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An Assessment of Rainwater Quality from the Tea Growing Areas of Sri Lanka

P. L. K. Tennakoon, L. S. K. Hettiarachchi, O. G. K. A. Gunaratne, R. G. A. Wijayawardhana and G. P. Gunaratne
(Soils and Plant Nutrition Division, Tea Research Institute of Sri Lanka, St. Coombs, Talawakelle, Sri Lanka)

ABSTRACT

Trends towards industrialization along with number of vehicles and their usage have markedly increased during the last two decades, adding a considerable load of undesirable emissions to the environment. There is a distinct possibility of acidic precursors, being transported away from their points of generation because Sri Lanka is situated below the south Asian periphery of the Indian Ocean as an island, underlying prevailing wind directions of the region receiving both south west and north east monsoon rains. Therefore monitoring rainwater quality is thought to be important as it reflects on the extent of air pollution. At the same time since tea is the most important plantation crop, increased soil acidity and mineral loads arising from wet precipitation become a matter of concern.

Hence, a monitoring programme was launched in May 1999, to collect rainwater samples from six meteorological centres of the Tea Research Institute covering a range of agro ecological regions.

Analysis of data over a three year period revealed that highest NO_3^--N, SO_4^{2-}-S and Ca^{2+} loads have been reported from Ratnapura centre. Mean monthly pH of rainwater was in the range of 5.67 - 6.35. Mean values of monthly NO_3^--N deposition was in the range of 0.47 - 1.55 kg ha^{-1}, while corresponding SO_4^{2-}-S, Mg^{2+} and Ca^{2+} were in the ranges of 0.85-2.43, 0.15-0.46 and 0.64-1.54 kg ha^{-1} respectively. Compared with other regional studies, the results of this study indicate that the rainfall receiving for tea growing regions of Sri Lanka is not significantly acidic...

Key words: Acid rain; Wet depositions; pH; Sulphate-S; Nitrate-N

INTRODUCTION

One of the important indicators of environmental pollution is the quality of rainwater. Rainwater is naturally acidic, and the pH of rainwater is primarily controlled by the carbonate buffer reaction and normally has a value around 5.6, depending on the extent of solubility of carbon dioxide in water (Likens *et al.*, 1972). When pH is lower than 5.6, it is generally regarded as an acidic wet-precipitation.

Acid rain is a serious threat to environment in many industrialized countries of the world. The detrimental effects of acid rain include acidification of surface and ground waters with their associated biological consequences, acidification of soils, increased rates of corrosion of building materials and monuments and formation of secondary pollutants such as ozone that are at times hazardous to human and plant life, by complex photochemical reactions taking place in the lower atmosphere from NO_x (NO and NO_2, collectively termed NO_x) and volatile organic compounds (Joshi *et al.*, 1998).

Acid rain is generally due to the presence of sulphuric and nitric acids produced by the atmospheric oxidation of SO_2 and NO_x, primary pollutants emitted from industrial and mobile sources. Sulphur dioxide is emitted mainly from coal burning power stations, smelting operations in the mining industry and also from combustion of fossil fuels in motor vehicles and power plants while nitrogen dioxide is emitted mainly from oxidation of nitrogen gas in the internal combustion engines (Norman *et al.*, 2001).

Sri Lanka, unlike many of its Asian neighbours, is very fortunate that it does not yet have acute air quality problems with its town suburbs including the only metropolitan city, Colombo, as a result of industrial expansion and increased vehicular traffic (Ileperuma and Premakeerthi, 1998). Although there is a tendency to believe that acid rain is not a major problem affecting Sri Lanka, there are a number of locations where acid rain (rain water having pH <5.6) is indeed present which should be of concern to the country (Ileperuma and Premakeerthi, 1998).

Since tea is the most important plantation crop in Sri Lanka, contributing approximately 16% of foreign exchange earnings to the economy, mineral loads and the effects of increased soil acidity arising from acid precipitation in the tea growing regions should at least be a matter of concern to the growers. A study was therefore commenced in May 1999 to monitor rainfall, with the objectives of determining the quality of rainwater and assessing the current mineral loads by wet precipitation, from the six meteorological centres covering different agro-ecological regions in the tea growing areas of the country. This paper reports the outcome of the above, in the first three years.

