Molecular-level ¹⁴C analysis: A review and forecast

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Molecular-level ¹⁴C Analysis in the Past Decade

Over the past decade or so, the advent of novel separation methods [1] and low-level ¹⁴C analysis (micrograms of C) by accelerator mass spectrometry (AMS) [2] has provided new insights into numerous environmental studies. The most diverse results have focused on the source apportionment of combustion-derived polycyclic aromatic hydrocarbons (PAHs) in aquatic sediments [3, 4], household dust [5], and aerosols [3, 6]. For example, Manolokis *et al.* [6] estimated that biomass burning contributed ~10 and 50% of the total PAHs in atmospheric samples collected from southern Europe and central Sweden, respectively.

Other studies have employed ¹⁴C to differentiate natural verus industrial compounds [7] and the uptake of fossil C into biomass [8-9].

Future for Molecular-level ¹⁴C Analysis

Two important methods will change the face of \$^{14}\$C analysis. One will be the ability to make continuous-flow \$^{14}\$C analysis. In one possible approach, a gas chromatograph could be interfaced to a combustion reactor that will convert the organic molecules into \$CO_2\$ and then deliver the latter directly in an AMS [10]; hence, eliminating the time consuming steps currently necessary to isolate pure compounds for AMS analysis. The other approach will be using low-level \$^{14}\$C-labelled molecules for tracer experiments. Such low amounts are too low to be measured by liquid scintillation, but are easily measured via AMS. With such labels, it will be possible to perform experiments at environmentally relevant concentrations.

[1] Eglinton et al. (1996) Anal. Chem. 68, 904-912. [2] Pearson et al. (1998) Radiocarbon 40, 61-75. [3] Reddy et al. (2002) Env. Sci. & Technol. 36, 1774-1782. [4] Mandalakis et al. (2004) Env. Sci. & Technol. 38, 5344-5349. [5] Reddy et al. (2003) Env. For. 4, 191-197. [6] Mandalakis et al. (2005) Env. Sci. & Technol. 39, 2976-2982. [7] Teuten et al. (2005) Science, 307, 917-920. [8] Wakeham et al. (2006) GCA 70 1761-1771. [9] Slater et al. (2005) Env. Sci. & Technol. 39, 2552-2558. [10] Schneider et al. (2004) Nucl. Instrum. Meth. B. 223, 149-154.

Dimethyl sulfide production in a saline eutrophic lake, Salton Sea, California

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The Salton Sea $(33.3^{\circ}, -115.8^{\circ})$ is regionally recognized for its high and annually increasing salinity (currently 48), warm surface water temperatures (14°C) in winter to 32°C in summer), high algal productivity, and high sulfate concentrations $(110 \text{ to } 135 \text{ mmol } \text{L}^{-1})$. It is a large (980 km^2) , shallow (maximum depth of 15 m), discontinuous polymictic lake located in an arid terminal basin in Southern California. The surrounding areas are dominated by irrigated agriculture, which contributes to high nutrient loading into the Salton Sea. Furthermore, the phytoplankton assemblage in the Salton Sea is typical of marine rather than freshwater environments, due to intentional and unintentional introductions.

The concentrations and distribution of volatile organic sulfur compounds (VOSCs), were quantified over a 13-month period (May 2006 through June 2007). The concentrations of dimethyl sulfide (DMS) appear to be the highest reported thus far for any natural body of water, with an average surface (0 to 2 m depth) concentration of 2.5 μmol L⁻¹. DMS concentrations as high as 11 µmol L⁻¹ were measured, and the concentrations of DMS strongly correlated with chlorophyll a ($R^2 = 0.62$, n =265, p < 0.05). Very high concentrations of dimethylsulfoniopropionate (DMSP), an osmolyte in marine algae, were also measured (average total DMSP of 2.4 µmol L 1), with concentrations strongly correlated with chlorophyll a $(R^2 = 0.88, n = 36, p < 0.05)$. The biomass of the Salton Sea is composed mostly of marine phytoplankton species that are high DMSP producers; based on the correlations of DMS, chlorophyll a, and DMSP, it appears that the DMS in the Salton Sea is directly linked to algal biomass through DMSP. As a result of its very high DMS concentrations, the average estimated volatilization at the Salton Sea (480 µmol m⁻² d⁻¹) was greater than the estimates of other lakes and the open ocean. We calculate approximately 9.6 x 10⁵ mol of DMS was volatilized off the surface of the Sea during the course of this study.