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Prospects of acid rain over India

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सार — भारत में अम्लयुक्त वर्षा की संभावनाओं का पता लगाने के लिए भारत के वायु प्रदूषण मानीटरन संजाल के 10 पार्श्व स्टेशनों से एकत्रित वर्षा के नमूनों के pH आंकड़ों तथा भारत के विभिन्न भागों के साथ-साथ विश्व के अन्य भागों के बारे में वर्षण रसायन एवं वायु प्रदूषण से संबंधित अन्य प्रकाशित आंकड़ों का भी अध्ययन किया गया है ।

देखा गया है कि भारत में बिना प्रदूषण वाले क्षेत्रों में pH का मान 6 से 7 के बीच होता है । pH के मानों का अम्लीयता के मानों (5.6 से कम) तक गिरना कदाचित बम्बई (pH 4.5) जैसे अति औद्योगिक नगरों के इर्द गिर्द तक ही सीमित है । अत : अम्लयुक्त वर्षा की संभावना किसी प्रादेशिक स्तर पर नहीं हो सकती । उपरोक्त का मुख्य कारण क्षारीय पदार्थों की कणिकाओं की उपस्थिति के फलस्वरूप अम्ल-क्षार का परस्पर निष्प्रभावी हो जाना है ।

आगरा के वायुमंडल में निलंबित कणिकीय पदार्थों का अनुपात काफी ऊंचा होने के कारण जिनमें काफी संभावना स्थूल पदार्थों के रूप में होने की भी होती है, आगरा की वर्षा में pH का मान अम्लीय परिसीमा में आने की संभावना नहीं रहती । मथुरा रिफाइनरी के स्थापित होने के बाद भी आशा है इसके मान अम्लीय मानों में परिवर्तित नहीं होंगे, क्योंकि आगरा में प्रदूषक की सांद्रता (विशेषत: SO_2) में वृद्धि होने की अधिक संभावना नहीं है ।

ABSTRACT. pH data of monthly rainwater samples collected from 10 Background Air Pollution Monitoring Network stations in India alongwith the other published data regarding the precipitation chemistry and air pollution from different parts of India as well as from other parts of world have been studied for assessing the prospects of acid rain over India.

It has been found that pH values over unpolluted areas in India may be between 6 and 7 and lowering of pH to acidic values (less than 5.6) might be restricted close to highly industrialized cities only such as Bombay (pH 4.5) and the phenomenon of acid rain may not be of regional nature. Main reason for the above is acid-base neutralization reactions in presence of basic particulate matter.

Due to high level of suspended particulate matter over Agra of which a good fraction is expected to be of coarse mode, pH of rainwater over Agra is not expected to be in the acidic range. It may also not change to acidic values after the commissioning of Mathura Refinery, since the expected increase in concentration of gaseous pollutants at Agra (specially SO_2) is not large.

1. Introduction

Due to increasing industrialization and urbanization air pollution is on increase in India. With a view to assess the prospects of acid rain over India, pH data collected from 10 stations (Fig. 1) situated at Allahabad, Jodhpur, Kodaikanal, Minicoy, Mohanbari, Nagpur, Port Blair, Pune, Srinagar and Visakhapatnam [these are known as Background Air Pollution Monitoring Network (BAPMoN) stations] have been studied alongwith the other published data regarding the precipitation chemistry and air pollution from different parts of India as well as from other parts of world and inferences have been drawn. Location details of nearby pollution sources and weather systems affecting BAPMoN stations are given elsewhere (Krishna Nand *et al.* 1979).

2. Concept of pH

Acidity in a solution is synonymous with the presence of hydrogen ion (H^+) and a common measure of acidity is pH. pH equal to 7 indicates neutral solution whereas less than 7, is acidic and greater than 7 is basic. Rainwater, thus, in its purest form should exhibit pH =7. However, due to the dissolution of carbon dioxide (CO_2) in rainwater (and formation of carbonic acid), pH of even uncontaminated rainwater reduces to 5.65 at 20 deg. C. Further, according to Junge (1963), in the presence of ammonia (NH_3) also (even though in trace quantity $\simeq 3\ \mu g\ /m^3$), the equilibrium value of pH of rainwater would be 7. In fact, absorption of NH_3 is not the only way in which pH value can be raised. Soil particles which are usually alkaline (basic), when swept into the atmosphere by wind get dissolved in the rainwater and release into the solution base cations (positive ion) such as calcium, magnesium, potassium and sodium with bicarbonate usually forming the corresponding anion or negative ion and thus pH value is raised.