MATERIALS AND METHODS

Collection of water samples

Monitoring of rainwater quality was commenced in May 1999 in collaboration with the meteorological centres of the Tea Research Institute of Sri Lanka (TRI), situated at St. Coombs in Talawakelle, St. Joachim in Ratnapura, Hantane in Kandy, Pelagahatenne in Passara, Kottawa in Galle and Diyadawa in Deniyaya town suburbs. Elevations of the centres are 1382, 29, 762, 1120, 30, and 250 m (amsl) and agro-ecologically the regions are Wet Up country 2 (WU2), Wet Low country 1 (WL1), Wet Mid-country 3(WM3), Intermediate Mid-country 2 (IM2), Wet Low country 4 (WL4) and Wet Mid-country 1 (WM1) respectively (Panabokke and Kannangara, 1975). Rainwater samples were collected using a home made wet-only collector (20 cm internal diameter and 75 cm height) and analysed since May 1999. According to Krupa (2002), the method used to collect rainwater for chemical analysis and the duration used to collect a sample plays a pivotal role when interpreting the data. Therefore, these factors were taken into consideration when collecting water samples. The sample was collected into a plastic bottle kept inside the collector, specifically designed to prevent dust contamination, and the collector in fact was kept above the ground surface of the meteorological station. As the sampling duration has an important influence on the chemical composition of rainwater (Krupa, 2002), samples were collected daily to minimize this error. In contrast to the bulk samplers, in this wet-only collector the bottle was kept inside the rainwater collector to prevent the collection of dust, absorption of gasses and to minimize the risk of contamination from insects and other debris. A piece of plastic net was kept at the lower part of the funnel to prevent debris from falling into the bottle. The connection between the funnel and the bottle was kept narrow to prevent evaporation of the water in the bottle.

The pH was measured *in situ* every morning following a rainfall not less than 0.1mm, using standard pH meters with a minimum accuracy of ± 0.01 calibrated between 4.00 and 7.00 buffers. Sample was preserved by adding a bactericide (1 ml of 0.1 M Thymol) following the determination of pH *in situ*.

Rainfall was recorded at each centre and samples were labelled and transported to St. Coombs laboratory for subsequent chemical analysis. Samples were kept inside a deep freezer until the analyses were carried out.

Analysis of water samples

Analyses were carried out to estimate nitrate nitrogen (NO_3^--N), by using colorimetric method (Keeney and Nelson, 1982) whilst turbidimetric method was used to determine the concentration of sulphate sulphur (SO_4^{2-}-S) in the sample (Buttlers and Chenery, 1959). To estimate SO_4^{2-}-S, 10 ml of the sample was transferred to a 50 ml beaker, and 1 ml of acid seed solution, containing 20 ppm S as $MgSO_4$ was then added which was followed by an addition of 0.5 g $BaCl_2$ crystals (30 - 60 mesh). This was stirred with a glass rod for 1 minute until the $BaCl_2$ dissolved. The contents were transferred to a spectrophotometer curvette, allowed to stand for 5 minutes and the turbidity read at 420 nm. Ca^{2+} and Mg^{2+} ions were directly read by Atomic Absorption Spectrophotometer and pH of the samples were measured using standard pH meters with a minimum accuracy of ± 0.01 calibrated between 4.00 and 7.00 buffers.

Analysis of data

Statistical analyses were carried out with the aid of Statistical Analysis System package (SAS) Version 6 (Anon, 1995). The data were subjected to the uni-variate procedure of the SAS system, to analyse centres as well as parameters within a centre separately, since the number of rainfall occurrences and number of measurements estimated such as pH, NO_3^--N, Mg^{2+}, and Ca^{2+} etc. differ from centre to centre. Data analysis were carried out for the period from May 1999 to April 2002 by taking the daily rainfall and thereby daily mineral loads. The daily mineral loads were summed up to obtain monthly mineral loads per hectare.

