

## Reconstructing intermediate watermass variability in the Southeast Pacific during the past ~30ka: A multi-proxy study

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Three high sedimentation rate cores from the SE Pacific, which sample three different intermediate waters, provide a means to reconstruct past variability of these climatically-important yet poorly studied watermasses. ODP Site 1233, at 41°S, sits in the core of Antarctic Intermediate Water (AAIW) at 838 m depth. Site 1234 (1015m), at 36°S, is located between AAIW and the underlying southward-flowing, low-oxygen Pacific Central Water (PCW) mass. Site 1235 (489 m) is located close to Site 1234, but is between AAIW and the overlying, southward-flowing, low-dissolved-oxygen Gunther Undercurrent (GUC) water mass.

To understand the history of this region we use redox-sensitive authigenic metals (Mo, U, Re, Cd) to characterize sedimentary redox conditions; C, N, and opal to characterize biogenic production; and Ti and Al to characterize lithogenic inputs. At site 1233 the redox-sensitive elements remain close to detrital levels throughout the record, while these elements show enrichment at sites 1234 and 1235 beginning at about 12 ky and extending through the Holocene. This Holocene dissimilarity in elemental distributions can be explained by a reduced penetration of AAIW; greater influence of northern-sourced, lower-oxygen waters (PCW and GUC); a constant influence but a decrease in oxygenation of these water masses; an increase in biological production at sites 1234 and 1235 over pre-Holocene levels; or as an artifact of changing sedimentation rates.

Records of sedimentary organic carbon and biogenic silica show that the enrichment of the redox-sensitive metals at sites 1234 and 1235 was not accompanied by an increase in biological productivity. It is also unlikely that the oxygenation of both PCW and GUC would covary, given that they come from different source regions. At this point it is difficult to unequivocally separate the possibility of changing watermass influence from the change in sedimentation rates during deglaciation, but our data does seem to suggest that some of our observed changes may be ventilation-driven rather than being an effect of dilution from lithogenic sources.

## Secondary consequences of Uranium concentration limits for geologic isolation of high-level nuclear waste

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Although the radioactivity of uranium isotopes is low in high-level nuclear waste compared to that of fission products and other actinides, uranium geochemistry exerts important secondary controls on geologic isolation of radioactivity. At the proposed nuclear waste disposal site at Yucca Mountain, Nevada, the  $\text{UO}_2$  matrix of spent nuclear fuel will oxidize, and uranyl mineral alteration products will dominate near-field uranium chemistry. Other radionuclides (e.g.  $^{237}\text{Np}$ ) can coprecipitate in uranyl minerals, and uranyl mineral stability may control source term concentrations of these radionuclides. Using reversed solubility data for uranophane at low temperature [1], total equilibrium uranium concentrations are calculated to be near  $10^{-7}$  M in Yucca Mountain groundwater. This value is consistent with model calculations of uranium concentrations for the Nopal I natural analog system [2]. Models for groundwater in the Yucca Mountain environment at equilibrium with uranophane containing 0.001 mole fraction Np-uranophane yield aqueous Np concentrations that are low compared to values used in Yucca Mountain performance assessment calculations [3].

Ingrowth and release of uranium decay series radionuclides (e.g.  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$ ) constitute principal natural environmental radiation hazards. In a steady-state scenario relevant to the period of geologic stability at Yucca Mountain, uranium concentration limits in the waste emplacement environment would control aqueous uranium concentrations along flow streamlines to the accessible environment, which would in turn control decay series isotope radioactivities. Natural analog data for groundwater that is conditioned by long term contact with uranyl minerals provide realistic values for aqueous concentrations of decay series isotopes including those of Ra and Rn.

[1] Prikryl (in prep.) [2] Pickett & Murphy (1999) *MRS Proc.* **556**, 809-816. [3] Murphy & Grambow (2008, in press) *Radiochimica Acta*.