

Mechanisms Responsible for Abrupt Climate Change

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On millennial time scales, the paleodata suggests marked variability in the climate of Europe and Northern Africa that appears to be concomitant with changes in the strength of the thermohaline circulation in the N. Atlantic. Indeed, it has been suggested that the Little Ice Age and the Medieval warm period were the most recent manifestations of the extremes of this millennial-scale thermohaline-climate oscillation, and that this "mode" of climate variability extends throughout the majority of Northern Hemisphere and, perhaps, the globe. During glacial epochs, the global extent of these climate excursions is unambiguous and the climate changes are large in amplitude and often abrupt in time.

The leading hypothesis for the millennial-scale thermohaline-climate "mode" invokes a switch in the overturning circulation in the (N. Atlantic) ocean, say from "on" to nearly-off, which then causes an abrupt, large cooling over the whole northern hemisphere; the millennial time scales come from the adjustment time of the global ocean.

In this talk I will present observations and calculations that suggest the overturning circulation in the N. Atlantic today has only a modest effect on the wintertime climate of Europe, and an even smaller effect elsewhere. Our results, when applied to the conventional view of the millennial scale climate variability, suggests that the ocean thermohaline circulation changes may be a response to the abrupt climate change, rather than a driver. In support of this view, calculations will be presented that illustrate the potency for small changes in the tropics as the driver of large, abrupt global climate changes during glacial times.

Time permitting, I will present a model that features the tropics as the driver for abrupt climate change.

References

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The Binding of Selenate and Selenite to Cement Minerals

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Selenium is one of the radionuclides that must be considered in nuclear waste management. It is also found in high concentrations in wastewater from coal-fired power plants, and, in minor concentrations, in air pollution control residues from municipal solid waste incineration. Cement is commonly used to solidify and stabilise hazardous waste material. In cementitious systems selenium selenite (SeO_3^{2-}) and selenate (SeO_4^{2-}) are the stable redox states of selenium. It is known that selenate is extremely soluble and only sorbs weakly on mineral surfaces, while selenite sorbs relatively well, and calcium selenite limits its solubility.

In this study, sorption experiments were performed adding selenite or selenate to suspensions of ettringite, monosulfate or C-S-H at a liquid to solid ratio of 500. The initial Se concentrations (both IV and VI) ranged from 0.3 up to 4500 $\mu\text{mol L}^{-1}$. Equilibrium between the calcium sulfoaluminates and selenate was reached quickly; the solution composition was constant only minutes after the Se addition to over 80 d equilibration time. After similar checks with selenate on C-S-H and selenite, all sorption experiments were performed with 7 days of equilibration. While almost no selenate was sorbed on ettringite and C-S-H, it sorbed well on monosulfate. The percentage selenate uptake was independent of the added Se-concentration. Selenite was sorbed to all three solids in similar amounts, which agreed well with values determined for different cement types. Freundlich sorption isotherms were calculated. At high loadings, the ettringite surface became saturated with respect to SeO_3^{2-} . When selenite was sorbed to monosulfate, precipitation of CaSeO_3 was observed at higher selenite additions.

X-ray diffraction analysis of dried samples showed the change of d-spacings when selenate was added to monosulfate. This suggests that the added SeO_4^{2-} anions replace the SO_4^{2-} anions in the monosulfate interlayers. The released sulfate was precipitated as ettringite. In contrast, neither Se(VI) on ettringite nor Se(IV) on ettringite or monosulfate caused a change in d-spacings, suggesting that in these cases, selenium was sorbed by surface complexation and not by ion exchange as in the case of Se(VI) on monosulfate.

It is concluded, that the sorption of SeO_3^{2-} on cement is unspecific and relatively independent of the occurring minerals, whereas the SeO_4^{2-} sorption to cement depends strongly on the amount of monosulfate present in the system.