RESULTS AND DISCUSSION

Rainfall distribution

Results of the data analysis for the period from May 1999 to April 2002 are discussed in this section. Table 1 compares the mean annual rainfall received for each centre with the corresponding 75% expectancy value of the annual rainfall (mm) in the respective agro-ecological regions as defined using long term data by Panabokke and Kannangara (1975). Fig. 1 presents the distribution of mean monthly rainfall values for the centres. According to Table 1, the mean annual rainfall values of all the centres are above 75% expectancy values of those respective agro-ecological regions, except for Deniyaya. This may have resulted in with a shortfall of rain over a few weeks, during the study period or else the centres may not fully represent the agro-ecological region.

Sub regions in the Wet zone viz. WU1, WM1 and WL1 receive the highest amount of rainfall while WU3, WM3, WL3 and WL4 receive much lower amount (Panabokke and Kannangara, 1975). However, hardly any pronounced dry periods occur between the two monsoons of the sub regions. In the Intermediate zone, the rainfall pattern is significantly different from that of the Wet zone, having a bimodal pattern of rainfall distribution with a short and a less prominent dry season from May to September (Panabokke and Kannangara, 1975). The mean monthly rainfall values of Hantane, Kottawa and Passara centres were comparatively higher than that of the 75% probability of monthly rainfall values in the agro-ecological regions (Fig. 1). Though the rainfall in the Intermediate zone was expected to be significantly different from that of the Wet zone with bimodal distribution pattern, short and less prominent dry season from May to September (Panabokke and Kannangara, 1975), no such dry spell was observed from Passara centre situated in the Intermediate zone (Fig. 1).

pH variation in wet deposition

Table 2 gives the minimum and maximum, and mean monthly pH values and estimated mineral loads for the meteorological centres where rainwater was collected for the period May 1999 to April 2002. Table 3 gives the daily minimum, maximum and mean concentrations of ions estimated for the meteorological centres for the same period.

Generally, pH value of wet precipitation is determined by the nature and relative proportions of acids and bases present in the solution. According to the current knowledge on atmospheric chemistry, the chief contributors to atmospheric acidity are nitrate and non-sea salt sulphate ions. These are sometimes buffered by the action of cations such as Ca and Mg (Shin- ichi Fujita *et al.*, 2000). The accepted mechanism for the formation of acid rain is by solubilising sulphur oxides and nitrogen oxides to form sulphurous, sulphuric, nitrous and nitric acids which have greater ionisation in the solution than that of carbonic acid formed by carbon dioxide. Thus, the term acid rain has been used to describe rainfall with a pH value less than 5.6 because this is the pH value of distilled water at 25° C in equilibrium with air containing 365 ppm CO_2 at a total pressure of 1 atmosphere or 101.3 kPa (Krupa, 2002).

$$CO_{2\,(gaseous)} + H_2O_{\,(liquid)} + H_2CO_{3\,(aqueous)} = H^+_{\,(aqueous)} + HCO_3^-{}_{\,(aqueous)} \quad pH = 5.6$$

Mean pH values of monthly rainfall over the study period varied from 5.67 to 6.35 for all the centres (Table 2). However, a minimum pH value of less than 5.6 was recorded from all the centres except Deniyaya (Table 2). Of the monthly mean pH values of 36, the pH values that are lower than 5.6 were found mostly in Ratnapura, Kottawa and Talawakelle centres and Ratnapura being the most (data not shown). The results therefore indicate that the acidification of rain has not occurred significantly in all the tea growing regions of the country. However, it appeared in some centres at random particularly when pH values of daily rainfall from Hantane, Kottawa, Passara, Ratnapura and Talawakelle meteorological centres were examined and the Ratnapura centre showed the most.