In Europe (Barrett and Brodin 1955) and USA (Likens *et al.* 1979) the definition of acid precipitation

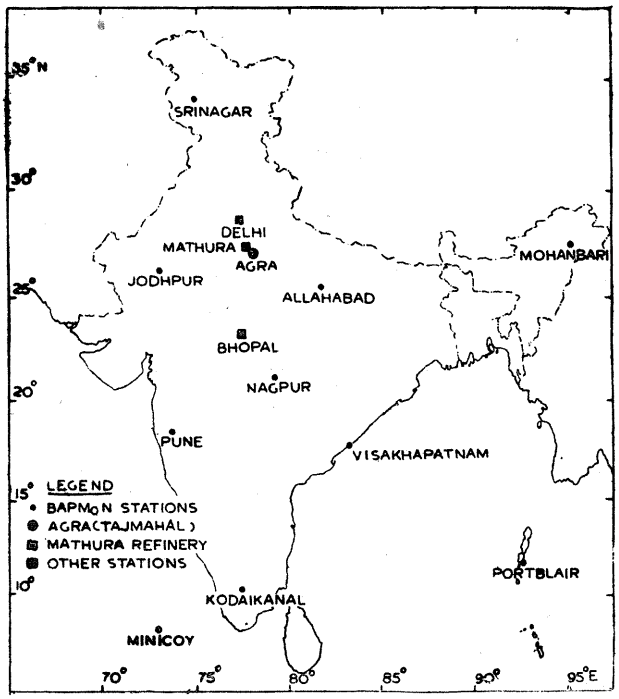

Fig. 1. Location of Background Air Pollution Monitoring Network (BAPMoN) stations in India

is, rain or snow at a pH below 5.6 which is assumed to be the equilibrium value for pure distilled water. pH data as collected from various sources have been studied in light of the above equilibrium value.

3. Details of sampling and pH measurements in India

Rainwater samples are collected by means of a specially designed collector in polyethylene bottles. pH is measured with the help of a standard pH meter.

4. Analysis of pH data from background air pollution monitoring network stations in India (1974 - 1980)

Monthly mean values of pH for different Background Air Pollution Monitoring Network stations are plotted in Figs. 2 and 3. Since arithmetic mean of pH is influenced by extreme values of rainfall amount, rainfall weighted mean values of pH have been used in the present study for the calculation of mean pH spread over number of years for a station. For the study of trend in pH, annual rainfall weighted mean (ARWM) have been also calculated when data for atleast eight months during a year was available for a station and are given in Table 1. Variations in pH at different stations are discussed in the following paragraphs.

4.1. *Allahabad*

pH at the station (Fig. 2) had been almost always more than 7.0. Rainfall Weighted Mean (RWM) for the years 1976 to 1980 is 7.16. Data do not show any definite trend expect that pH at the station was in basic range.

4.2. *Jodhpur*

pH at the station (Fig. 2) had been always more than 7.0 and had large annual variation. RWM value for the years 1977 to 1980 is 7.35. Data do not show any definite trend except that rainwater samples were highly basic in nature.

4.3. *Kodaikanal*

Continuous pH data for a long period are not available from the station. Large fluctuations in the pH values are observed at this station which is somewhat unexpected since it is a hill station and there is no industry surrounding the station. Values of pH as low as 5.8 were observed during 1976. However, RWM value (1976 - 1980) is 6.37 which is in the basic range.

4.4. *Minicoy*

Continuous data for the station are not available for a long period. There is large fluctuation (Fig. 3) in pH values. RWM value for 1977 is 6.45 which is in the basic range.

4.5. *Pune*

It can be seen from Fig. 3 that pH values at the station were in general more than 7.0 and during some months only it had decreased to 6.0. However, the rainfall weighted mean of pH for the years 1974 to 1980 is in basic range (7.02). Study of ARWM data indicate a decreasing trend in pH from 1977 to 1979.

