



# 15,000 years of black carbon deposition – A post-glacial fire record from maar lake sediments (Germany)



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## ABSTRACT

Fires accompanied human development throughout the Holocene, leaving behind black carbon (BC) as residues from incomplete biomass burning. Here we used molecular fire markers, benzene polycarboxylic acids (BPCAs), to reconstruct fire history in two Eifel maar lakes, Germany. We hypothesized to find indications for (i) changes in BC related to ecosystem changes, (ii) an increase in BC influx at the onset of agriculture until modern times, and (iii) a change in BC quality due to technical progress in combustion, e.g., at the beginning of agriculture and at the onset of the Bronze Age. To calculate absolute BC influx into the maar lakes, we multiplied BC contents with sedimentation rates. The BC influx rates were elevated during tundra-like vegetation in the Late Pleistocene (up to  $7.7 \text{ g BC m}^{-2} \text{ a}^{-1}$ ), followed by relatively constant  $2.5 \text{ g BC m}^{-2} \text{ a}^{-1}$  from the Bølling interstadial ( $>13.7$  kilo years before present, ka BP) until the early Atlantic when forest began to develop. Thereafter, BC influx increased with the onset of land use of Neolithic cultures in the region from 7.5 ka BP to rates of  $7\text{--}9 \text{ g BC m}^{-2} \text{ a}^{-1}$ . Noteworthy, also the quality of BC changed: higher ratios of five- to six-times carboxylated benzenes (B5CA/B6CA) pointed at colder, arable fires approximately 1000 years after first Neolithic activity from 6 to 4 ka BP (B5CA/B6CA increased from 1.0 to 2.0). From 4 ka BP (Bronze Age) to modern times increasing burning temperatures as indicated by dropping B5CA/B6CA ratios (from 2.0 to 1.0) were related to metallurgy and industrialization. Between 2.5 and 1 ka BP maximum BC influx rates were reached with ca  $15 \text{ g BC m}^{-2} \text{ a}^{-1}$ . With increasing combustion efficiency and a reduction of wild fires during the last centuries, total BC influx decreased, suggesting that fossil fuel combustion contributed less to total BC input into the lake sediments than former vegetation fires.

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## 1. Introduction

Since the mid Holocene humans started to notably influence ecosystems in Europe (Dearing and Jones, 2003), including increasing burning activities (Marlon et al., 2013). In addition to human impact ecosystems also changed because of climatic fluctuations. To decipher to which degree man or climate triggered prehistoric fires, Holocene charcoal records from different terrestrial archives have been investigated (e.g. see review by Marlon et al., 2013). However, a differentiation of prehistoric combustion processes has not been achieved so far.

Natural fire frequencies vary in different types of ecosystems. Ecosystems with high biomass production and pronounced minima in precipitation, e.g., savannas, are most affected by fire. However, the relationship between fire frequency and climate is non-linear; e.g., in dry regions biomass production may be too low to sustain burning events, as it is the case in extreme cold and hot regions (Thonicke et al., 2010). High moisture contents may affect ignitability and hence fire frequency and fire intensity (e.g., Marlon et al., 2013). Marlon et al. (2013) and other authors reported consistent relationships between fire frequency and climate by evaluating palaeosol (Kaal et al., 2011; Schatz et al., 2011; Wang et al., 2012) and lake sediment records (Sadori and Giardini, 2007; Hallett and Anderson, 2010; Tan et al., 2011). Fire intensity was reported to increase in mountainous regions due to reduced snowpack and warmer summer temperatures, i.e. when trees are pushed uphill by expansion of lower-elevation species (Hallett and Anderson,

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2010). In the Mediterranean high fire activity was correlated with drought events (López-Blanco et al., 2011; Bernhardt et al., 2012; Colombaroli and Tinner, 2013). In other studies from China and Hungary, cooling lead to an increase in grassland vegetation and hence to higher fire activity, particularly at the Pleistocene/Holocene boundary (Schatz et al., 2011; Wang et al., 2012). Daniau et al. (2012) conclude that over the past 21 ka temperature increase is the key driver of fire with a peak at intermediate moisture levels. Thonicke et al. (2010) showed that today climate and fuel availability are key factors to fire frequency. They found that in seasonally dry regions, e.g., in savannas, where vegetation recovers quickly, fire frequency is highest with up to one fire per km<sup>2</sup> and year, whereas in wetter and colder regions with deciduous forests fire frequencies are by a factor 4 to 25 lower. Apart from the pure presence or absence of charcoal, it has not yet been tested whether changes in fire residue quantity in Holocene sediments correlate with the fire frequency of the respective ecosystem due to the lack of methodological sensitivity.

The interaction of men and fire becomes evident in sediments of the Mid Holocene: the beginning of agricultural activity (in Europe < 8 ka BP) went along with a higher fire frequency, due to population increase (Marlon et al., 2013). With the onset of agriculture, fire had increasingly been used for forest clearings, fire management, cooking and industry. Nevertheless, population growth stopped correlating with increased charcoal deposition towards modern times (Marlon et al., 2013). Especially during the last centuries, a decrease in fire frequency was reported for densely populated regions, due to landscape fragmentation (preventing ignition of larger plots) and the need to suppress open fires for safety reasons and it remained challenging to differentiate human from climate impact on fire residue deposition (Marlon et al., 2013). Assumedly this is because most studies only investigated charcoal abundances but did not consider variations in the complete combustion residue spectrum by means of geochemical black carbon (BC) analysis.

