63	3.74	0.22	4.36	1.41	0.39	0.55
Aug	207	6.55	29.9	0.60	3.65	0.13	3.14	1.21	0.28	0.36
Sep	175	6.46	25.7	0.42	2.46	0.03	2.41	1.14	0.32	0.47
Mean for the monsoon season	855*	6.61	32.9	0.69	3.48	0.15	3.46	1.28	0.32	0.54

*Total rainfall during the monsoon season (long period average).

TABLE 4

Monthly mean chemical composition of monsoon rainwater at Port Blair (1979-1982), concentrations are in mg/litre

Month	Rainfall (mm)	pH	Conductivity (μs/cm)	SO_4	Cl	NH_4	Na	Ca	K	Mg
Jun	478	6.31	34.8	0.60	3.16	0.08	3.04	0.79	0.38	0.32
Jul	406	—	33.1	0.48	3.52	0.01	2.50	0.84	0.35	0.40
Aug	387	6.44	30.5	0.51	3.90	0.01	4.04	0.68	0.21	0.35
Sep	419	6.15	31.8	0.30	2.36	0.06	2.22	0.33	0.28	0.13
Mean for the monsoon season	1977*	6.33	32.6	0.48	3.10	0.04	3.04	0.68	0.31	0.32

*Total rainfall during the monsoon season (long period average)

3. Meteorological features

As per the classification of different seasons in India by Meteorological Department, monsoon season covers the period between June and September. However, rains over Minicoy and Port Blair start in May itself. Monthly mean values of different meteorological parameters at Minicoy and Port Blair are given in Tables 1 and 2 respectively.

Climatological charts showing the wind pattern for July (typical monsoon period for the country) for 850 and 700 mb levels of the atmosphere over the Indian subcontinent and its neighbourhood indicate that dust from Sahara, Arabia, Iran, Somalia and also from northwest India can be transported over Arabian Sea and also Bay of Bengal.

Existence of large quantity of dust over Arabian Sea has been well documented Bunker and Chaffee (1969) had concluded that sources of dust for haze over Arabian Sea are Somalia, Arabia and Iran. Prospero (1979) had got quite high mineral aerosol concentrations over the Indian Ocean. He suggested that Arabian Sea and to a lesser extent the Bay of Bengal are subjected to a very high input rates of mineral dust, rates in the Arabian Sea perhaps being comparable to those experienced on the west coast of north Africa. Source of this material would be the Arabian Peninsula and the arid regions of the northern India, the former being the most important.

Since, the dust particles are known to influence singnificantly the chemistry of rainwater it is expected that samples collected at Minicoy and Port Blair would be also influenced by dust.

4. Discussions

4.1. General considerations

Monthly concentrations of various constituents of rainwater over Minicoy and Port Blair are given in Tables 3 and 4. It can be seen that, in general, the concentrations of Cl and Na are quite high in comparison to other constituents. Surprisingly, C1 concentrations are not that high as obtained in other coastal areas of the world (Eriksson 1952, 1957 and 1960; Junge and Werby 1958 and Sequeira 1976b). Cl and Na concentrations have been observed to decrease significantly during September which represents the withdrawal phase of monsoon and is associated with decrease in monsoon activity and wind speed as well as the roughness of the sea. Kulkarni *et al.* (1982) during their studies at Tarapore had also observed the increase in the concentration of CI and Na with increasing roughness of the sea. Identical pattern for Ca and Mg has been also observed for Port Blair but for Minicoy the above pattern is not clearly discernible. One of the reasons for similar pattern is wind dependence of the concentration of these chemical constituents though they are not fully derived from single source (sources may be sea or soil). Potassium concentrations broadly remained constant at both the stations. NH_4 concentrations have been observed to be significantly higher at Minicoy than Port Blair. This appears to be due to some local source at Minicoy and in absence of sufficient data it is rather difficult to explain the exact reasons for the above.

In general, concentrations of various constituents are low at Port Blair in comparison to Minicoy. pH values which are somewhat higher at Minicoy (between 6.1 and 6.7) indicate basic nature of precipitation. Quite interestingly, conductivity values at both the stations are almost similar. According to Junge (1963), for C1 and Na, sea is the main source and in the present case Arabian Sea and Bay of Bengal are the main sources. Junge (1963) had also mentioned that Ca, Mg and K in rainwater are derived mainly from soil, though sea also contributes to their concentrations.