4.6. *Srinagar*

Quite interestingly, almost all the pH values (except in November 1978) were more than 7.0 (Fig. 3). RWM for the years 1976 to 1978 is 7.11 indicating highly basic nature of precipitation samples. ARWM values do not indicate any definie trend in pH.

4.7. *Visakhapatnam*

It can be seen from Fig. 3 that all the pH values during 1976 were less than 7.0. However, subsequently, majority of the pH values during 1977 to 1979 were more than 7.0. pH values during 1980 were again quite low. Since, sufficient data for 1976 are not available, it has not been possible to calculate ARWM value for 1976 and compare the increase in pH between 1976 and subsequent years. At this stage it is also not possible to identify the reasons of such an increase in pH values. However, the study of ARWM for different years indicate a decreasing trend in pH from 1978 to 1980. The rainfall weighted mean for all the years is 6.61 which is basic in nature.

Study of trend in pH based on ARWM values is greatly hampered due to nonavailability of sufficient data throughout the year. As such, in addition to the study of ARWM values, rainfall weighted (MSRWM) means for monsoon season (June to September) have been also studied and the results thus obtained are given in Table 2 and are discussed in the following paragraphs. MSRWM value for any station has been calculated for any year only when data for all the four months, *viz.*, June, July, August and September had been available during the year.

TABLE 1

Annual rainfall weighted mean of pH values for BAPMoN stations in India

BAPMoN station	1975	1976	1977	1978	1979	1980	Rainfall weighted mean pH for all the years
Allahabad	—	—	—	6.99	7.46	—	7.16
Jodhpur	—	—	—	—	—	—	7.55
Kodaikanal	—	6.42	—	—	—	—	6.37
Minicoy	—	—	—	6.91	6.61	6.59	6.74
Mohanbari	—	—	—	—	—	—	6.02
Nagpur	—	—	—	—	—	—	6.32
Port Blair	—	—	6.44	—	—	—	6.50
Pune	—	—	7.21	—	6.78	—	7.02
Srinagar	—	7.54	7.85	6.88	7.30	6.87	7.11
Visakhapatnam	—	—	7.24	7.43	6.90	6.17	6.61

4.8. *Allahabad, Jodhpur, Kodaikanal, Minicoy, Nagpur, Port Blair and Srinagar*

Mean MSRWM values at Allahabad (7.28), Jodhpur (7.34) and Srinagar (7.35) are quite high and there is large annual variation at Allahabad and Jodhpur. A significant fall in MSRWM for 1980 has been observed in comparison to its value during 1978 at Srinagar. MSRWM data from remaining stations are available for one or two years only and they indicate rainfall of basic nature with pH values less than 7.

4.9. *Pune and Visakhapatnam*

Mean MSRWM value for Pune is 7.06 whereas for Visakhapatnam it is 6.62. MSRWM data for different years for Pune (1976 to 1980) indicate a decreasing trend in pH which is quite significant. Decreasing trend at Visakhapatnam has been also observed.

4.10. *pH data from other parts of India*

Mukherjee (1957, 1964) had made first systematic pH observations in India. He had observed pH values between 6 and 7 at Calcutta and Bombay. Recently, Mukherjee (1978) reported pH values at 6.75 for rainwater samples collected on board ship, far away from the land in the Bay of Bengal (excluding the possibility of contamination due to pollution). For Amritsar pH values close to 7 and lower values for Calcutta were reported by Handa (1969) from the measurements carried out during 1967 and 1968. Sequeira (1976) reported pH values as low as 4.5 over some parts of Bombay from measurements made during 1975 and the above trend is still continuing (Mahadevan 1981). Subramanian and Saxena (1980) reported the value of pH over Delhi (close to a thermal power station) between 7 and 8.4 during the monsoon season of 1978. Das *et al.* (1981) reported the value of pH during April to September of 1978 over Bhopal between 6 and 7.2. Agarwal and Lahiri (1979) reported pH value between 7.2 and 8.3 for the monsoon season (1975 to 1977) over Rajasthan (Bikaner, Jaisalmer, Jodhpur, Pali and Palsana).