While charcoal counts allow for a reasonable estimation of relative differences in BC inputs, some char and especially soot remains are too small to be identified by eye and microscope and therewith escape this analytical window. Some studies have already shown the beneficial effect of combining charcoal analysis with geochemical approaches (e.g., Han et al., 2012). For example geochemical proxies allow studying the quantity and quality of submicron scale, molecular BC remains. In particular, the use of benzene-polycarboxylic acids (BPCAs) is suitable as marker for BC (e.g., Glaser et al., 1998; Brodowski et al., 2005). The method converts condensed aromatic BC moieties to BPCAs with different degree of carboxylation providing additional information about burning temperatures: the hotter a fire burns, the more benzene with 6 carboxyl rings are isolated from the combustion residue (see B6CA in Table 1; Schneider et al., 2010, 2013; Wolf et al., 2013). Based on a compilation of literature data, Wolf et al. (2013) differentiated forest, grass and shrub from domestic fires by characteristic fire temperature ranges, which are well reflected in the BPCA pattern. Hence, variations in the B5CA to B6CA ratio potentially allow deciphering changes in fire type in the surroundings of the lakes under study (Table 1). Therefore, this method appears

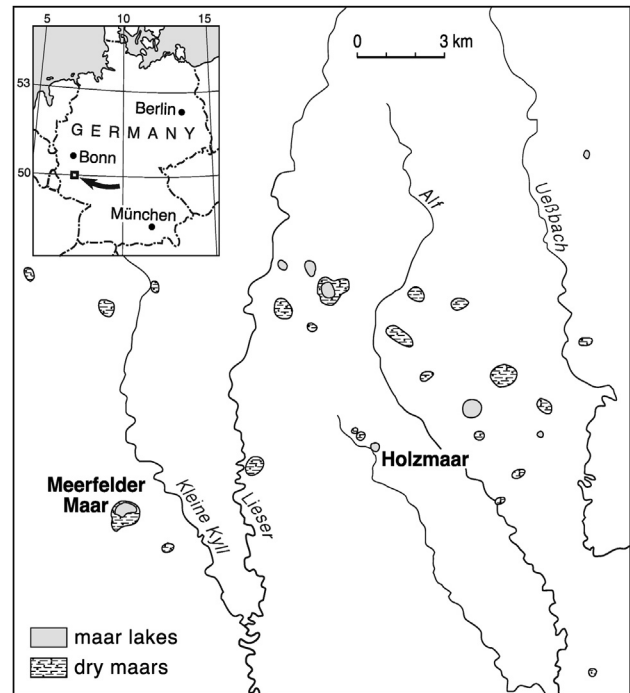


Fig. 1. Map of the Western Eifel Volcanic Field and location of the lakes Holzmaar and Meerfelder Maar in Germany (map from Litt et al., 2009).

suitable to study fire history in high-resolution and well-dated lake sediment records in regions that were settled since several millennia.

Sediment cores from Lake Holzmaar and Meerfelder Maar (Eifel, Germany) were shown to record Holocene vegetation and climate change (Fuhrmann et al., 2004; Lücke and Brauer, 2004; Litt et al., 2009; Martín-Puertas et al., 2012) as well as cultural history (Litt and Kubitz, 2000). The maar lakes started archiving in the Pleniglacial where vegetation cover was low and tundra-like as revealed by organic proxies (Zolitschka et al., 2000; Lücke et al., 2003; Baier et al., 2004; Fuhrmann et al., 2004) and pollen data (Leroy et al., 2000; Brauer et al., 2001; Litt et al., 2001, 2003). Likely due to the scattered vegetation cover, erosion rates and minerogenic input were at a maximum at this time and decreased until app. 10 ka when climate became, successively and with regressions, warmer and vegetation more dense (Litt et al., 2003, 2009; Lücke et al., 2003; Brüchmann and Negendank, 2004). Organic proxies, i.e., total organic carbon, stable carbon isotope composition, n-alkanes and lignin abundance (Lücke et al., 2003; Fuhrmann et al., 2004) and pollen data (Litt et al., 2003, 2009) indicated that the climate ameliorations were accompanied by shifts from tundra-like and tundra-steppe vegetation (Pleniglacial, Oldest Dryas and Younger Dryas) to shrub and forest dominated vegetation (Meiendorf, Bølling, Older Dryas, Allerød) until the beginning of the Holocene when forest was dominant (Litt and Stebich, 1999; Brauer et al., 1999a; Fuhrmann et al., 2004). From the Holocene on

Table 1

Individual BPCAs, their relative contribution to the sum of BPCAs and their dominant sources in the maar lakes.

Abbreviation	B3CAs	B4CAs	B5CA	B6CA <sup>a</sup>
Trivial name	Hemimellitic, trimellitic & trimesic acid	Pyromellitic, melophanic & prehnitic acid	Pentacarboxylic acid	Mellitic acid
Contribution to BPCAs	<2–12%	<4–14%	30–47%	21–38%
Dominant source	Organic matter << charcoal, soot	Organic matter << charcoal, soot	Soot << charcoal	Charcoal << soot

<sup>a</sup> Relative abundance of mellitic acid depends strongly on fire temperature (see text and literature for details).

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