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On the isolation of elemental carbon (EC) for micro-molar ^{14}C accelerator mass spectrometry: development of a hybrid reference material for ^{14}C-EC accuracy assurance, and a critical evaluation of the thermal optical kinetic (TOK) EC isolation procedure

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Abstract. The primary objective of the research reported here has been the development of a hybrid reference material (RM) to serve as a test of accuracy for elemental carbon (EC) isotopic (^{14}C) speciation measurements. Such measurements are vital for the quantitative apportionment of fossil and biomass sources of "soot" (EC), the tracer of fire that has profound effects on health, atmospheric visibility, and climate. Previous studies of ^{14}C-EC measurement quality, carried out with NIST SRM 1649a (Urban Dust), showed a range of results, but since the "truth" was not known for this natural matrix RM, one had to rely on isotopic-chemical consistency evidence (^{14}C in PAH, EC) of measurement validity (Currie et al., 2002). Components of the new Hybrid RM (DiesApple), however, have known ^{14}C and EC composition, and they are nearly orthogonal (isotopically and chemically). NIST SRM 2975 (Forklift Diesel Soot) has little or no ^{14}C, and its major compositional component is EC; SRM 1515 (Apple Leaves) has the ^{14}C content of biomass-C, and it has little or no EC. Thus, the Hybrid RM can serve as an absolute isotopic test for the absence of EC-mimicking pyrolysis-C (char) from SRM 1515 in the EC isolate of the Hybrid RM, as well as a test for conservation of its dominant soot fraction throughout the isolation procedure.
The secondary objective was to employ the Hybrid RM for the comparative evaluation of the thermal optical kinetic (TOK) and thermal optical transmission (TOT) methods for the isolation of EC for micro-molar carbon accelerator mass spectrometry (AMS). As part of this process, the relatively new TOK method was subjected to a critical evaluation and significant development. Key findings of our study are: (1) both methods exhibited biomass-C "leakage"; for TOT, the EC fraction isolated for AMS contained about 8% of the original biomass-C; for TOK, the refractory carbon (RC) isolated contained about 3% of the original biomass-C.; (2) the initial isothermal oxidation stage of the TOK method substantially reduced the transfer of artifact char to the RC fraction, improving isolation capabilities; (3) the Hybrid RM was not equal to the sum of its parts, with matrix interactions inducing premature loss of EC which, however, could be quantified and minimized; (4) the three-stage TOK method provided a superior capability for carbonate quantification at the sub-micromolar level, with "reagent-free" removal of carbonate-C from EC – essential for low-

level EC-^{14}C AMS.

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