

Ca and Sr dynamics in the Indo-Gangetic plains: Different sources and mobilization processes in northwestern India

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The leachable fraction of the sediments from the Thar Desert fringe and the adjacent Ganges alluvial plains, has been studied to determine the sources and the processes responsible for the mobilization of Ca and Sr using Sr isotopes and Ca/Sr ratios. In the desert the leachable fraction of the soil/sediments is probably derived from mixing of old marine carbonates, microfossils with the sea-spray of the Arabian Sea and rainwater. Aeolian reworking of soil carbonates of this mixed origin could have provided the carbonate found at the desert fringe. The sub-humid zone of the Gangetic plains, just outside the desert fringe, has relatively higher ⁸⁷Sr/⁸⁶Sr ratios with lower Ca/Sr ratios, indicating silicate weathering as the major contributor of leachable fraction. The spatial geochemical differences could also be related to the ineffectiveness of dust transport and accumulation processes in the humid Ganga plain. The ⁸⁷Sr/⁸⁶Sr ratio of the present-day dust leachate from the polluted city of Delhi indicates that its Sr source is petroleum burnt residues.

WE studied the sediments of the Thar Desert and Ganga plains for their sources and found that the Himalayas have been playing a dominant role in supplying sediments for these two geomorphic entities¹. During the course of the Sr and Nd isotopic studies of the sediments, we leached our samples with dilute hydrochloric acid to separate carbonates and other authigenic phases from the samples^{2,3}. Besides studying silicate chemistry, we also analysed the leachates for their ⁸⁷Sr/⁸⁶Sr isotopic composition. Because Sr replaces Ca in mineral structure, it can be used as a tracer for the source of Ca⁴. When we combine this information with Ca/Sr ratios of the leachates, we find that we have a powerful tracer pair to distinguish between different sources for the leachates. Such a geochemical approach can provide important information on the sources of calcium in various surface and ecosystem studies as shown by various investigations⁵⁻⁷.

The main sources which may supply Ca to the ecosystem could be weathering products of silicates and carbonates, sea spray and rainwater⁷⁻⁹. Besides the local sources, dust derived from a long distance could also provide Ca to the

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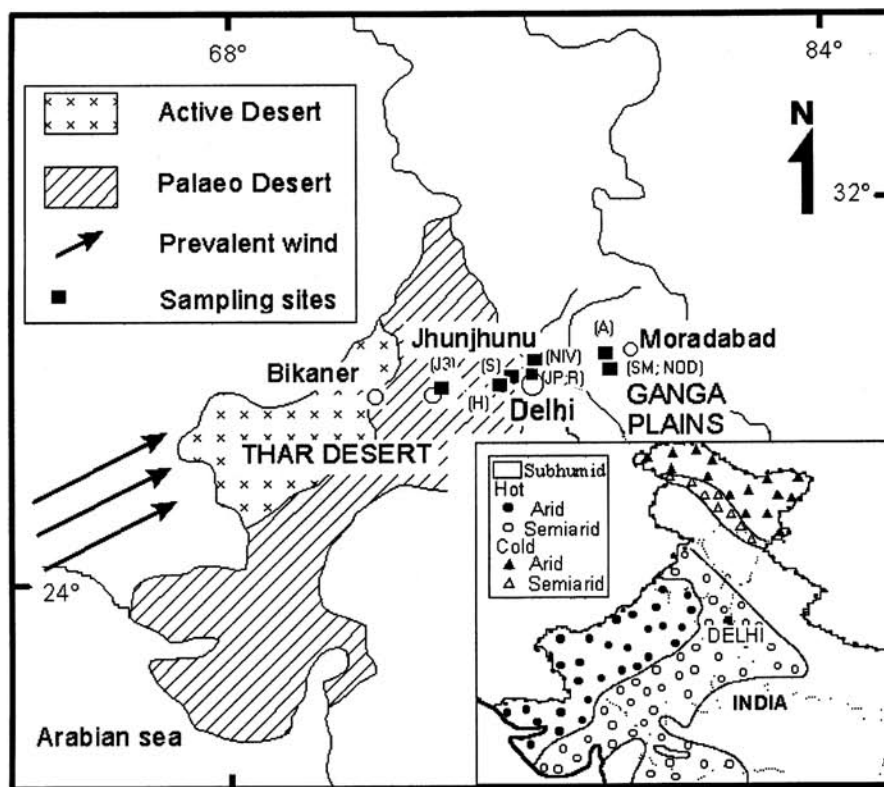


