

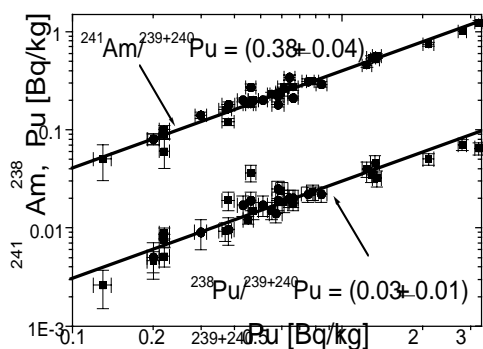
## Actinide isotope analysis for determining different nuclear fallout components in soils

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The Swiss immission surveillance program around nuclear installation includes analyses of soil samples for various cosmic, terrestrial and particularly anthropogenic radioisotopes such as <sup>7</sup>Be, <sup>14</sup>C, <sup>40</sup>K, <sup>89</sup>Sr, <sup>90</sup>Sr, <sup>134</sup>Cs, <sup>137</sup>Cs, U- and Th-series decay products as well as the actinides <sup>238</sup>Pu, <sup>239+240</sup>Pu, <sup>241</sup>Pu, <sup>241</sup>Am, <sup>244</sup>Cm. The isotopes of Pu and Am can be used to determine the fraction of hot particle fallout from different nuclear accidents, e.g. derived from surface atomic bomb tests in the 60ies or the Chernobyl accident, that occurred 16 years ago. Figure 1 shows that there is a strong correlation between <sup>238</sup>Pu, <sup>241</sup>Am and <sup>239+240</sup>Pu. The <sup>238</sup>Pu/<sup>239+240</sup>Pu ratio of  $0.03 \pm 0.01$  calculated via regression analysis agrees well with literature values obtained from various other sites in the northern hemisphere. For the <sup>241</sup>Am/<sup>239+240</sup>Pu ratio a value of  $0.38 \pm 0.04$  was obtained. This result is within uncertainty identical to the present day <sup>241</sup>Am/<sup>239+240</sup>Pu ratio of 0.36 calculated via <sup>241</sup>Pu/<sup>241</sup>Am progenitor/progeny relationship and additionally considering that the initial <sup>241</sup>Pu/<sup>239+240</sup>Pu ratio resulting from nuclear weapon testing in the 60ies was about 13. This result shows clearly that <sup>241</sup>Am measured in the samples analysed here can be well explained via support from decaying <sup>241</sup>Pu initially released during the surface A-bomb testing period meaning that contribution of heavy particles from the Chernobyl accident are not significant in Central Europe.

Figure 1: Correlation diagram with <sup>238</sup>Pu vs. <sup>239+240</sup>Pu and <sup>241</sup>Am vs. <sup>239+240</sup>Pu obtained on Swiss soil samples from various locations.



## Hydrogen-isotope geochemistry of the CM chondrites

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Hydration of initially anhydrous silicates and oxides is a common and significant early solar system processes recorded by the carbonaceous chondrites. We present new data constraining the hydrogen isotope geochemistry of the CM chondrites, which are characterised by a wide range in degree of hydration. Our study makes use of a recently developed method for analysis of D/H ratios of  $10^{-9}$  mol quantities of water released by heating small (10's of microgram) solid samples.

Data for four CM chondrites (Murchison, Mighei, Murray, Cold Bokkeveld) document a trend of monotonically decreasing matrix D/H with increasing  $[\delta^{18}\text{O}_{\text{matrix}} - \delta^{18}\text{O}_{\text{whole-rock}}]$ —a measure of the extent of aqueous alteration. The upper and lower limits in matrix D/H are 0.000148 (Murchison) and 0.000125 (Cold Bokkeveld). Mighei and Murray yielded intermediate D/H ratios of 0.000143 and 0.000137, respectively. A nearly anhydrous CM chondrite (L85311) was also analysed but was not sufficiently well resolved from background to yield a reliable D/H ratio. The D/H ratios of individual altered chondrules from both Cold Bokkeveld (highly hydrated) and Murchison (modestly hydrated) are equal to one another and to that for Cold Bokkeveld matrix; i.e., altered chondrules from both H-rich and H-poor samples have D/H ratios equal to that of the most extensively hydrated matrix. Replicate analyses suggest matrix is isotopically homogeneous within any one sample.

These data are not easily reconciled with either simple (0-dimensional) or chromatographic (1-dimensional) models of reaction between water and initially H-poor rock, in part because such models generally predict hyperbolic relationships between H-isotope composition and extent of aqueous alteration rather than the linear relationship we observe. Our results could be explained if the matrices of CM chondrites contained substantial amounts of hydrogen *before* they underwent the episode of aqueous alteration responsible for their first-order geochemical and mineralogical variations, or if they are mechanical mixtures of two components that did not share the same history of hydration and/or dehydration. Finally, our data suggest the D/H ratio of water that infiltrated the CM chondrites was ca. 0.000132. This value is interpretable as a constraint on the temperature at which infiltrating water last equilibrated with the dominant H reservoir of nebular  $\text{H}_2$ ; this temperature was  $282 \pm 50$  K if the D content of water was influenced only by  $\text{H}_2\text{O}(v) - \text{H}_2(v)$  equilibrium and  $390^{+110}_{-90}$  K if water was isotopically fractionated by formation of ice at a 'nebular snow line' before incorporation into the CM parent body. The principle source of uncertainty in these estimates is the protosolar D/H ratio (ca.  $0.000034 \pm 10$ ).