Deposition values of a few significant constituents such as SO_4, C1 and Na are given in Table 5. SO_4 deposition values are low in comparison to the values reported by others from coastal locations (Eriksson 1952). Cl deposition values are of similar order as observed in USA and South Africa at coastal locations. However, they are lower than the values reported from Europe, Newzealand, Spain as well as from Ceylon (Eriksson 1960). Sequeira (1976 b) data yield the Cl deposition at about 100 kg/ha which is quite high in comparison to deposition at Minicoy. Sequeira (1976 b) had estimated the Na deposition at 54 kg/ha at Bombay and is comparable to the value as obtained at Port Blair but is significantly high in comparison to deposition at Minicoy.

It is the practice (Sequeira 1976 a) to calculate the weight ratios of various chemical constituents with respect to Na to assess the marine influence on their concentrations. Accordingly, the weight ratios of various constituents are calculated and their values are given

TABLE 5

Deposition of SO_4, Cl and Na at Minicoy and Port Blair during the monsoon

Station	Deposition of		
	SO_4 (kg/ha)	Cl (kg/ha)	Na (kg/ha)
Minicoy	5.9	29.7	29.6
Port Blair	9.5	61.3	60.1

TABLE 6(a)

Ratios of chemical constituents during June

Station	Cl/Na	SO_4/Na	Mg/Na	Ca/Na	K/Na
Minicoy	1.07	0.23	0.18	0.37	0.09
Port Blair	0.99	0.19	0.11	0.28	0.12
Thumba*	1.80	0.17	0.11	0.18	0.052
Seawater	1.80	0.25	0.12	0.038	0.036

*Data are for the year 1975 (Sequeira 1976 a).

TABLE 6(b)

Ratios of chemical constituents during July

Station	Cl/Na	SO_4/Na	Mg/Na	Ca/Na	K/Na
Minicoy	0.86	0.15	0.14	0.34	0.08
Port Blair	1.07	0.26	0.21	0.36	0.16
Thumba*	1.60	0.17	0.09	0.18	0.040
Seawater	1.80	0.25	0.12	0.038	0.036

*Data are for the year 1975 (Sequeira 1976 a).

TABLE 6(c)

Ratios of chemical constituents during August

Station	Cl/Na	SO_4/Na	Mg/Na	Ca/Na	K/Na
Minicoy	1.12	0.21	0.16	0.41	0.09
Port Blair	1.25	0.11	0.09	0.20	0.06
Thumba*	1.70	0.07	0.09	0.22	0.043
Seawater	1.80	0.25	0.12	0.038	0.036

*Data are for the year 1975 (Sequeira 1976 a).

TABLE 6(d)

Ratios of chemical constituents during September

Station	Cl/Na	SO_4/Na	Mg/Na	Ca/Na	K/Na
Minicoy	1.12	0.18	0.15	0.58	0.14
Port Blair	1.21	0.20	0.04	0.17	0.16
Thumba*	1.50	0.70	0.08	0.24	0.063
Seawater	1.80	0.25	0.12	0.038	0.036

*Date are for the period 1975 (Sequeira 1976 b) in Tables 6 (a-d). The results are discussed below :

June — Cl to Na ratios are significantly lower than the seawater value (1.8) indicating that either loss in Cl has taken place or for Na there are other sources also in addition to sea water. SO_4/Na values are also low but Ca/Na and K/Na values are quite high. Mg/Na value at Minicoy is marginally higher but at Port Blair it is almost similar to sea water value. Above data suggests significant enrichment of K and Ca in relation to Na.

Enrichment E is defined as $E = \frac{(X/Na)\ R.W.}{(X/Na)\ S.W.} - 1$

where (X/Na) R. W. is the weight ratio in the sample of element X to Na, and (X/Na) S.W. is the same element ratio in seawater. Enrichment of K is of similar order at both the stations (200 per cent) but for Ca the value is significantly higher at Minicoy (870 per cent) than Port Blair (640 per cent).

July — This is a typical monsoon month. In comparison to the ratios obtained during June there has been a slight decrease at Minicoy and increase at Port Blair.

August — This is also a typical monsoon month. C1/Na value has increased slightly. Quite interestingly other ratios have somewhat increased at Minicoy and decreased at Port Blair.

September — C1/Na value has remained similar to August but Ca/Na has increased significantly at Minicoy. Other ratio values have not changed significantly.