Figure 1. Sampling sites of the sediment samples for leachate study from the Thar Desert margin and the adjacent Ganga plains. Thar Desert boundaries (palaeo and active) are shown (after Pant⁴⁰). (Inset) Climate zones⁴⁰. Sampling sites: J3, Jhunjhunu dust; S, Sahibi river reworked sands. Haryana; S, Sultanpur sand dunes, Haryana; NIV, Yamuna alluvium, Baghpat, UP; JP and R, Loessic sediments and Yamuna alluvium from Delhi ridge area; A and SM, Alluvium from Moradabad area; NOD, Fe/Mn nodule in SM alluvium.

soils¹⁰. We investigated the different sources for leachable Ca fraction of sediments from the semi-arid Thar fringe and sub-humid Gangetic plains of the Indo-Gangetic plains using Sr isotopes and Ca/Sr ratios.

Sediment samples of dunes, loessic sediments and alluvium were collected from sediment profiles of thickness up to 9 m on a sampling tract of arid–semiarid–humid regions for the study of their Quaternary geology and geochemistry (Figure 1). Except for A5, SM2 and Fe/Mn nodule (sampled from sub-humid Gangetic plains), all samples belong to the semi-arid Thar Desert fringe. Samples S5, R2, JP4 and NIV2, which were analysed for their leachates, were sampled at deeper levels (below a depth of >4 m). They are stratigraphically older, in age and could be older than 10–20 Ky (S. K. Tandon and A. K. Singhvi, pers. commun.). Other sediment samples are from shallower levels of the sediment profile (less than 3 m) in the arid desert fringe. The freefall dust samples were collected in a plastic pan with glass marbles for around a month (for details of freefall sampling see Yadav and Rajamani¹¹). Samples were ground to –200 mesh size for geochemical analysis. It has been found that acetic acid or weak HCl is generally employed to dissolve the carbonate fraction in

carbonate–silicate mixtures, assuming that these acids will not leach the Sr of the silicate fraction⁷. However, in some cases the Ca and Sr adsorbed to organic matter and clays could be leached. Quade *et al.*³ found that the adsorbed fraction was similar to the soil carbonate. Although leachates represent predominantly carbonates, a small contribution of the adsorbed fraction cannot be ruled out. Therefore, we avoid use of the term soil/sediment carbonate; instead we use the term leachable fraction of the sediments in this study.

We leached³ the sediment samples with 1.3 N HCl and the leachates were analysed for Ca and Sr using ICP-OES (JY-Ultima) at JNU, New Delhi and for ⁸⁷Sr/⁸⁶Sr at GEOMAR, Germany. A separate aliquot of the sample was leached to analyse Sr isotopes. The Fe/Mn nodule was leached¹² with 3.5 N HNO₃ + H₂O₂ and the leachate was analysed for Sr isotopes. Loss on ignition (LOI) on 1000°C has been determined after drying the bulk sample at 40°C in the oven for 24 h. The precision and accuracy of analysis for Ca and Sr concentrations were monitored using USGS rock standards (BHVO, STM and RGM) as well as in-house rock standards. They were found to be better than 5 and 2% respectively. Sr separation by column chemistry has been performed using the method described elsewhere¹³.

Sr isotopic ratios were determined on a Finnigan MAT-262RPQ²⁺ Thermal Ionization Mass Spectrometer and a Triton at GEOMAR. Sr isotopes were measured in the static mode and were normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$; all errors are 2σ . Over the course of this study, NBS 987 gave $^{87}\text{Sr}/^{86}\text{Sr} = 0.710215 \pm 0.000008$ ($n = 11$). For Triton, NBS 987 gave $^{87}\text{Sr}/^{86}\text{Sr} = 0.710254 \pm 0.000003$ ($n = 35$).

