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Rainwater chemistry at a regional representative urban site: influence of terrestrial sources on ionic composition

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Abstract

In the present investigation, the rainwater chemistry is studied over a one year period at a regional representative urban site, Tirupati. A continuous record of 67 events were included that ranged from 0.5 to 74.6 mm (annual total 850.2 mm), from which 35 events (52%) were chemically analyzed during the period for major inorganic ion composition viz. F, Cl, NO_3, SO_4, Na, K, Ca, and Mg as well as NH_4. The volume-weighted mean (VWM) of the measured ionic sum is 501.28 $\mu eq\,l^{-1}$ and the crustal ion contribution dominating over the acidic ions with a ratio of total cation to that of anions (Σcations/Σanions) is 1.4. Only the Ca and SO_4 together made up more than 55% of the total ion, whereas Ca alone contributed about 51% of the total cation. All the rain samples analyzed showed alkaline pH always, to that of reference level 5.6, with VWM of 6.78. On the basis of good correlation between the crustal ions (Ca, Mg, K and NH_4) and SO_4 ($r = 0.95$) and NO_3 ($r = 0.91$) as well as order of neutralization factors Ca (0.88) > Mg (0.33) > NH_4 (0.11), it has been determined that the acid neutralization is brought about by crustal ions, particularly calcium, in the region. The data were subjected for correlation analysis. A good correlation observed between Ca and SO_4 and NO_3 ($r = 0.92$ and 0.90) and Mg and SO_4 and NO_3 ($r = 9.83$ and 0.77), respectively, reveals that the ionic composition of rainwater in the region is strongly influenced by terrestrial sources rather than anthropogenic and marine sources.
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1. Introduction

Precipitation chemistry is an intricate result of a complex interaction between cloud dynamics and microphysical processes as well as a series of in-cloud and below-cloud atmospheric chemical reactions. The acidity and ion concentrations in rain depend on the source strength of the constituents, their physical corporation into the hydrological system, chemical transformation during cloud formation and below cloud scavenging (Kulshrestha et al., 1999). Incorporation of sulphur and nitrogen oxides resulting from fossil fuel combustion, to precipitation is particularly important as they are the precursors of major acids (H_2SO_4 and HNO_3). In poorly buffered watersheds, acid deposition leads to acidification and results in notable adverse effects on the ecosystem and indirect effects on human health along with changes in the species composition and abundance. Emissions of alkaline substances (dust particles and gaseous NH_3) significantly influence the

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precipitation acidity by neutralizing some fraction of the acids (Placet and Streets, 1987; Khemani et al., 1985). Wet deposition constitutes an important natural pathway for the removal of atmospheric pollutants. However, contamination of rainwater by atmospheric pollutants is of growing concern on both regional and global scales (Galloway et al., 1984). Determination of chemical composition of rainwater, provides an understanding of the source types that contribute to rainwater chemistry, and enhances the understanding of the local and regional dispersion of pollutants and their potential impacts on ecosystems through deposition processes. Thus, precipitation chemistry has been the subject of intense research since the last two decades (Galloway et al., 1984; Loye-Pilot et al., 1986; Lacaux et al., 1992; Smirnioudi and Siskos, 1992; Ayres et al., 1995; Tuncel and Sevgi, 1996; Galpin and Turner, 1999; Lee et al., 2000; Marquardt et al., 2001).

In contrast to Europe and the US, emissions of air pollutants in Asia are increasing rapidly (Higashino et al., 1997) resulting from its large population, rapidly growing economy and the associated systems of energy consumption and production. The potential for air pollution problems in Asia is great due to high emissions and close proximity of many major industrial and urban centers. In India, soil has been considered as a major contributor of the particulate in the atmosphere, owing to its dusty nature (Khemani et al., 1985; Mahadevan et al., 1989). In Indian context, both the rainwater and aerosols are mostly alkaline, which is due to low contribution of secondary aerosols (SO_4 and NO_3) from anthropogenic sources and high contribution of primary aerosols (Ca, K and Mg) from soil (Khemani et al., 1985; Mahadevan et al., 1989; Naik et al., 1995; Kulshrestha et al., 1999; Saxena et al., 1996; Satsangi et al., 1998; Balachandran and Khillare, 2001; Thakre and Joshi, 2001; Singh et al., 2001). Knowledge about ion composition of rainwater can provide information on the prevailing state of the atmospheric environment of that area.

The aim of this paper is to gain an initial understanding of the rainwater chemistry, to identify possible sources that contribute to its chemical composition and finally to establish a baseline data at an urban locality in peninsular India, where the proper data are not available.

2. Experimental

2.1. Demographic details of the study area

Tirupati, a holy pilgrimage town, for devotees of Lord Sri Venkateswara, is situated in southern peninsular India at an altitude of 182.9 m (13.05° N latitude; 79.05° E longitude). The town is considered to be a high profile center for education, tourism (mainly due to pilgrimage) and business with a population of 3,09,000 (2001 census) and affected by a floating population of more than 60,000 (per day). It is a semi-arid region encompassed by agricultural land, forest and by major to minor industrial activities related to chemical and pharmaceutical based units, metal based units, brick and refractory kilns and lime oxidation and pulverization. Besides these, the mobile sources also contribute a significant amount of atmospheric pollutants.

Though a continental type of climate prevails in the region with three distinct seasons, winter from January to February, summer from March to May and monsoon from June to December (S–W monsoon from June to September and N–E monsoon from October to December), it is clear that the sampling site is mostly wet during the monsoon period (July to December) and dry in the summer as well as winter months (January to June). There is seldom much rain during the early months of the year until May. The southwest monsoon sets during the last weeks of June and brings some rain until the end of September. The northeast monsoon, which breaks in the month of October, brings heavier rains till December. The local ambient temperature (which is affected by solar radiation) starts to increase in February and reaches its maximum in April–May (approx. 43 °C) after which it decreases to a minimum of 13 °C during January, while the wind speed is varied between 18.9 km h^{-1} (during summer) and 2.5 km h^{-1} (during monsoon).

2.2. Sample collection

The rainwater samples were collected on event basis with wet-only collectors, for a period of one year (July 2000–June 2001), in a university campus located almost in the central part of the study area. The site is surrounded uniformly by almost all activities that exist in the study area. A national highway lies about 2.5 km from the sampling site. Samples were collected on the roof of the physical sciences building about 6 m from ground level. The sampler was placed 1 m from the floor of the roof. Sampling was done manually on an event basis, using only wet collectors, which were fitted with a 2 l polypropylene bottle and a polypropylene funnel 22 cm in diameter. Sample collection equipment used on an event basis were washed with 10% HCl and then rinsed with double distilled water (DDW). The collectors were deployed as soon as the rain began and were withdrawn immediately after they were filled up or on stoppage of the event. A total of 35 samples had sufficient volume for chemical analysis, which represented about 52% of the total rainfall during the study period. Upon arrival at the laboratory, the pH was measured in an aliquot of the sample using pH meter (Metrohm-632). The pH meter was calibrated before and after each measurement. After pH measurements,

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