Uranium isotopes in surface waters from the Mount Cameroon: tracing water sources or basalt weathering?

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TIMS analysis of the ^{234}U/^{238}U radioactive disequilibrium in river and spring waters of the Mt Cameroon volcano has been undertaken (1) to constrain the parameters controlling the supply of U isotopes to freshwaters, and (2) to evaluate the potential interest of the U activity ratio in dissolved loads of river waters as a tracer of rock weathering. The Mount-Cameroon volcano has the advantage to provide a wide but nearly monolithological geographical area, which consists essentially of alkali basalts and to present an intense tropical weathering at different evolution stages depending on the age of the successive volcanic flows.

Thirty six river and spring waters draining and flowing over the Mount Cameroon volcano were collected during two campaigns in December 1996 and May 1997. These samples were filtered on site at 0.1μm, and splited in different aliquots used for analysis of the major and trace elements, and the oxygen, Sr and U isotope compositions. In twelve sampling sites, about 40L of water were also treated by ultrafiltration techniques using Sartorius ultra-filtration cartridges with successive cut-off at 0.22μm, 100kDalton (~0.02μm) and 5kDalton (~0.002μm). For each ultrafiltration experiment, the ultrafiltrates and the "colloid concentrates" were collected and treated for chemical and isotope analysis.

Results obtained for the (^{234}U/^{238}U) ratio and the U, Rb and Sr concentrations are presented here. The (^{234}U/^{238}U) isotope ratios were measured with a precision of 0.5% (2σ error) on a VG sector mass spectrometer. The concentrations of U, Rb and Sr were determined by an ICP-MS Fisons VG-PlasmaQuad with a detection limit of 0.01mg/L.

Ultrafiltration experiments

The preliminary results obtained by ultrafiltration techniques reveal that the U concentrations in the filtrates may strongly decrease when the filtration size decreases. For waters enriched in organic matter, i.e. with a Total Organic Carbon (TOC) above 2mg/L, the U concentration of the "dissolved load" decreases by about a factor two when the filtration size decreases from 0.22μm to 5kD. This U decrease is associated with a significant decrease of the TOC in the filtrate, which certainly indicates that the transport of U is influenced by the presence of organic colloids for these samples. A decrease of the U concentration upon successive filtrations at decreasing pore size, can also be observed for samples less enriched in organic matter, i.e. with TOC below 1mg/L. In these samples, the decrease of the U concentrations is less pronounced than for samples enriched in organic matter, and it is never associated with a decrease of the TOC. In this case, the occurence of inorganic colloids, i.e. oxides and hydroxides of Fe and Al, is *a priori* required for explaining the decrease of the U concentrations in the filtrates. Contrasting with the U concentrations, those of Sr and Rb do not vary or only slightly when the size of filtration decreases. Overall, these results confirm the importance of organic but also of inorganic colloids in the transport of U in freshwaters (Anderson *et al.* 1994; Porcelli *et al.* 1997), and they demonstrate that the U-Sr and U-Rb soluble elements are quite significantly fractionated by the occurence of colloids in the waters. Because of this chemical fractionation, the determination of the U/Rb or U/Sr ratios in dissolved loads of river waters could be difficult especially when the rivers are rich in organic matters.
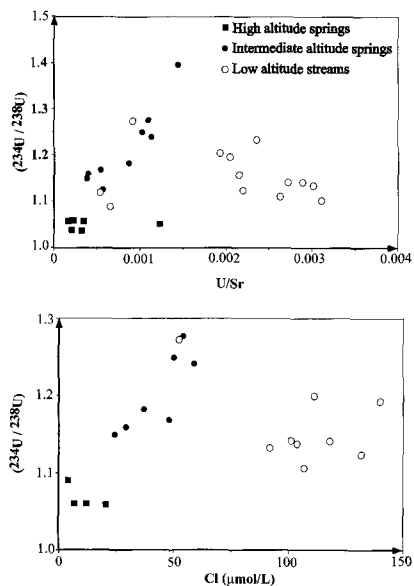
FIG. 1: Variations of $(^{234}U/^{238}U)$ activity ratios against U/Sr ratios (*top*) and Cl concentrations (*bottom*) in waters from the Mount-Cameroon.

For the ultrafiltrated samples that had their U isotopic ratio analysed, no significant differences were observed between the $(^{234}U/^{238}U)$ activity ratios of the different filtrate and concentrate fractions recovered from a single sample. This means in turn that $(^{234}U/^{238}U)$ activity ratios in dissolved loads of a fresh water, can be determined from any filtrate fractions.This result also indicates that the U of the dissolved and colloidal loads of the samples are in isotopic equilibrium, implying either an in situ precipitation of colloids from chemical species dissolved in the water, or/and a complete exchange of U between the colloids and the water, if colloids are thought to have derived from mechanical erosion of soils and weathering profiles.

Geographical variations

At the scale of the Mount Cameroon volcano, the data obtained on the 0.1 μm-filtered waters outline important variations of both the $(^{234}U/^{238}U)$ activity ratios and U concentrations, from 1.04 to 1.4 for the $(^{234}U/^{238}U)$ ratios, and from 0.3ppb to the detection limits of the ICP-MS techniques for the U concentrations. On the basis of the available results, the analysed samples can be subdivided into 3 groups. One includes the high altitude springs (2400−1000m high) which have low ^{234}U-^{238}U

disequilibrium, between 1.04 and 1.08. One is formed by the sources of intermediate altitude (900−200m high) which are marked by higher $(^{234}U/^{238}U)$ activity ratios, from 1.1 to 1.4. These waters together with those from former sources define a reasonably good positive correlation in both the $(^{234}U/^{238}U)$ vs. U/Sr diagram and the $(^{234}U/^{238}U)$ vs. Cl diagram (Fig 1a-b). The remaining water samples from lower altitude (<100m high), constitute the third group of data, defining a very scattered domain when plotted in the two former diagrams. The Cl concentrations of these waters reach higher values than in waters of the two other groups.

The correlations of figure 1a-b, obtained for the springs of high and intermediate altitude, could be interpreted in terms of an increase/intensification of the water-rock interactions. The Longer the water rock interactions, the higher are the dissolved loads and the $(^{234}U/^{238}U)$ activity ratios of the waters. A possibility could be that the increase of the disequilibria in these waters reveal a sequential weathering of the lavas : (1) dissolution of the basaltic glass which only moderately fractionates the ^{234}U-^{238}U isotopes, then (2) a subsequent weathering of the lava minerals with a much stronger ^{234}U-^{238}U fractionation. In this interpretation the $^{234}U/^{238}U$ disequilibria in waters would average the degree of the basalt alteration of the watershed of the springs. However, this explanation is not univocal and the correlations in fig 1a-b could be also interpreted in terms of mixings between different aquifers with different Cl, U/Sr and $^{234}U/^{238}U$ signatures. For the waters of the third group, the scatter of the data in the $(^{234}U/^{238}U)$ vs. U/Sr diagram can be explained, at least for some samples, by an U/Sr fractionation due to the presence of colloids. The data points of these samples, move towards the trend defined by the waters of the groups 1 and 2, when the 5kD filtrates are used instead of the 0.1μm filtrates. These waters have also high Cl concentrations. A simple explanation to conciliate a significant presence of colloids with a high concentration of Cl in the waters of low altitudes would be to involve an input of water and particles from surficial horizons of the soils and the weathered profiles.

References

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