The area of study covers the northeastern part of the Thar Desert margin and the adjacent Gangetic alluvial plain. The Thar Desert extends from the Sind Province of Pakistan to Gujarat, Rajasthan, Haryana, Punjab and Delhi in NW India. During the Quaternary, fluvial and aeolian processes have shaped the Thar Desert, while fluvial processes of Himalayan rivers created the Gangetic plains, supplying sediments mainly from the Higher Himalayas^{1,14}. Aeolian sand ridges of local origin are also reported from the Gangetic plains, which are locally known as Bhur sands¹⁵. The basement of the Thar Desert is mainly composed of peneplained surfaces of Precambrian rocks in the east and gradually thickening deposits of Mesozoic and Tertiary age westward¹⁶. The Aravalli ranges, containing the Aravalli and Delhi Supergroups, mark the eastern boundary of the Thar Desert, which also contains Quaternary loessic deposits in its depressions on top of the hills¹⁷. Calcretes are ubiquitously present in the soils and sediments of the Thar Desert¹⁸.

The hot arid to semi-arid Thar region experiences hot summers, with a mean minimum and maximum temperature of 26 and 42°C respectively during April to June. The winter mean minimum and maximum temperatures are 7 and 22°C respectively between October and March. Rainfall shows an increasing trend from west to east and from south to north. The desert region receives an annual rainfall of 100–300 mm. The region around Delhi, at the northeastern fringe of the desert, receives an average annual rainfall of about ~700 mm, whereas the sub-humid Moradabad area of the Gangetic plains receives ~1000 mm of rainfall. Most of the rainfall (80%) falls in the summer monsoon period from July to September¹⁹. In this region, the prevailing winds during summer are southwesterly and are largely controlled by the seasonal heat low in Sind (Pakistan) and western Rajasthan (India). These winds carry large amounts of dust. Presently, the maximum wind activity and development of new aeolian bedforms take place mostly in the western part of the desert, because the efficacy of aeolian processes increases with decreasing rainfall from east to west²⁰. The average number of days of dust storm per year also increases from east to west¹⁹. The average soil loss in the Thar Desert is estimated to vary between 30 and 60 kg/m² per day during April to June²⁰. During the winters, the northwesterly light winds blow through the Delhi and Moradabad areas, which are not effective in sediment transport.

In comparison with the Thar region of Rajasthan and Haryana, the Delhi region has gone through heavy industrial growth and therefore is now ranked as one of the world's

most polluted urbanized city. Vehicular emissions, thermal power plants and other industrial units are the major polluting sources²².

Weathered material of crustal rocks with high Rb content provides higher $^{87}\text{Sr}/^{86}\text{Sr}$ bearing run-off to the ocean than weathering of basalts or hydrothermal input within the ocean. Therefore, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the leachable fraction of the sediments can be used to distinguish between material resulting from the weathering of crustal and oceanic components (e. g. marine carbonates, sea spray and rain-water). Dust, which incorporates marine carbonate grains and sea spray, is an important carrier of these components to the continent¹⁰. To demarcate the dust and desert encroachment on land in the study area, we plotted the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios on the sampling traverse, from the desert margin to the Gangetic floodplains (this is the direction of the prevalent southwesterly winds), which is shown in Figure 2.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the leachates in the study area abruptly increase from the desert margin to the Gangetic plains of the Moradabad area (Figure 2). The Sr isotopic ratios of soil leachate of sand dunes and loessic sediments vary between 0.7110 and 0.7166 (Table 1). The leachates of Yamuna alluvium carry isotopic ratios of 0.7188 (NIV6) and 0.7154 (R2) despite the host alluvial silicate fractions having values of 0.7428 and 0.7670 respectively. In the Gangetic alluvial plains, isotopic ratios of the leachate are 0.7337 (SM2) and 0.7496 (A5), consistent with the silicate fraction values of 0.7739 and 0.7893 respectively. The free-fall dust leachate sampled at Jhunjhunu (J3), which lies far away and upwind of Delhi, has a $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.7117 (silicate fraction: 0.7275). Delhi free-fall dust (D99) also has low radiogenic Sr isotope composition (0.7088) despite its radiogenic silicate fraction composi-

