

Low latitude surface ocean contribution to the deglacial atmospheric radiocarbon decline

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The bulk of the atmospheric radiocarbon decline across the last deglaciation was likely driven by a release of ¹⁴C-depleted CO₂ from the deep ocean. Renewed deep mixing in the Southern Ocean is most often cited as the main mechanism of that release. However, observations from the intermediate-depth Indo-Pacific suggest that ¹⁴C-depleted waters were also present at low latitudes during deglaciation. If those waters impacted the sea surface via upwelling, then part of the atmospheric $\Delta^{14}\text{C}$ drop might be attributable to radiocarbon isofluxes across the air-sea interface in the tropics and subtropics.

Here we present new radiocarbon results from two species of shallow-dwelling planktonic foraminifera in Baja California core MV99-PC08 (23.5°N, 111.6°W, 705 m water depth). Calendar ages are estimated by stratigraphic correlation to the GISP2 ice core in Greenland, allowing us to calculate seawater $\Delta^{14}\text{C}$ through time. We find that the two deglacial $\Delta^{14}\text{C}$ minima previously seen in intermediate waters at this site are also reflected in the planktonic foraminifera, but in attenuated form. The rate of decline in *Globigerinoides ruber* $\Delta^{14}\text{C}$ across Heinrich Stadial 1 was slower than in intermediate waters, but faster than in the atmosphere. These observations are consistent with partial upwelling of the $\Delta^{14}\text{C}$ minima to the sea surface. Additional constraint on the vertical transfer is provided by new benthic $\Delta^{14}\text{C}$ data from nearby Soledad Basin (290 m sill depth). We diagnose the potential impact of low latitude ¹⁴C exchange on the atmosphere using a simple numerical model of the atmospheric radiocarbon balance.

Phases and phase transitions of tropospheric aerosols

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Submicron aerosol particles exert a potentially large but not well constrained effect on the radiative balance of Earth's atmosphere by direct interaction with solar radiation and indirectly by modifying cloud properties. They typically consist of a high variety of organic substances, sulfate and other inorganic ions such as ammonium and nitrate. Knowledge of the physical state and morphology of these particles is needed to predict hygroscopicity and gas/particle partitioning of semivolatile species, to improve knowledge of heterogeneous and multiphase chemistry, and to quantify light scattering and absorption.

However, it is difficult to infer the physical state of atmospheric aerosol particles directly from field measurements. Laboratory experiments on model mixtures and thermodynamic models are thus required to gain insight into the phases and phase transitions of aerosols. The high variety of organic substances impedes crystallization and the organic fraction remains as a liquid or glass even at low relative humidity (RH) and cold temperatures. The way this organic phase interacts with inorganic salts depends on its hydrophilicity. We use Raman microscopy to investigate the phase transitions of micrometer-sized particles during humidity cycles and a high-speed video camera to observe the efflorescence process. For particles consisting of dicarboxylic acids and ammonium sulfate, liquid-liquid phase separation into an organic-rich and an aqueous electrolyte phase occurs for O:C ratios of 0.7 or lower. Using the thermodynamic model AIOMFAC (Aerosol Inorganic-Organic Mixtures Functional groups Activity Coefficients) we can also model liquid-liquid equilibria and gas/particle partitioning of such particles. While crystal growth occurs within milliseconds in the aqueous electrolyte phase, it is drastically slowed in the highly viscous organic-rich phase. Slow diffusion in glassy aerosol particles indeed keeps particles off thermodynamic equilibrium during humidity cycles, as we have shown for sucrose particles levitated in an electrodynamic balance. This highlights the need to also consider deviations from thermodynamic equilibria caused by diffusion limitations especially at cold and dry conditions.