Results of a monitoring programme on rainwater quality in Sri Lanka carried out by Ileperuma and Premakeerthi (1998), over a 3 year period covering the Wet, Intermediate and Dry zone using 18 meteorological stations since early 1995 also showed similar findings. They showed that although very low pH (< 5.6) values were found at random, the mean pH values of rainwater varied between 5.89 -7.00. A rainwater monitoring study conducted in India during 1991-1996 covering 10 major metropolitan cities viz. Ahmedabad, Calcutta, Chennai, Delhi, Hyderabad, Jaipur, Kanpur, Kochi, Mumbai and Nagpur showed that the average annual pH was in the range of 6.0-7.9 while the lowest was recorded as 4.8 at Kochi and Delhi in 1991 and 1994 respectively (Joshi *et al.*, 1998). It has also been reported that a common feature in most studies of the chemistry of Indian precipitation is that the high pH values (pH≥6) prevailed during all seasons and all over the country. The lack of acid rain despite relatively large man-made emissions of acidifying oxides of sulphur and nitrogen indicates the presence of neutralizing compounds like calcium carbonate and ammonia in rainwater (Norman *et al.*, 2001; Barman, 1998).

Unlike in India the results of the chemical composition of precipitation from 18 sites in East Asia, from a cooperative monitoring performed during the period from 1992-1993 showed that the annual mean pH of the 18 sites was approximately 4.9. The maximum mean annual pH was observed at Jinan of China (pH 6.1) and minimum at Tokyo in Japan (pH 4.5). Although north - eastern China had the sources of largest anthropogenic emission causing precipitation with relatively low pH in the same area precipitation with relatively high pH value was observed in Islands in the north Pacific, which is much far from major anthropogenic emission sources (Shin- ichi Fujita *et al.*, 2000).

Wet deposition of ions

The analysis of nitrate and sulphate gives an indication as to the sources of environment pollution. The results of this study show that the sulphate loads are fairly high (Table 2) compared to other mineral loads. Sulphate in rainwater arises mainly from two factors; exhaust fumes from mobile sources and industrial stacks, thermal power plants and seawater spray, especially in coastal areas (Shin - ichi Fujita *et al.*, 2000). The highest mean monthly depositions were at Ratnapura, Kottawa and Deniyaya (Table 2). Out of these three centres Kottawa and Deniyaya are closer to the sea and Kottawa is the closest (less than

5 km in displacement). Therefore the high sulphur loads in these regions may be partly due to the sea spray containing SO_4^{2-} - S (As evident from the primary data presented in Table 3). The mean of monthly rainfall distribution during the period of study for the centres, are shown in Fig. 1 while Table 2 gives mean values. According to Table 2 and Fig. 1, locations receiving higher rainfall have greater depositions of sulphur. Similar relationships have also been observed from a study carried out over a period of 12 months to assess sulphur deposition by rainfall in some agriculturally important areas of Sri Lanka covering Wet-, Intermediate- and Dry- zones (Amarasiri and Lathiff, 1982).

Extreme outliers of nitrate, sulphate, magnesium and calcium loads have also been observed from the data pools subjected to the univariate procedure of the SAS system, from all the centres. Although the mean of monthly SO_4^{2-} - S loads are comparatively lower in Talawakelle and Hantane centres (Table 2), extremely high SO_4^{2-} - S loads have also been found at random in July 2001 from these two centres, and the loads being 4.66 and 5.43 kg ha^{-1} respectively. Extremely high SO_4^{2-} - S loads have also been observed from Passara during the periods of October and November 1999 and August 2000 and the loads are 4.68, 1.35 and 2.3 kg ha^{-1} respectively. During these periods, the above centres had received much higher rainfall than their mean monthly rainfall values, except Hantane (data not shown). Therefore, it is apparent that the high sulphur loads would have probably been brought down by the high rainfall as opined by many researchers (Amarasiri and Lathiff, 1982; Shin-ichi Fujita *et al.*, 2000 and Irwin *et al.*, 2002).

As in the case of SO_4^{2-} - S, highest NO_3^- -N loads have also been recorded from Ratnapura (Table 2). High NO_3^- -N loads arise mainly due to automobile exhaust fumes. The NO_3^- - N loads in Ratnapura centre may have resulted partly from the acidic fumes transported from the western province, which usually shows high levels of pollution in atmosphere, depending on the movement of wind direction and other climatic factors. Extremely high NO_3^- -N loads in rainwater have also been observed from all the centres. Like in the case of SO_4^{2-} - S deposition, relatively high rainfall figures have also been reported during the periods of extremely high NO_3^- -N loads (data not shown), except in Deniyaya during March 2002. It has also been reported that the rain act as an effective cleansing agent, flushing particulate and other water-soluble gases out of the atmosphere (Samarakkody *et al.*, 1998 and Waite *et al.*, 1999). Therefore, high NO_3^- - N depositions observed in this study would also have been brought down with high rainfall. Precipitation monitoring studies carried out by Shin-ichi Fujita *et al.*, (2000) in east Asia and by Irwin *et al.*, (2002) in United Kingdom also showed that higher the rainfall, higher the SO_4^{2-} - S and NO_3^- -N depositions.

