Plutonium and uranium isotope ratio measurement by MC-ICP-MS

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Multi-collector plasma source mass spectrometry has revolutionised the measurement of plutonium and uranium isotopes in geological and environmental studies. Chief improvements are the increase in ionisation efficiency and collection, and the predictability of instrumental fractionation between measurements.

We present high precision analytical protocols for uranium isotope ratios using MC-ICP-MS and a ^{233}U-^{236}U double spike to correct for instrumental fractionation. This is compared with sample-standard alternation correction on the same instrument and external fractionation corrections using thermal ionisation mass spectrometry. For samples with a natural uranium isotopic ratio $^{238}U/^{235}U$ =137.88, and with >20 ng U, the reproducibility is 0.06% 2sd using the double spike technique. This represents a factor of three improvement over traditional TIMS measurements involving a separate estimation of fractionation. If total U available is 1ng - 50pg then Faraday-Daly combinations are used in conjunction with external fractionation and gain calibrations. Such inter-detector analyses achieve a reproducibility of 0.15% - 0.4%, and enable low-concentration studies of depleted uranium contamination.

The acquisition of high-resolution plutonium isotope data is a fundamental benefit of MC-ICP-MS. Overall efficiencies of the measurement approach 1% for Pu, forming an alternative for low-level Pu analysis to accelerator mass spectrometry. We present both single and multi-ion counting techniques, which enable measurement of 1pg to 10fg Pu with reproducibilities of 0.15% - 0.5%.

Combining these U and Pu analytical techniques, we have measured ice core and vegetation samples spanning the last 50 years. The result is a reference curve for the changes in Pu and U environmental fallout, to serve as a baseline for local contamination studies.

Emission of Soil Dust Aerosol: Anthropogenic Contribution and Future Changes

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Introduction

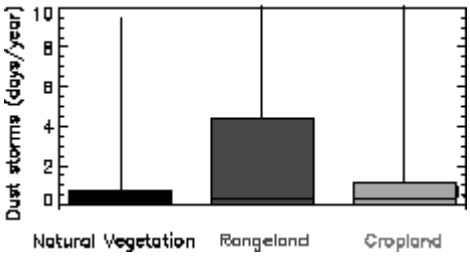
Soil dust aerosol is an important factor in the climate system, impacting on the radiation balance, atmospheric chemistry and biogeochemical cycles. Anthropogenic disturbance of soil surfaces has been estimated to contribute as much as 50% to the modern global dust load in the atmosphere (IPCC 2001), but this estimate is highly uncertain. We need to quantify this contribution more accurately in order to understand the historical dust record and estimate future changes in dust emissions.

Method

We compare a global compilation of dust storm frequencies, based on data from more than 2000 stations, to vegetation cover and the distribution of cultivated and rangeland areas. The global emission of natural and anthropogenic dust is computed with a dust source model which explicitly includes the contribution of preferential source areas and vegetation phenology.

Results and Discussion

Figure 1: Comparison of global dust storm frequencies in areas of natural vegetation, croplands and rangelands, the bars indicate the 25th to 75th percentiles of the data



We find a slightly higher dust storm frequencies in rangeland and cropland areas than in undisturbed regions. The increase is small, and, according to our model anthropogenically-disturbed soils contribute only 5 to 15% to the modern global dust emission. Using these results, we estimate the role of future changes in anthropogenic dust loading in offsetting natural changes in dust emissions under a global warming scenario.

References

IPCC (2001), Climate Change 2001, edited by J. T. Houghton et al., Cambridge University Press, New York.