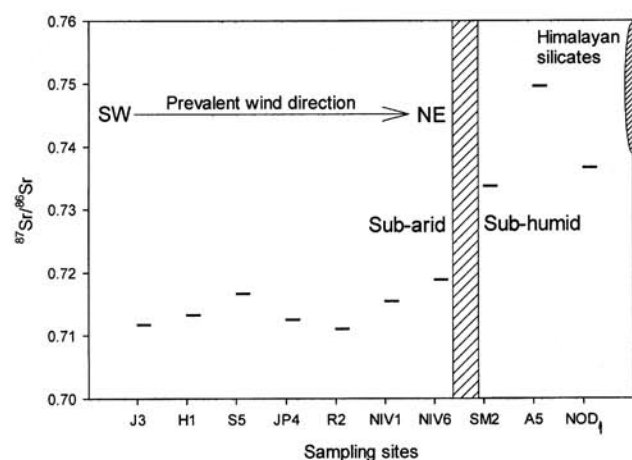


Figure 2. Spatial variation in $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the leachates in the northeastern margin of Thar Desert and adjacent Ganga plains. Sampling points follow nearly the prevalent wind direction during summer. The abrupt increase in $^{87}\text{Sr}/^{86}\text{Sr}$, coinciding with the climatic variation, points to a connection with the humid conditions in the Ganga plains. The $^{87}\text{Sr}/^{86}\text{Sr}$ value of the leachate of Fe/Mn nodule (NOD) is similar to that of the leachates of sediments in that region, indicating similar silicate sources for Sr and Ca.

Table 1. Ca/Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of leachates (L), silicate fraction (S) and loss on ignition (LOI) values of bulk sediments of Thar Desert and Ganga plains

| Sample | Ca/Sr | $^{87}\text{Sr}/^{86}\text{Sr}$ | 2σ | $^{87}\text{Sr}/^{86}\text{Sr}(\text{S})$ | 2σ | LOI (%) |
|--------------|--------|---------------------------------|-----------|---|-----------|---------|
| NIV1 | 940.3 | 0.71542 | 0.000008 | 0.72895 | 0.000010 | 6.79 |
| NIV6 | 456.03 | 0.71884 | 0.000008 | 0.74276 | 0.000009 | 4.79 |
| R2 | 1549.9 | 0.71101 | 0.000008 | 0.76732 | 0.000010 | 9.58 |
| JP4 | 824.9 | 0.71250 | 0.000009 | 0.72942 | 0.000007 | 7.37 |
| H1 | 627.3 | 0.71323 | 0.000008 | 0.72895 | 0.000010 | 8.77 |
| S5 | 331.1 | 0.71661 | 0.000007 | 0.73323 | 0.000009 | 3.22 |
| J3 | 574.5 | 0.71170 | 0.000003 | 0.72753 | 0.000004 | 6.98 |
| SM2 | 409.8 | 0.73368 | 0.000010 | 0.77388 | 0.000010 | 7.44 |
| A5 | 300.9 | 0.74956 | 0.000008 | 0.78932 | 0.000005 | 6.53 |
| Fe/Mn nodule | | 0.73659 | 0.000008 | | | |
| D99 | 534.8 | 0.70892 | 0.000009 | 0.73387 | 0.000009 | 17.9 |

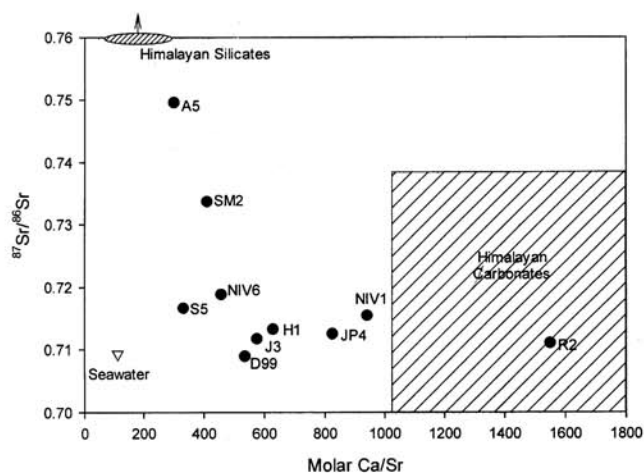


Figure 3. Three end-members, sea water⁷, Himalayan carbonates and silicates²⁷ are plotted along the leachates of sediments for their $^{87}\text{Sr}/^{86}\text{Sr}$ and molar Ca/Sr ratios. All the marine carbonates also plot within the Himalayan carbonate field, including those precipitating now. If the leachate from the Ganga plain had predominantly silicate sources because of the weathering of silicate minerals in humid conditions, those of the desert margin have their origin from soil carbonates/calcretes. These soil carbonates/calcretes would have formed from the marine carbonates and sea spray and rainwater in the upwind exposures. Mixing of these two components in different proportions explains the spread of the datapoints along a mixing line between sea water and marine carbonates.