5. Discussions

It is well-known that sulphur-dioxide and nitrogen oxides are oxidized and hydrolized in the atmosphere to sulphuric and nitric acids respectively and when these acids are present in significant quantities, they can give rise to acid rain. However, as mentioned earlier, pH values in rainfall would decrease only if the concentration of hydrogen ion (H^+) is increased by some process. In presence of sufficient ammonia in the atmosphere, emission of sulphur-dioxide (SO_2) and oxides of nitrogen (NO_x) need not necessarily lower the pH, since SO_2 and NO_x might not get converted to H_2SO_4 and HNO_3 for subsequent hydrolysis to produce H^+ positive ion. Instead, they may be converted to $(NH_4)_2SO_4$ and NH_4NO_3. Since, solubility of ammonia is very large, small quantity of ammonia which is always present in the atmosphere is able to affect the pH of the rainwater in a big way. In addition to above, due to the dissolution of wind blown soil particles in rainwater, an increase in pH can also occur. The influence of particulate matter in altering the pH depends on its particle size distribution. Particles of coarse mode which are usually basic since they are formed from basic materials such as soil, cement and flyash can increase pH significantly.

As such, pH values in rainwater may not change much from 7.0 unless almost all the ammonia in the atmosphere has been converted to $(NH_4)_2SO_4$ or NH_4NO_3 and the base cations from atmospheric particulates have been neutralized. With a view to assess the influence of particulate matter on pH, rainwater