Decrease in the C1 to Na ratio with respect to seawater had been observed by other workers also in the past. High ratios between Ca and K with Na in comparison to seawater indicates that in addition to marine sources there are other continental sources also for Ca and K. The values of Mg/Na close to the seawater ratio suggests that for Mg marine source is predominant.

A comparison of the ratios of Minicoy and Port Blair with the values reported for Thumba (Lat. 08° 30′ N, Long. 77° E) by Sequeira 1976(b) as given in Tables 6 (a-d) yield a few interesting results. Thumba is situated on the west coast and is in between Minicoy and Port Blair. Sampling station selected for the measurements at Thumba was about 0.1 km from the coast.

One striking observation is that C1/Na value at Thumba is close to the seawater value. In addition to above. in general. SO_4 deficiency has been observed, an observation which has been made in the present studies also for Minicoy and Port Blair (SO_4/Na values lower than seawater ratio). C1/Na value close to seawater is as per normal expectation but SO_4 deficiency is not normally expected. Sequeira 1976(a) had tried to explain the SO_4 deficiency in terms of very small concentration of SO_2 over the Arabian Sea. Junge (1972) is also of the opinion that the tropical oceans may represent a sink at their surface for SO_2. Sequeria 1976(a) had conjuctured that SO_4 deficient waters may even be present in certain areas of the sea surface releasing SO_4 dificient salt nuclei which are likely to take part in precipitation. Marine upwelling off the coast might be responsible for the above situation.

TABLE 7

Ratios of mean concentrations in rainwater sample during the monsoon season

Ratio	Seawater	Average soil (Vinogradov 1959)	Present work: Minicoy	Present work: Port Blair
Ca/Na	0.038	2.17	0.43	0.25
K/Na	0.036	2.60	0.10	0.12
Mg/Na	0.12	1.00	0.16	0.12
Cl/Na	1.80	—	1.04	1.13
SO_4/Na	0.25	—	0.19	0.19

Complete monsoon season — Mean ratios of various constituents for the complete monsoon season are given in Table 7. A comparison of mean ratios with seawater ratios suggests that rainwater samples at Minicoy and Port Blair get enriched to large extent in terms of Ca and K. However, there is a definite deficiency in SO_4 and C1. Maximum enrichment has been observed for Ca (1030 per cent at Minicoy and 560 per cent at Port Blair). Sequeria 1976(a) had observed Ca enrichments at Thumba between 374 and 532 per cent. Enrichment for K has been about 200 per cent at Minicoy and Port Blair whereas at Thumba it was only about 44 per cent. Considering the fact that during monsoon, wind kick up of soil particles may not be an important process, the probability that the excess Ca is contributed by local soil is quite remote. Thus, Ca excesses in rainwater appear to be due to some foreign continental source.

K/Na ratio is slightly higher than the value obtained (Lodge cited by Rasool 1973) for US in regions close to the sea (0.075). In the present case the sources for excess K and Ca appear to be same, *i.e.*, foreign continental source.

Simultaneous deficiency in Cl and SO_4 present an interesting case at Minicoy and Port Blair. The above deficiency in SO_4 is corroborated from measurements at Thumba also. However, for Cl no deficit was observed at Thumba. The practice of calculating the ratios with respect to Na for assessing the marine contribution assumes that other significant contributors to Na are not present. However, in case, Na is being contributed significantly by continental sources also, Cl/Na ratio would be reduced since it has a lower value in comparison to seawater. It is important to mention here that Rossby and Egner (1955) had also observed Cl/Na values less than 1.5 at number of coastal stations. According to them, one would have to assume that the precipitation falling from air masses near the coast contain a mixture of sodium chloride and some other sodium salt. Once, Na (marine contribution) value has been over estimated, the computation of SO_4 from the above value would give a misleading picture and may yield deficits in SO_4. It is important to mention here that for continental aerosol, value of Cl/Na (water soluble) is less than the seawater. As discussed earlier, atmosphere over Minicoy to a great extent and Port Blair to a lesser extent are influenced by the aerosols from continental sources, and they are contributory to high concentrations of Ca and K (as is evident from high enrichment factors) as well as possibly to Na. From the chemical analysis of aerosols collected on board ships during the monsoon of 1977, Horwath *et al.* (1981) had found the value of Cl/Na as 1.3 for Arabian Sea aerosols (near Minicoy) whereas for Bay of Bengal (near Port Blair) the same was about 1.5. However, Horwath *et al.* (1981) had observed enrichment in SO_4. Above observations are not fully in agreement with the results as obtained at Minicoy, Port Blair and Thumba. It is generally believed that the Cl loss is due to the release of gaseous HCl owing to the interaction of HNO_3 vapour and sea salt particles (Martens *et al.* 1973). However, low levels of NO_x, HNO_3 and nitrate present over marine atmosphere are not sufficient to explain the above phenomenon. Hitchcock *et al.* 1980 had suggested that Cl loss may result from other processes, *e.g.*, from the coagulation of H_2SO_4 particles and sea salt droplets and thus liberate HCl vapour into the air. Thus excess sulphate and chloride loss will be correlated. However, such correlations were not observed even by Harwath, *et al.* (1981). It appears that from limited data it is rather difficult to derive any definite inclusion regarding the mechanism of loss of Cl as well as of SO_4 in rainwater samples over Minicoy and Port Blair. Eventwise sampling and trajectory analysis of the wind may indicate a clue to the above abnormal behaviour.