tion (0.7388). This clearly shows that there is an increase in radiogenic Sr in the soil leachate beyond the semi-arid fringe area of the Thar Desert. As seen by the comparison of the host silicate fraction and its leachates in the fringe area, the leachate chemistry is not affected by the chemistry of the host silicate sediments. The increase in radiogenic component in the leachates of samples from the sub-humid Ganga plains, could be either explained by distinct sources for the leachate fraction as carbonates or the influence on the leachate components from silicate weathering. The low molar Ca/Sr ratios for the leachate indicates that the contribution of carbonates to Ca is negligible relative to

the silicate sources, as discussed later. Alternatively, the low radiogenic components are not available to the Ganga plains by aeolian processes. The increase in humidity and vegetation at the semi-arid to sub-humid transition zone²³ and the swing of the SW winds from NE to E and therefore the decrease in wind velocity causes a trapping effect on dust-containing low radiogenic mobile phases of Sr. It is interesting to find that the change in leachate geochemistry coincides, with the spatial change in the climatic regime.

Surficial processes do not fractionate Sr isotopes; therefore, isotopic signatures can be used to distinguish between various sources (silicate vs non-silicate) for Ca. However, discrimination between various low radiogenic sources (sea spray, rainwater, carbonates) is difficult using only isotopic signatures. If Ca/Sr ratios are also taken into account, distinction among various non-silicate sources is also possible. Biogeochemical processes of rock-weathering, nutrient uptake and redistribution through different trophic levels results in different Ca/Sr ratios, because Ca is preferentially taken up by the biological systems^{4,24}. This causes a large variation in Ca/Sr ratios of biogenic and non-biogenic materials. This has been successfully used to understand the source and path of Ca in the ecosystem studies on land^{5,25} and to trace the sources for Ca in the rainwater and soil carbonates^{7,26}. Ca/Sr ratios have also been used in the study of stream waters of the Himalayas to suggest carbonate sources against the previously assumed silicate sources for the radiogenic Sr²⁷. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are plotted against molar Ca/Sr ratios for the leachates in Figure 3. The data plot along a curve nearly parallel to the axes, possibly towards the two end-members, silicates and carbonates (Figure 3). The turning point of this curve could be a third component (sea water), which is also plotted for reference.

One of the end-members is identified as a silicate source for the leachates having high $^{87}\text{Sr}/^{86}\text{Sr}$ and low Ca/Sr ratios. The Himalayan silicate rocks and Ganga plain sediments derived from the Himalayas, also plotted for reference, corroborate the inference that the weathering of these

sediments may provide radiogenic labile phases. The leachate of the Fe/Mn-nodule, present in the SM2 sediments, also has a $^{87}\text{Sr}/^{86}\text{Sr}$ value similar to the silicate host (SM2) leachate. Such Fe/Mn oxide nodules have been used as proxy to the weathering solution that has deposited the nodules²⁸. Therefore, we suggest that Sr and Ca present in the leachate of the nodule are mainly derived by chemical weathering of silicates under sub-humid conditions of this region. The SM2 (sandy mud) leachate shows relatively less radiogenic Sr composition and higher Ca/Sr values with respect to A5 (mud). It is possible that the detrital grains of marine carbonate rocks of the Himalayas present in the alluvium could bring down the $^{87}\text{Sr}/^{86}\text{Sr}$ values and increase the Ca/Sr ratio²⁷.

Marine carbonates with low $^{87}\text{Sr}/^{86}\text{Sr}$ and high Ca/Sr ratios could be another end-member component. The low radiogenic Sr isotope value of R2, with the highest Ca/Sr ratio among all the samples studied, indicates that the contribution of marine carbonate was larger in R2. We also observe high Ca/Sr values for JP4 and NIV1, which are lower than those of R2, but much higher than those expected from silicate weathering or sea water. This could be due to the mixing with calcium of mixed origin (discussed below) from upper horizons by water. Marine carbonates are characterized by low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (<0.7092) and high Ca/Sr ratios (>1100)⁷ (see caption, Figure 3). Limestones and dolomite of the Aravalli and Delhi Supergroups as well as Mesozoic and Tertiary sediments occur in the Rajasthan and Gujarat parts of the Thar Desert. The miliolite foraminifera deposits occur in Saurashtra, Gujarat²⁹. Wasson *et al.*³⁰ suggested that these forams could have been the source for microfossils occurring in the dune-sands of the Thar. Gaudie and Sperling³¹ have found foraminifera in the Thar dune fields up to 800 km inland from the Arabian Sea. It is possible that they could have been transported in suspension by the prevailing winds. We suggest here that the smaller forams could have been transported directly in suspension, but bigger forams could break during saltation and attrition. These broken forams could have been weathered under favourable conditions and reprecipitated as finer secondary phases. The finer carbonate grains can be further transported easily for long distances³². Palaeo-karstic topographic features in the Jaisalmer basin¹⁶ and Gujarat²⁹ indicate that the carbonate sediments have been supplied to the desert by dissolution, precipitation and erosion throughout the Quaternary under favourable conditions.