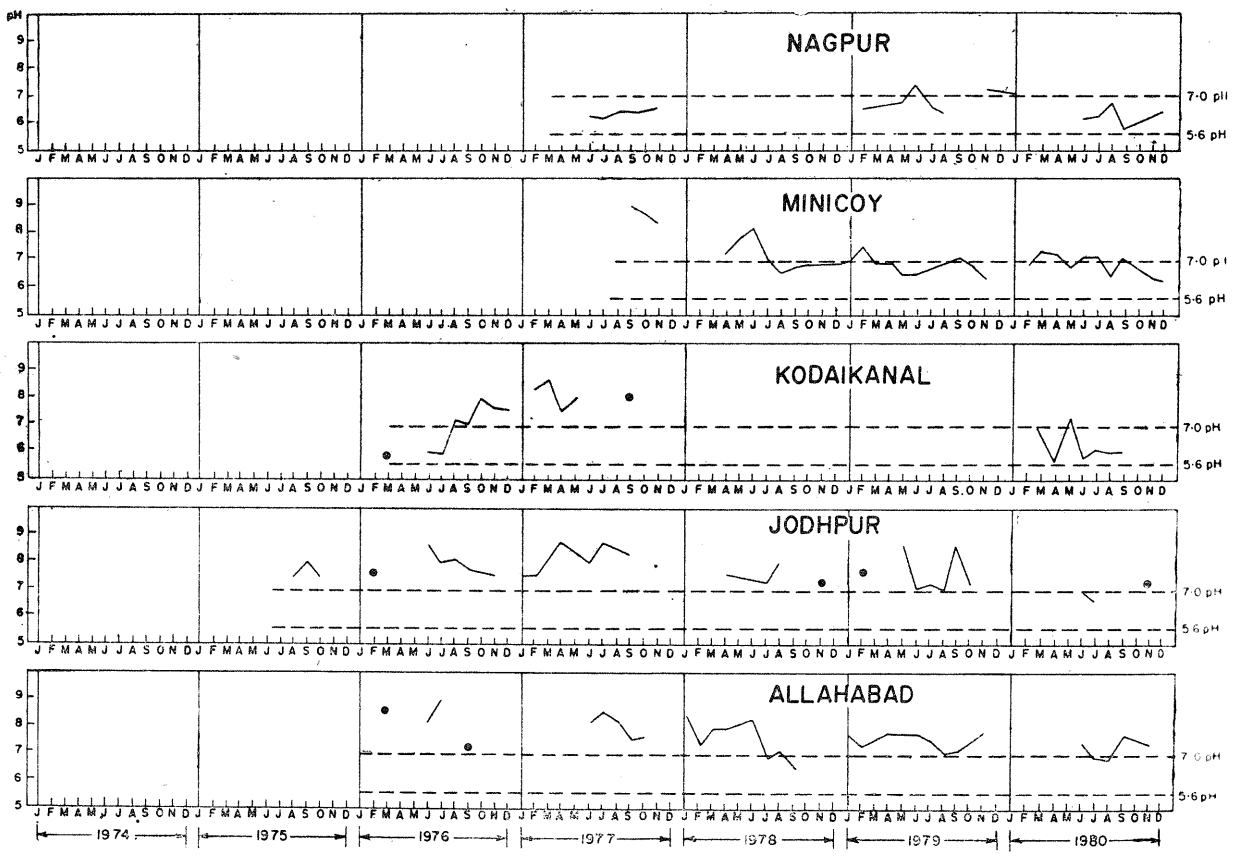

Fig. 2. pH monthly values

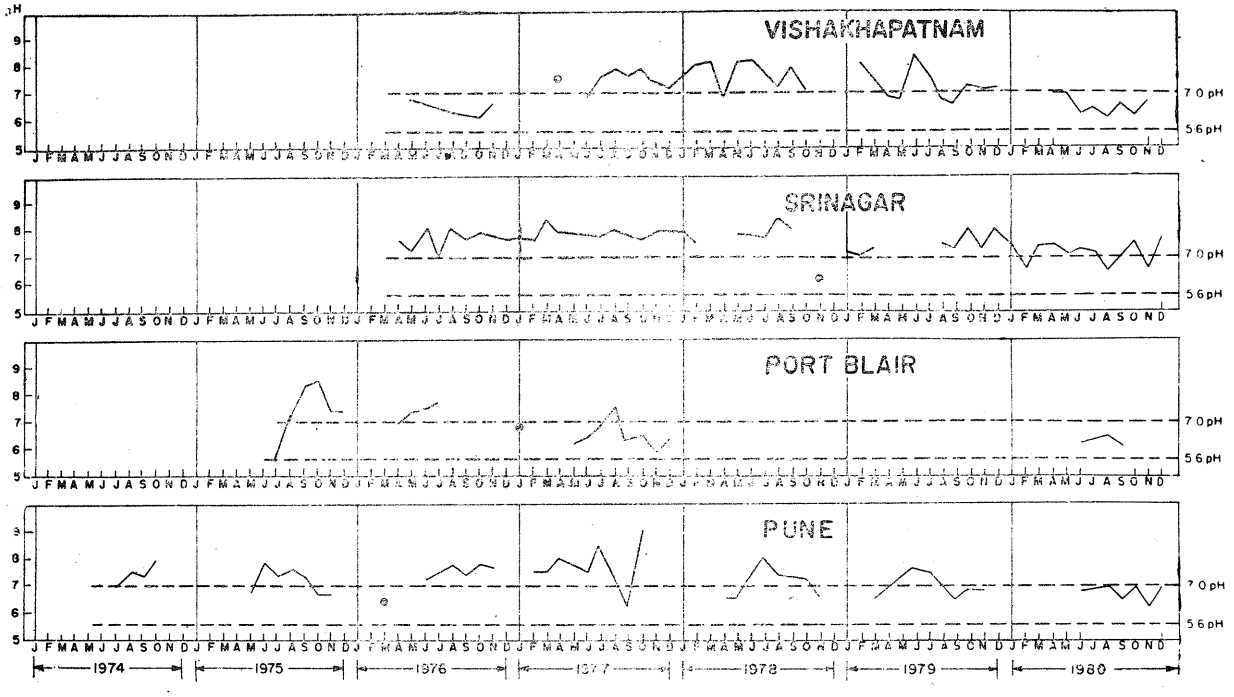

Fig. 3. pH monthly values

samples have been analysed for base cations, *viz.*, calcium, sodium and potassium and the total concentration of all these base cations (rainfall weighted mean) for the monsoon season of 1979 and 1980 in milli equiv/litre have been computed. Since, base cations, specially calcium and potassium are mainly soil derived, their concentrations can be taken as representative of the particulate matter load over the station. It has been found that high pH values are associated with comparatively high values of base cation concentrations. It has been also observed that pH is affected more by the concentration of calcium.

In the light of above discussions, high pH values at Allahabad, Jodhpur, Srinagar, Pune and Visakhapatnam can be probably explained in terms of the dissolution of soil particles (contributing to high base cations) which are swept in the atmosphere from the fields surrounding the station as well as due to the dissolution of ammonia present in the atmosphere. High pH values at Srinagar are due to the high concentration of base cations which are probably transported by winds from deserts mainly situated west of Srinagar. The decreasing trend in mean pH values at Pune and Visakhapatnam suggest that industrialization and urbanization have been able to influence the pH of rainwater at these stations. Kodaikanal, Minicoy and Port Blair represent typical unpolluted stations and yield pH values between 6.5 and 7.0 as observed by other workers also in India.