Alkaline components like Mg^{2+} and Ca^{2+} has a neutralizing influence on the acidity of rainwater and thereby in general pH in rainwater increases (Barman, 1998). However, when means of monthly pH values observed from the Ratnapura and Kottawa centres in this study were examined together with their base depositions (Table 2), no significant association between these two components was apparent, and this is likely to be due to the presence of higher acidifying components in rainwater than the basic ions.

Results from this study showed that the mean values of monthly SO_4^{2-}-S and NO_3^--N depositions were in the range of 0.85 - 2.43 kg ha^{-1} and 0.47 - 1.55 kg ha^{-1} respectively in the tea growing regions in Sri Lanka. Ion depositions received over 12-month period was worked out by summation of average monthly depositions and presenting as kg per hectare per year. Accordingly, SO_4^{2-}-S and NO_3^--N depositions have been arrived at and the values were in the range of 10.4 - 29.2 and 5.2 - 18.6 kg ha^{-1} yr^{-1} respectively. The study carried out by Amarasiri and Lathiff (1982) on sulphur depositions by rainfall using rain gauges for collection in some 10 agriculturally important areas of Sri Lanka covering Wet-, Intermediate- and Dry- zones showed that the annual SO_4^{2-}-S loads were in the range of 7.0-23.9 kg ha^{-1} yr^{-1}. Hardly any other literature is available as far as Sri Lanka is concerned. Apart from the differences in the collector, when results from these two studies are compared it indicates that the annual SO_4^{2-}-S loads received have marginally increased during the last two decades.

In comparison to the nitrogen requirements of the tea crop, it is unlikely that the quantity of nitrogen brought down by rainfall would have significantly contributed to compensate the plant needs. Contrary to nitrogen, sulphur requirements is relatively low as the sulphur concentration in the harvestable young shoots are as low as 0.25 to 0.4% when compared to N, concentration of N being 3 to 4%. Thus the S removal with harvested tea crop is small and only 8 - 12 kg S is removed even with a harvest of 3000 kg made tea ha^{-1} yr^{-1}. Hence when taking into comparison the S needs and monthly S quantities brought down as wet deposition, it is probable that the quantities brought down would have partly supported to meet up the requirements.

In comparison with some regional studies carried out by Joshi *et al.*,(1998) in India and Shin- ichi Fujita *et al.*, (2000) in East Asia, though the types of samplers used differ, it can be clearly seen that the chemical loads deposited in tea growing regions in Sri Lanka are relatively at a low level (Table 4).

CONCLUSION

Highest NO_3^--N, SO_4^{2-}-S and Ca^{2+} loads were reported from Ratnapura. Mean monthly pH was in the range of 5.67 - 6.35 among the stations. Mean values of monthly NO_3^--N deposition was in the range of 0.47 - 1.55 kg ha^{-1} while SO_4^{2-}-S, Mg^{2+} and Ca^{2+} depositions were 0.85 - 2.43, 0.15 - 0.46 and 0.64 -1.54 kg ha^{-1} respectively. When compared with other regional studies, the results of this study clearly indicated that the tea growing regions of Sri Lanka is not receiving significant levels of acid rain at present condition of anthropogenic activities.

