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Variation of trace element concentration in rainwater with rainfall rate and rainfall amount

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ABSTRACT. The variation in trace element concentration in rainwater with rainfall rate and rainfall amount for collections during monsoon at Lonavala and Bombay are discussed. The results confirm the known inverse relationship between concentration and rainfall rate as well as rainfall amount. The decrease in concentration with rainfall rate is related to increasing drop diameter and liquid water content of the precipitating cloud. Effect of washout on the trace element concentration in rainwater is not significant. The maximum to minimum concentration ratio is significantly less for I, K and Br as compared to other elements like Na, Cl, Ca or Mg.

1. Introduction

The concentration of trace constituents in precipitation reaching ground level is governed by physical processes like formation of cloud droplets, growth of the raindrops, the scavenging of particles and gases by the various precipitation elements in the cloud and the sub-cloud layer washout of atmospheric aerosols. The concentration in rainwater of any contaminant is generally known to vary inversely with the amount of rainfall and a similar relationship is also obtained in individual rainfalls between the concentration and precipitation rate (Sugawara *et al.* 1949, Woodcock and Blanchard 1955, Mukherjee 1956, Facy 1962, Junge 1963 and other references therein). The inverse relationship was attributed by Junge (1963) to droplet evaporation, amount of liquid water content in cloud and the contribution of washout. Engelmann (1970) suggested that the observations can be explained by the varying contribution of coalescence and condensation to raindrop growth since the former process will tend to keep the specific concentration constant while the effect of the latter process will be to decrease the specific concentration. This paper presents and discusses the variation of the trace element concentrations in rainwater at Bombay and Lonavala with rainfall amount and rainfall rate.

2. Data used

The sampling and analysis method and the complete data are available elsewhere (Sadasivan *et al.* 1974, Sadasivan 1977, Sadasivan 1979). Essentially the trace element concentration in daily or twice daily rainwater samples collected during monsoon at Bombay and the concentration variation in short duration samples collected near cloud base at Lonavala have been discussed.

2.1 *Variation with rainfall amount*

The variation in Ca content with rainfall amount in twice daily samples collected during monsoon at Colaba, Bombay, during 1973 is presented in Fig.1. The concentration decreases with increase in rainfall amount, following the familiar inverse relationship. An analysis of the data shows that this inverse relationship holds for all the elements measured. Such a result was also obtained for specific radioactivity of rainwater collected at Bombay (Sadasivan 1967). For rainfall amounts less than 1 mm the concentrations show tremendous increases while for 1 to 5 mm rainfall there is a scatter. Typically, the values for Cl lie between 5-40 μg/ml. The scatter in the values is further reduced for higher rainfall amounts as is reported in the literature (Junge 1963, Georgii and Weber 1960).

The contribution from washout and droplet evaporation to the specific rainwater concentration will not explain the observed inverse relationship in the present case. For most of the sampling periods the cloud bases were very low while relative humidities were very high. In a few instances in July and August when collection conditions like rainfall amounts, rainfall modes (*e.g.*, very short duration, continuous precipitation etc) and surface air concentrations were similar, the rainwater concentration was lower in August than in July. Hence the ground level

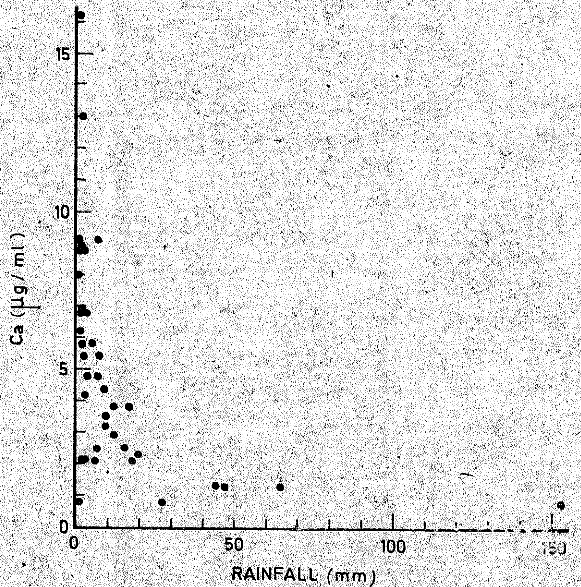

Fig. 1. Variation in Ca concentration with rainfall amount in rainwater samples from Colaba

