

Identification and ^{14}C analysis of black carbon and elemental carbon in sediments

C. Steiner¹, M. Vonwiller¹, G. Salazar¹ and S. Szidat¹

¹ Department of Chemistry and Biochemistry & Oeschger Centre for Climate Change Research, University of Bern, 3012 Bern, Switzerland

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Presenting author email: matthias.vonwiller@dcb.unibe.ch

Sediments are an important sink of air-borne particulate matter (PM). Consequently, lake sediments may serve as environmental archives of PM concentrations of the atmosphere of the past. However, two drawbacks limit the potential of lake sediments as such archives. On the one hand, the regional atmospheric input needs to be distinguished from the local input of the catchment of the lake, which has been deposited by surface run-off or resuspended by wind erosion. On the other hand, tracing back atmospheric PM concentrations from sediment records is often hardly possible due to the lack of quantitative information on scavenging and deposition factors.

One of the few examples of PM components that have been studied qualitatively and quantitatively in lake sediments is black carbon (BC). This investigation is mainly driven by the determination of the residence time and the fate of this refractory constituent within the carbon cycle (Hammes et al., 2007). It should be noted, however, that the terminology of BC within the sediment and atmospheric communities is not used consistently. Whereas BC is defined as an ideally light-absorbing substance composed of carbon and discriminated from elemental carbon (EC) as derived from evolved thermal methods in atmospheric sciences (Petzold et al., 2013), it describes a continuum of slightly charred biomass, charcoal and soot in the sediment community (Bird and Ascough, 2012; see Figure 1). Of these, only soot originates from regional atmospheric input and can, therefore, be regarded as synonymous with the definitions of BC and/or EC in atmospheric sciences.

Although strongly differing results of various BC methods in sediments have been discussed controversially in ring trials (Hammes et al., 2007), only few procedures aimed at the identification of the atmospheric component in sediment BC based on methods used in the aerosol community (e.g., Han et al., 2007 and 2009; Khan et al., 2009). These procedures remove inorganic and labile organic sediment components by acid treatment and determine the remaining EC using thermal-optical protocols. For the purpose of isolation of EC for radiocarbon (^{14}C) analysis for apportionment of fossil vs. wood-burning emissions (Szidat, 2009), however, these methods are not suitable due to strong charring of many of the current thermal-optical protocols. Nevertheless, this drawback has largely been overcome by the Swiss_4S protocol, which applies organic carbon (OC) removal from the PM filters by oxidation instead of evaporation (Zhang et al., 2012). Therefore, we investigate in this work the combination of the acid treatment with the Swiss_4S protocol in order to remove the inorganic sediment matrix, OC and other organic components completely and monitor charring as well as EC losses for an optimized source apportionment of sediment EC into fossil and wood-burning emissions with ^{14}C analysis.

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	slightly charred biomass	charcoal	soot
pyrogenic carbon content	lower		higher
chemical structure	'disorganized' low aromaticity		'organized' high aromaticity
common particle size	mm and larger	mm-cm	μm and smaller
common formation temperature		<350°C - >350°C	>500°C
O/C and H/C		>0.5	<0.5

Figure 1. BC fractions in sediments as modified from Bird and Ascough (2012). The BC fraction soot represents the atmospheric input as a synonym for BC and/or EC according to Petzold et al. (2013).