The role of particulate matter in increasing the pH on the acidity of rainfall over India can be better understood when the pH data from Delhi (Subramaniam and Saxena 1980) close to a thermal power station is considered where the sulphur-dioxide and nitrogen dioxide concentrations are expected to be much higher in comparison to Background Air Pollution Monitoring Network stations. As per the National Environmental Engineering Research Institute's (1980) report, annual arithmetic mean sulphur-dioxide concentrations at Delhi during 1978 were between 7 (residential area) and 61 $\mu g/m^3$. NO_2 concentrations varied between 23 and 37 μ g/m^3 whereas the concentrations of total suspended particulate matter (TSPM) varied between 331 & 431 $\mu g/m^3$. The annual concentration of SO_2 though not exceeding the safe specified limits of 80 μ g/m^3 (United States Environmental Protection Agency, USEPA) from health point of view, was quite high when compared to the values as obtained from other parts of India (NEERI 1980). Alongwith the above, the TSPM concentration were very high and even exceeded the health standards of USEPA (75 μ g/m^3 annual geometric mean). A comparison of concentration of potassium in the rainwater samples at Delhi (0.3 to 10.5 ppm) and Bhopal (0.002 to 0.78 ppm) indicates that the samples at Delhi were influenced by the flyash from thermal power station also in addition to the contribution from other sources.

5.1. *Implication of high suspended particulate matter over India*

From NEERI's (1980) report it can be seen that in general, suspended particulate matter concentrations at all the ten stations in India (Ahmedabad, Bombay, Calcutta, Cochin, Delhi, Hyderabad, Jaipur, Kanpur, Madras and Nagpur) are quite high (ranging between 200 and 800 μ g/m^3, annual arithmetic mean) and it can be reasonably assumed that a good fraction of the above particulate matter will be of coarse mode, except probably at few locations in Bombay where industrial pollution is very high as is evident from NEERI's (1980) data.

Taj Mahal situated at Agra (Fig. 1) which is close to Rajasthan is predominantly influenced by the winds from southwest/west/northwest directions and is likely to be influenced to a great extent in terms of particulate matter and that too of coarse mode. Results of studies conducted at Agra during 1976 showed that annual SO_2 and TSPM concentrations were 15 and 300 μ g/m^3 respectively and NO_2 concentration was 15 μ g/m^3. A comparison of the above data with values at Delhi indicates that NO_2 concentration was quite low whereas SO_2 concentration was high in comparison to residential area at Delhi but was significantly low when compared with the values from industrial area. However, TSPM concentration at Agra was broadly similar to the values as observed in residential area at Delhi but was about 25% lower than industrial area. From the above comparison, it can be argued that the pH of rainwater at Agra may not be in the acidic range and also may not change to acidic values after the commissioning of the Mathura refinery, since the expected increase in the concentration of gaseous pollutants at Agra (specially SO_2) is not large (Das *et al.* 1977).

5.2. *Comparison of pH data as obtained from India and other countries*

Visser (1961) had reported very high pH values (between 5.7 and 9.8) for individual rainwater samples in Kampala, Uganda with medium value at 7.9. Dust was attributed by Visser (1961) as one of the main sources of some of the constituents of the Kampala rainwater and this must have also contributed positively to the high pH. Measurements made by Kasina (1980) indicated that pH values in southeastern Poland were between 4.30 and 5.30. Quite interestingly mean pH values in city industrial area were found to be higher in comparison to remote area. This was explained by Kasina (1980) in terms of higher water soluble content of calcium sulphate and the neutralization of acidic component in the city industrial area. The annual mean weighted pH values in SE Poland were higher (Kasina 1980) to those reported for UK (Marsh 1978), Norway (Dovland *et al.* 1976), NE United States (Galloway *et al.* 1976) and in SE Canada (Whelpdale 1978). It is interesting to mention here that in the eastern USA (where quite low values of pH have been measured) the fine particle mode in the urban aerosol generally dominates the total mass and thus the net aerosol pH is likely to be acidic whereas the aerosols in western USA have greater tendency to be basic. In addition to above, pH data as published by WMO (1981) from the global BAPMoN stations for the year 1978 are also reproduced in Table 3.