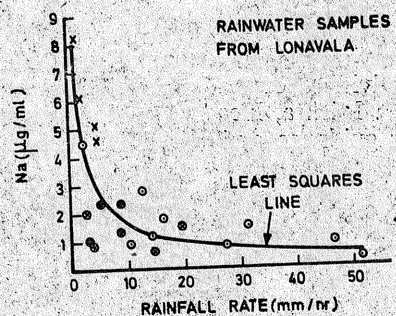

Fig. 2. Variation in Na concentration with rainfall rate

rainwater concentrations are seen to be influenced only by the concentration in the cloud droplets. Also the inversion relationship cannot be explained by rainfall rates since even low rainfall amounts were quite often the result of passing showers with high rainfall rates. The spacing between rainfall periods, that is the length of dry period in between two collections and fairly equal rainfall duration do not correlate with increased concentration in small rainfall amount samples which again indicates that the effect of washout is not significant. So the measured inverse relationship can be due only to the element concentrations in cloud water, the absolute humidity of the entraining air and the efficiency of water removal (Engelman 1970).

The data also showed that the maximum to minimum concentration ratio is more for Na or Cl than for I and Br. These variations result from the differences in particle size distribution of the elements. Investigations on the maritime aerosol compositions showed (Sadasivan (1978) that the ratio of concentration in > 0.1 μm radius particles (impactor stages) to < 0.1 μm radius particles (back-up filter) is more by a factor of 2 to 3 for Na and Cl as compared to the ratios for Br and I. Also the ratio of maximum to minimum concentration, in impactor stages first to fourth is more for the two elements Na and Cl. Hence, addition of large and giant particles to precipitation elements in its formation and growth is likely to cause larger variation in rainwater for Na and Cl. The variations in Ca and Mg concentrations with rainfall amount were similar to Na and Cl while K variation was comparable to I and Br.

2.2 *Variation with rainfall rate*

The variation in Na concentration with rainfall rate for four sets of samples from Lonavala, where continuous, short duration sampling was carried out during July 1971, is shown in Fig. 2. The four sets of samples are represented by different symbols. It is seen that the concentration decreases with increasing precipitation rate, as has also been reported in the literature (*e.g.*, Woodcock and Blanchard 1955). The other trace elements measured also vary similarly. All the points can be adequately fitted by a curve of the form $k_{Na} = AR^{-B}$, where k_{Na} is the Na concentration, R is the rainfall rate in mm/hr and the constants have values $A = 7.95 \pm 0.95$ and $B = 0.70 \pm 0.10$. The raindrop diameter (D_n) as well as liquid water content (L) are related to rainfall rate by similar function with a positive exponent value. The value of the exponent for Indian orographic rains is 0.90 for estimating the liquid water content (Ramana Murty and Gupta 1959) while for Hawaiian orographic rain at cloud base, the exponent value for calculating drop diameter is 0.37 (Blanchard 1953). The decrease in concentration with precipitation rate is thus seen to be related to increasing drop diameter as well as increasing liquid water content of the precipitating cloud. The samples were collected near cloud base and hence there can be no effect due to below cloud washout. Also the total contribution to element concentration in rainwater at cloud base by in-cloud scavenging of aerosols by precipitation elements is not very significant and it is only at the nucleation stage that the contaminant incorporation is maximum (Junge 1963, Engelmann 1970, Lee and Dingle

1974). Thus the measurements confirm (Engelmann 1970) that the observed inverse relationship between concentration and rainfall rate is the result of varying contribution by condensation and coalescence to raindrop growth. A similar conclusion was arrived at in an earlier investigation on the specific radioactivity variation in individual showers (Bhatnagar and Sadasivan 1965).

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