ABSTRACT

The role of silicate and carbonate weathering in contributing to the major cations and Sr isotope geochemistry of the headwaters of the Ganga-Ghaghara-Indus system is evaluated by making ***new*** chemical and isotopic measurements of Precambrian carbonates from the Lesser Himalaya and from the available data on silicates. Samples of Precambrian carbonate outcrops collected across the Lesser Himalaya have been analysed for their mineralogy, chemical composition and isotope ratios of Sr, O and C. Their Sr concentrations range from 20 to 363 ppm with $^{87}Sr/^{86}Sr$ 0.7064 to 0.8935, $\delta^{18}O_{PDB}$ -1.4 to -12.8 ‰ and Mn 11 to 2036 ppm. The petrography of the samples, their low Sr concentrations and wide range of $\delta^{18}O$ values are all suggestive of their postdepositional alteration.

Comparison of the $^{87}Sr/^{86}Sr$ and Sr/Ca ratios among the carbonates and silicates from the Lesser Himalaya and the source waters of the Ganga, Ghaghara and the Indus shows that the values for the source waters overlap with those of the silicates but are much higher than those in carbonates. An upper limit of carbonate Sr in the various source waters is calculated to be between 6%-44% (mean 18%), assuming that *all* the Ca in the rivers is of carbonate origin. The results show that on the average, weathering of the Precambrian carbonates is unlikely to be a major contributor to the highly radiogenic Sr isotope composition of these source waters; however, they can be a dominant supplier of radiogenic Sr to some rivers on a regional scale.

The silicate Sr component in these headwaters range from 2% to 100% with a mean of 40%. The calculation shows that only in few of the headwaters Sr balance would be achieved based on a two component, silicates and Precambrian carbonates, mixing. This indicates the need of a third end member. Tibetan/Tethyan carbonates, evaporites, phophorites are the possible end member.

The second part of the thesis deals with the study of Re-Os isotope systematics in black shales from the Himalaya for determining the chronology and evaluating their potential to contribute to the steady increase in $^{187}Os/^{186}Os$ of the oceans since past ~25 Ma. Towards this, chemical procedures and Negative Thermal Ion Mass Spectrometry techniques have been established for the precise measurements of Re-Os concentrations in environmental samples and the Os isotope composition in them for the first time in our laboratory. The precision of $^{187}Os/^{186}Os$ ratios and Os concentration determination are

better than 1% ($\pm 2\sigma_\mu$) and 2-3% ($\pm 2\sigma$) respectively in a few tens to few hundreds of picograms of Os.

Re and Os abundances and Os isotope composition have been measured in a number of black shales sampled from outcrops and two underground phosphorite mines, Maldeota and Durmala, in the Lesser Himalaya. The black shales from the Maldeota and Durmala mines, collected ~15 m above the Krol-Tal (Pc-C) boundary, yield ^{187}Re-^{187}Os isochron ages of 535 ± 11 Ma consistent with stratigraphy and those assigned for the Pc-C boundary at various other locations. The age of these samples from the outer belt seems to be a few hundred millions of years younger than the ^{187}Re-^{187}Os age of 839 ± 138 Ma for black shales from the inner belt.

The role of weathering of these black shales in contributing to the Os isotope evolution of seawater over the past ~25 Ma was assessed using a simple budget model. If the ^{187}Os/^{186}Os of global rivers (including those draining the Himalaya) is kept constant at today's value of 11.0 (based on available data for rivers) then the Os flux required from HTP rivers to reproduce the oceanic ^{187}Os/^{186}Os would have to increase from 0.51 moles y^{-1} at 16 Ma ago to 830 moles y^{-1} at present. The present day flux would correspond to Os concentration of ~40 pg ℓ^{-1} in HTP rivers. This concentration is sustainable by weathering of black shales from the region, though it is about a factor of ~2 more than the highest Os concentration reported in rivers.

Similar calculations, assuming global ^{186}Os flux to be constant over the past 16 Ma at 15.8 moles y^{-1} (calculated from available data for rivers), show that ^{187}Os/^{186}Os in HTP rivers have to increase from 7.7 at 16 Ma ago to 40.5 at present to reconstruct the observed seawater ^{187}Os/^{186}Os variations. The present day ratio of 40.5 is within the range measured in black shales from the Himalaya, but it is higher than the mean of ~23 in the Maldeota and Durmala black shales and the reported value of ~16.2 in the leachable fraction of the Ganga river sediments in the plains. The measured Os concentrations and ^{187}Os/^{186}Os in black shales from the Lesser Himalaya though can meet the model requirements, the demands on them can be more easily accommodated if the ^{187}Os/^{186}Os of HTP rivers is taken as 16.2 and that in all the other rivers as 11.0.

This study has brought the need (i) to look for other possible source(s) to balance the Sr budget in the head waters and (ii) more detailed study of black shales from the inner and outer belts to determine their chronology and inter-relation.