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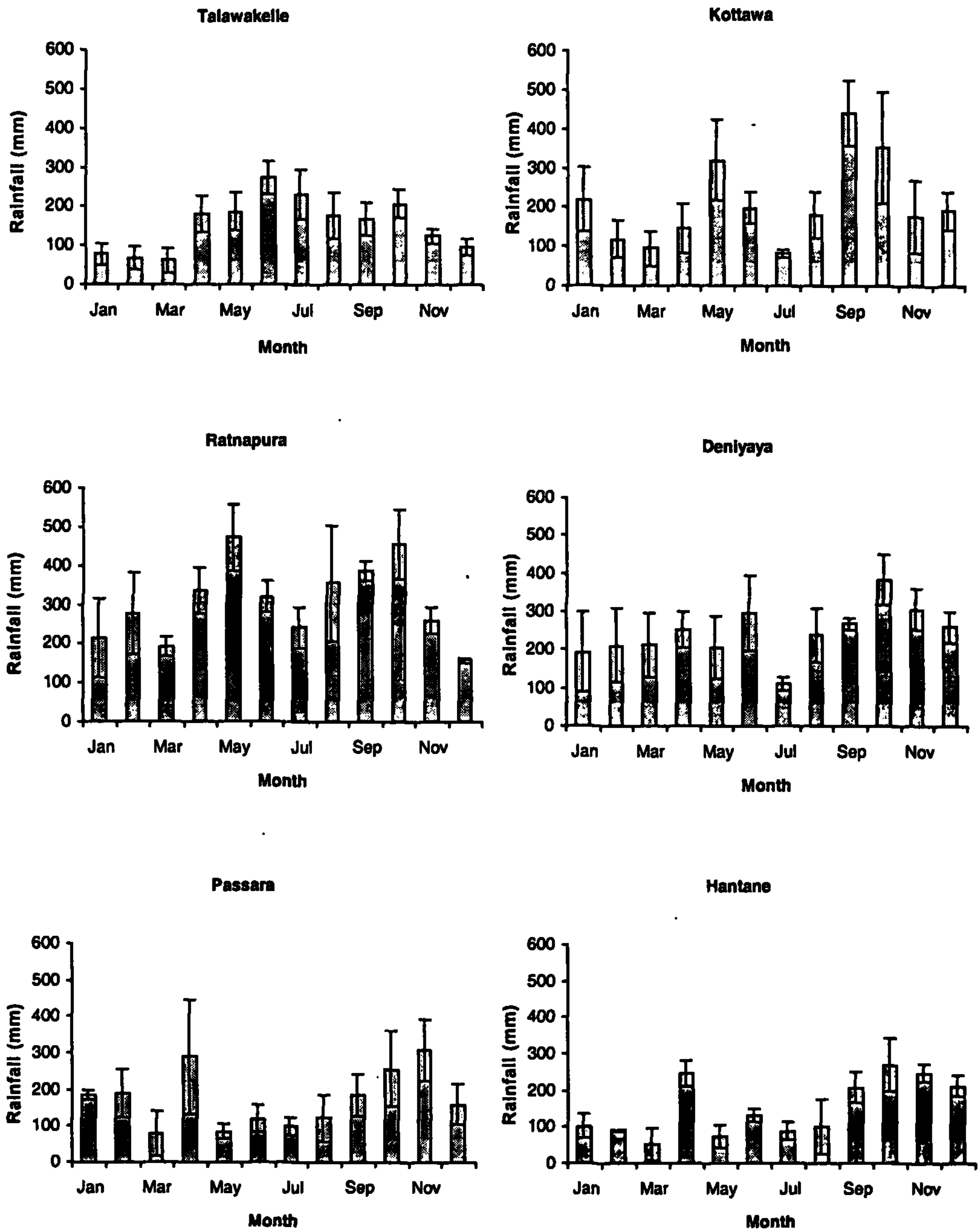

Fig. 1. **Monthly rainfall distributions at the meteorological centres during the period of study; Vertical bars represent the standard errors of the means.**

Table 1. Mean annual rainfall between meteorological centres and agroecological regions

Centre	Agro-Ecological Region	Mean annual rainfall (mm)	75% expectancy value of mean annual rainfall (mm)
Deniyaya (DN)	WM1	2988	>3150
Passara (PS)	IM2	2088	>1150
Hantane (HN)	WM3	2002	>1250
Ratnapura (RP)	WL1	3540	>2525
Kottawa (KT)	WL4	2400	>1525
Talawakelle (TK)	WU2	2004	>1900

