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Isotopic Black Carbon in the Environment: New Metrology for ^{14}C and its International Impact

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Objective: To develop highly reliable isotopic-chemical reference methods and reference materials for the apportionment of sources and the assessment of temporal and spatial distributions of black carbon (BC) in the global environment.

Problem: Black carbon, known also as elemental carbon and soot carbon has long been one of the most elusive yet most critical species demanding high quality measurements and standards for the urban, regional, and global environments. The importance of fine particle BC derives from its unique role as a tracer of fire, combined with its impacts on visibility, health, and climate change. Isotopic speciation (^{13}C , ^{14}C) in BC is essential for the quantitative assignment of sources (apportionment), including anthropogenic which are subject to control, and natural, which are not. BC metrology is beset with twin problems: First, since BC is not a unique chemical substance, but a continuum, the resulting method dependence has led to many discordant results and erroneous conclusions. Second, since isotopic measurements require isolation of the BC often in minute amounts, the preferred optical methods do not apply, and the more vigorous chemical methods may lead to severe problems with contamination and losses.

Approach: The first problem has been addressed through international comparison involving a multidisciplinary team representing some of the most competent workers in the field and a broad range of chemical, thermal, and optical methods, all applied to the same NIST SRM 1649a (Urban Dust). The results of the intercomparison were presented at the Ninth Annual Goldschmidt Conference [1], and they are being incorporated in the new Certificate of Analysis for SRM 1649a. This represents the first comprehensive effort to provide "operational" (method-specific) BC Reference Values. The second problem has been attacked by developing a new "clean chemistry" method for BC analysis and isolation, the Thermal Optical Kinetic (TOK) technique. The TOK method, which was first introduced also at the Goldschmidt Conference [2], uses only high purity gases as reagents and a small quartz oven as reaction chamber. Both absorbance and carbon reaction rate are monitored with time, permitting the deconvolution of reactivity classes of components and sensing the onset of BC oxidation. By stopping the reaction at the appropriate time, minute BC residues can be isolated without losses and without reagent contamination, for high sensitivity ^{14}C accelerator mass spectrometry (AMS). With TOK, valid AMS results have been obtained with as little as 6 mg BC, some 50 times smaller than that measured with (wet) chemical methods of isolation.

Results and Future Plans: The increasing importance of BC in the atmospheric, earth, and marine sciences was evident in another outcome of the Goldschmidt Conference, the formation of the International Steering Committee for the Development of Black Carbon Reference Materials. This six-member committee, which includes NIST representation (LAC), has joint chairs at UC Irvine and the Max Planck Inst. für Biogeochemie, Jena. We expect it to serve as the central international vehicle for generation of suitable reference materials,

method-specific reference values, and accuracy control through international comparisons. The TOK method shows great promise for high quality BC carbon isotopic data at the mg level. The small sample size, combined with the simultaneously produced chemical data, makes it an excellent candidate for objective and quantitative apportionment of atmospheric soot deriving from a variety of fossil and biomass combustion sources.

[1] L.A. Currie et al. (15 coauthors), Sympos. Black Carbon in the Environment, paper 7149 (Aug. 1999).

[2] L.A. Currie and J.D. Kessler, Sympos. Black Carbon in the Environment, paper 7148 (Aug. 1999).

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