The low $^{87}\text{Sr}/^{86}\text{Sr}$ and low Ca/Sr leachate values can be attributed to the third component, which could be the sea salt and rainwater derived from sea water. Among the possible airborne sources, sea spray and rainwater cannot be distinguished on the basis of their Sr isotopes because both of them had the same origin³³. However, the rainwater chemistry can get changed during reaction with tropospheric dust. The average rainwater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the downwind direction of SW monsoon in Nepal and Bangladesh have been estimated as³⁴ as 0.714. The elevated $^{87}\text{Sr}/^{86}\text{Sr}$

values of rainwater than sea water suggest that this has been equilibrated with fine-grained tropospheric dust that consists of material of the desert and Indo-Gangetic plains. High amounts of crustal-derived Ca were also inferred in rainwaters of northern India and Nepal³⁴⁻³⁷. It has been found that the coastal rainwater also contains Ca, but less than that in inland sampling sites³⁴. The saline depressions in the Gujarat Rann of the Kutch region are integral part of the Thar landforms. Their origin is thought to be linked with the recession of the sea and aeolian transportation of salt particles³⁸. Thick horizons of illuviated caliche deposit and/or gypsum occur within the older alluvium in the desert³⁸. From the chemical composition of the different components discussed above, we infer that the mixture of rainwater and sea spray with the marine carbonates could give rise to the required intermediate range of chemistry found in this study. We suggest that soil carbonates/calcretes going through processes of congruent dissolution and precipitation, and aeolian transportation are the main sources of the leachable fraction in the Thar Desert and its margin. In the sub-humid regions of the Ganga plains, Ca in the leachate is mainly derived from silicate weathering.

The leachable from free-fall dust sampled during May–June 1999 in Delhi (D99), interestingly, has a $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.7089, which is lower than that of the present-day sea water. However, the free-fall dust from Jhunjhunu (J3; sampled during May–June, 2000) has given a leachate $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.7117. No significant differences have been noticed in their Ca/Sr ratios. Even the present-day sea water ($^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.7092) and rainwater composition (0.714) in this region have higher values than dust (D99) leachate, indicating that a low Sr isotope source must have contributed significantly to this leachate. The free-fall dust sample in Delhi is blackish in colour and shows the highest loss of ignition (17.9%) in comparison to other sediments and free-fall dust J3. We may suspect Phanerozoic carbonate detritus as the possible source, but the high LOI and intermediate Ca/Sr values point to volatile phases and carbonaceous material present in this sample. It has been reported that the production of black carbon due to diesel combustion in road transport is highest for Delhi, Kolkata, Chennai and Mumbai²². We suggest that the residue of fossil fuel burning is likely the low radiogenic isotopic source in the Delhi region. The $^{87}\text{Sr}/^{86}\text{Sr}$ value for the Delhi dust leachate corresponds to the Mesozoic sea water composition³⁹ when petroleum was formed.

Sr isotope chemistry in the leachate component of surface sediments changes abruptly across the boundary between the semi-arid desert margin and sub-humid Ganga plains in northwestern India. Secondary carbonates present in the Thar Desert as soil carbonate/calcrete, which formed by congruent weathering processes incorporating a diversity of sources external to the sediments, provided the leachable fraction in the sediments of the Thar fringe. The Sr isotope chemistry of the free-fall dust in Delhi points to the environmental pollution pertaining to the fossil-

fuel burning in this region, as also reported by other independent studies.

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ACKNOWLEDGEMENTS. We thank the Department of Science and Technology, New Delhi and DAAD, Germany for financial support during this work. J. K. T. thanks Prof. S. K. Tandon for discussions and suggestions during fieldwork in Ganges plain and Ana Kolevica for her assistance during geochemical lab work at Geomar. Unknown referees are acknowledged for their reviews.

Received 16 February 2004; revised accepted 1 July 2004