From the pH data as collected from different countries, it can be seen that in general pH values are

TABLE 2

Monsoon period (June to September) rainfall weighted mean of pH values for BAPMoN stations in India

BAPMoN Station	1974	1975	1976	1977	1978	1979	1980	Monsoon period rainfall weighted mean pH for all the years
Allahabad	—	—	—	8.22	6.93	7.41	7.15	7.28
Jodhpur	—	—	7.96	8.28	—	7.16	—	7.34
Kodaikanal	—	—	6.18	—	—	—	6.06	6.13
Minicoy	—	—	—	—	6.87	6.67	6.67	6.74
Nagpur	—	—	—	6.21	—	—	6.36	6.36
Port Blair	—	—	—	6.61	—	—	—	6.49
Pune	—	7.44	7.46	7.08	—	6.79	6.77	7.06
Srinagar	—	—	7.68	—	7.74	—	6.95	7.35
Visakhapatnam	—	—	—	7.11	7.53	6.76	6.22	6.62

TABLE 3

Annual 1978 mean of pH values for BAPMoN stations in different countries

S. No.	Country	Mean pH	Remarks
1	Cananda	4.23-5.96	—
2	Czechoslovakia	4.18-4.34	—
3	Denmark	4.25	Data from one station only
4	El Salvador	5.40	Do.
5	Faeroe Island	4.46	Do.
6	Finland	4.34-4.74	—
7	France	4.39-5.25	—
8	German De. Rep.	4.05-4.25	—
9	Greenland	4.39	Data from one station only
10	Hungary	5.11	Do.
11	Ireland	5.17	Do.
12	Italy	5.10-6.37	—
13	Japan	4.83	Data from one station only
14	Malaysia	5.15	Do.
15	Netherlands	4.46	Do.
16	Norway	4.10-4.40	Do.
17	Poland	4.67	Do.
18	Switzerland	5.83-6.09	—
19	Sweden	4.32-4.97	—
20	U.K.	5.23	Data from one station only
21	U.S.A.	4.15-6.19	—
22	U.S.S.R.	5.78-6.64	—
23	Yugoslavia	4.73-4.88	—

high in tropics in comparison to extratropical countries. This is mainly due to the fact that background concentration of particulate matter over tropics is quite high and also the gaseous pollution load in tropics specially in India is significantly low except in few cities. As an example it may be mentioned here that the annual suspended particulate matter concentration in Canada (NAPS 1980) was about 60 $\mu g/m^3$ in 1979 which is much lower than the values reported for India.

In view of the above discussions it can be reasonably argued that over India, lowering of pH to acidic values might be restricted close to highly industrialized cities only such as in Bombay (pH 4.5; Sequeira 1976) and the phenomenon of acid rain may not be of regional nature which is corroborated from the pH data collected from different parts of the country also.

6. Conclusions

1. pH values of rainwater over India is influenced to a large extent by the particulate matter which is present in large (varying) quantities throughout the country.

2. pH values over unpolluted areas in India may be between 6 and 7 and they may be modified to higher values due to further dissolution of wind blown soil particles and ammonia in the rainwater while falling through the atmosphere.

3. A decreasing trend in pH (ARWM values) has been observed at Pune and Visakhapatnam which can be ascribed to the increasing trend in industrialization and urbanization of these places. However, due to random variation in ARWM values of pH for Allahabad and Srinagar it has not been possible to draw any definite conclusion regarding the trend in pH for these stations.

4. Over India, lowering of pH to acidic values might be restricted close to highly industrialized cities only such as Bombay (pH = 4.5) and the phenomenon of acid rain may not be of regional nature. Main reason for the above is acid base neutralization reactions in presence of basic particulate matter. This is evident from the pH data also which has been collected from different parts of the country.

5. Due to the high level of suspended particulate matter over Agra of which a good fraction is expected to be of coarse mode, pH of rainwater over Agra is not expected to be in the acidic range. It may also not change to acidic values after the commissioning of Mathura refinery, since the expected increase in concentration of gaseous pollutants of Agra (Specially SO_2) is not large.

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