Atmospheric Forcing of Sulphate in Speleothem Carbonate

Sulphur in the atmosphere is an important agent of climatic forcing and is known to originate from a variety of sources including combustion of fossil fuels, sea salt aerosol, biological emissions and volcanic eruptions. Just as ice core records from Greenland are suggested to reflect changing atmospheric loads of sulphur, this project aims to utilise speleothem carbonate as an archive of atmospheric sulphur concentration over a much more extensive geographical region.

The degree to which speleothems capture regional variations in atmospheric sulphur loading will depend upon the extent of ecosystem modification. Field sites are currently in use in Italy, Austria, Ireland, Scotland and Ethiopia, where natural environmental isotopes of sulphate-S and sulphate-O are being used to improve our understanding of sulphur cycling in the karst ecosystem and to define the method of sulphate incorporation into speleothem calcite.

The basis for this project arose from a study by Frisia et al. (Earth and Planetary Science Letters, 2005) in which it was demonstrated that at an Alpine cave site in North East Italy subject to moderate levels of pollution, host stalagmites recorded a rise in sulphur concentration from the late nineteenth to late twentieth century. This was thought to reflect increasing anthropogenic emissions and sulphur isotope ratios are now being used to test this assertion. The first inter-comparison between tree rings and speleothems is also being undertaken at this site enabling an assessment of whether Alpine trees or speleothems provide a more direct record of sulphur variability.

In coastal locations, much of the sulphate is sourced from seasalt aerosol as opposed to emissions from anthropogenic pollution. As variations in the winter North Atlantic Oscillation (NAO) are associated with changes in oceanic circulation patterns, these in turn affect the transport of sea salt aerosol onto land and therefore the availability of sulphate for incorporation into speleothem calcite. At Crag Cave, Ireland, the presence of isotopically light sulphur in the surrounding bedrock contrasts markedly with the heavier seasalt aerosol to produce two distinctive sources of sulphur. The relative contributions of these may possibly be used as a proxy for Holocene circulation patterns and will complement work being undertaken in NERC's RAPID programme.

At the laboratory scale, experiments are being undertaken to identify the controls upon sulphate incorporation into speleothem calcite. Growth of experimental crystals under different environmental conditions in association with field studies of biogeochemical sulphur cycling should enable a comprehensive understanding of how the sulphate ion contained within speleothem calcite can be used as a palaeoclimatic indicator of atmospheric sulphur loading.

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Publications

Publications arising from this grant:

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