4.2. *pH data*

pH values are more than 6 at both the stations. However, values at Minicoy are slightly higer in comparison to Port Blair for individual months as well as in the mean for the complete monsoon season. A study of the ionic balance indicates that at both the places the balance between cation and anion is positive, with higher positive balance at Minicoy (in the mean) where higher pH value (mean) has been also observed. Above pH data clearly indicates that pH of rainwater over oceanic stations also is in the basic range which has been the case for other Indian locations (Mukherjee 1957, 1964, 1978; and Krishna Nand 1984).

4.3. *Correlation studies between various chemical constituents*

Minicoy — Significant positive correlations have been observed between Ca, Mg, Na and K. Quite interestingly SO_4 is also well correlated with NH_4. *Insignificant correlation* between SO_4 and Ca indicates that for SO_4 continental sources may not be very important. As expected, correlation between Cl and Na is very good but highest correlation has been observed between Ca and Mg (0.90) which suggests that continental sources are quite important for these elements.

Port Blair — Pattern to correlations is slightly different at Port Blair than Minicoy. Significant positive correlations have been obtained between Ca, Mg and Na only. K is not significantly correlated with Ca, Mg and Na. It indicates that for K some important uncommon source is present. One of the sources could be the vegetation (Crozat and Domergue 1981) which is quite extensive at Port Blair. Cl is significantly correlated with Na as well as with Ca and Mg which suggests important common source for the above elements. SO_4 is significantly correlated with Mg which was not seen for Minicoy. Exact reason for the above is not very clear.

5. Conclusions

Based on the study of rainwater chemistry data from Minicoy and Port Blair during the monsoon season (1979-1982) following conclusions can be drawn :

· Maritime air is the major source of Cl and Na at both the stations. However, for Na, remotely situated continental sources also appear to be quite important. For other cations, continental sources are predominant. For SO_4, sources other than sea are not important.

Deposition values of SO_4 are low in comparison to the values reported by others for coastal stations. Cl deposition values are of similar order as observed in USA and South Africa at coastal locations but are low in comparison to the value reported for Bomb: Na deposition value reported for Bombay is comparable to the Port Blair but is significantly high in comparison to deposition at Minicoy.

Study of ratios between various constituents and a comparison with seawater ra'o suggest that rai.water samples at Minicoy and Port Blair get enriched to a large extent in terms of Ca. E.r chm:nt dec eases from M.nicoy o Port Bla:r wh:h sugres:s th:t h influence of remote continental source decreas d w'th distance, poss'bly due to the effect of washout. Studies also show deficiency in Cl as well as 'n SO_4 at both the stations. Deficiency in Cl had been observed by other workers also. However, deficiency in SO_4 is somewhat unexpected. One of the reasons for the above 's that source of Na at both the stations is not restricted to sea only and thus the value of Cl/Na and SO_4/Na is less than the seawater.

pH data clearly indicates that pH of rainwater over oceanic areas is also in the basic range which has been the case for other Indian locations. A study of the ionic balance indicates that at both the places the balance between cation and anion is positive wh'ch is suggestive of the importance of dissolution and exchange reactions involving particulate material (calcareous minerals) by which H^+ ions are consumed. Insignificant correlation between NO_3, SO_4 and H^+ ion indicates that pH is not controlled by HNO_3 and H_2SO_4.

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