Table 2. Minimum, maximum and mean values of monthly rainfall, pH and estimated mineral loads from the sample collected

Centre	Rainfall (mm)			pH			NO_3^--N (kg ha^{-1})			SO_4^{2-}-S (kg ha^{-1})			Mg^{2+} (kg ha^{-1})			Ca^{2+} (kg ha^{-1})		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
DN	8.2	517	249 (496)	5.69	6.89	6.35 (130)	*	3.71	0.67	*	5.41	1.78	*	1.82	0.23	*	2.89	0.80
HN	6.8	415	166 (386)	5.58	6.57	6.13 (280)	*	1.58	0.47	*	5.43	0.85	*	0.43	0.17	*	3.85	0.92
KT	1.8	628	200 (525)	5.07	6.67	5.86 (501)	*	3.93	0.61	*	5.74	2.06	*	3.09	0.46	0.14	7.79	1.29
PS	24.4	473	146 (399)	5.45	6.35	5.96 (182)	*	3.85	0.92	*	4.68	0.95	*	0.99	0.21	0.13	2.38	0.64
RP	42.2	691	295 (572)	4.83	6.42	5.67 (561)	*	5.58	1.55	0.22	8.30	2.43	0.05	0.89	0.38	0.16	5.53	1.54
TK	11.6	350	167 (511)	5.21	6.46	6.08 (283)	*	3.01	0.60	*	4.66	0.98	*	0.46	0.15	0.08	2.92	0.64

* Values less than 0.05 recognized as negligible

** Number of rainfall occurrences and pH measurements are given in the parenthesis

** Percentage of acid rain (pH<5.6) samples out of total pH measurements in HN, KT, PS, RP and TK were 4.6%, 18.2%, 5.4%, 31.9% and 8.5% respectively.

*** Number of measurements taken for NO_3^- - N, SO_4^{2-}- S, Mg^{2+} and Ca^{2+} in DN, HN, KT, PS, RP and TK were 130, 280, 501, 182, 560 and 283 respectively.

Table 3. Minimum, maximum and mean concentrations of ions estimated from the daily collected samples

Centre	Ionic Concentration (ppm)											
	NO_3^- - N			SO_4^{2-} -S			Mg^{2+}			Ca^{2+}		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
DN	ND	4.79	0.48	ND	5.48	1.09	ND	0.49	0.09	ND	3.45	0.64
HN	ND	4.91	0.34	ND	6.36	0.79	ND	3.05	0.13	ND	4.21	0.58
KT	ND	3.25	0.35	ND	24.6	1.36	ND	3.32	0.29	0.01	4.30	0.71
PS	ND	4.45	0.57	ND	2.82	0.72	ND	3.05	0.18	0.01	1.55	0.43
RP.	ND	8.12	0.66	ND	9.80	0.93	ND	0.98	0.15	0.01	4.88	0.56
TK	ND	4.38	0.42	ND	7.70	0.84	ND	0.98	0.12	0.01	2.96	0.51

* ND stands for values not detectable

** Number of measurements taken for NO_3^- - N, SO_4^{2-} -S, Mg^{2+} and Ca^{2+} in DN, HN, KT, PS, RP and TK were 130, 280, 501, 182, 560 and 283 respectively

Table 4. Average annual pH values observed and annual deposition rates of sulphate - S and nitrate - N from rainfall in some industrial areas of the world

Location	Year	pH	NO_3^--N ($kg\ ha^{-1}$)	SO_4^{2-}-S ($kg\ ha^{-1}$)	Sampler type
India (10 major metropolitan cities); Joshi *et al.*, (1998)	1991-1996	6.0 - 7.9	3.4 – 97	16 – 287	Bulk collectors (on daily basis)
East Asia (18 cities including Japanese Islands, Korean peninsula and central parts of China); Shin- ichi Fujita *et al.*, (2000)	1992-1993	4.5 - 6.1	4.8 – 26	27 – 71	Wet only collector (on 10 day intervals)
United Kingdom (32 cities); Irwin *et al.*, (2002)	2002	N/A	6.3 – 22	7.2 – 27	Refrigerated wet only collector (on daily basis)