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A European aerosol phenomenology -4: Harmonized concentrations of carbonaceous aerosol at 10 regional background sites across Europe

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HIGHLIGHTS

- Artefacts bias the sampling of carbonaceous matter by quartz fibre filters.
- Identical thermal protocols run on various instruments produce different results.
- Seasonal variations can be observed in intensive carbonaceous aerosol variables.
- TC/PM_{10} ratios range from 12 to 34% across European regional background sites.
- Site-mean EC/TC ratios range from 10 to 22% and get similar at all sites in winter.

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ABSTRACT

Although particulate organic and elemental carbon (OC and EC) are important constituents of the suspended atmospheric particulate matter (PM), measurements of OC and EC are much less common and more uncertain than measurements of e.g. the ionic components of PM. In the framework of atmospheric research infrastructures supported by the European Union, actions have been undertaken to determine and mitigate sampling artefacts, and assess the comparability of OC and EC data obtained in a network of 10 atmospheric observatories across Europe. Positive sampling artefacts (from 0.4 to 2.8 μg C/m^3) and analytical discrepancies (between −50% and +40% for the EC/TC ratio) have been taken into account to generate a robust data set, from which we established the phenomenology of carbonaceous aerosols at

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regional background sites in Europe. Across the network, TC and EC annual average concentrations range from 0.4 to 9 μg C/m^3, and from 0.1 to 2 μg C/m^3, respectively. TC/PM_{10} annual mean ratios range from 0.11 at a Mediterranean site to 0.34 at the most polluted continental site, and TC/$PM_{2.5}$ ratios are slightly greater at all sites (0.15–0.42). EC/TC annual mean ratios range from 0.10 to 0.22, and do not depend much on PM concentration levels, especially in winter. Seasonal variations in PM and TC concentrations, and in TC/PM and EC/TC ratios, differ across the network, which can be explained by seasonal changes in PM source contributions at some sites.

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1. Introduction

Carbonaceous aerosol is a complex mixture of many organics (the OC fraction) and elemental carbon (EC). As some of these organics are highly toxic and elemental carbon is present largely as solid insoluble nanoparticles, carbonaceous aerosol could have a larger health impact than other PM constituents (Cassee et al., 2013; WHO, 2013). Carbonaceous particles also play a clear role in climate change through direct and indirect radiative forcing, although the magnitude of these effects is still quite uncertain (Boucher et al., 2013). During the last decade, OC and EC data have been measured at many sites across Europe, (e.g. Pio et al., 2007; Yttri et al., 2007a; Querol et al., 2013). Such measurements are extremely valuable for assessing temporal trends and spatial variability in OC and EC concentrations (Yttri et al., 2007b; Putaud et al., 2010; Tørseth et al., 2012). In-situ measurements in general are also essential for calibrating or validating data retrievals from remote sensing and model outputs. However, the accuracy and precision of particulate OC and EC data is particularly questionable since various factors can lead to large errors in OC and EC data, both at the sampling and analysis stages.

Artefacts can affect the sampling of particulate organic carbon, which is always carried out on quartz fibre filters. They have been extensively studied in the USA for more than 2 decades (e.g. McDow and Huntzicker, 1990; Turpin and Huntzicker, 1994; Mader et al., 2001; Watson et al., 2009). They found positive sampling artefacts ranging between 0.2 and 3 μgC/m^3, increasing with the particulate total carbon (TC) concentration, and decreasing with the sampling face velocity. In Europe, less information is available. From studies by Viana et al. (2006) and Schwarz et al. (2008), it could be estimated that the contribution of positive artefacts to the total amount of OC collected by a quartz fibre filter was on average about 30% in Ghent (Belgium), and Prague, (Czech Republic). At Nordic sites for 1 week sampling times, the mean positive sampling artefact ranged from 11% to 18% of OC (Yttri et al., 2011a).

Analytically, atmospheric particulate carbon has traditionally been split into OC and EC, although drawing a clear border between organic macro-molecules (OC) and small clusters of (possibly amorphous) EC is challenging (Baumgardner et al., 2012). Furthermore, charring can transform a part of OC into species looking like EC during the analysis, which must be accounted for (Chow et al., 1993; Birch and Cary, 1996). Eventually, OC and EC are operationally defined, and values produced by various laboratories using identical or different methods can be very different from each other, especially for EC. Various studies report differences up to a factor of 2 when comparing EC resulting from different methods, and reproducibility standard deviations in the range of 10–25% for the determination of EC by a given method (e.g. Watson et al., 2005; Karanasiou et al., 2015).

The current study reports on a specific action aimed at providing robust and comparable data on particulate carbonaceous aerosol across Europe. This long-term action was carried out under the European Research Infrastructure projects EUSAAR (European Supersites for Atmospheric Aerosol Research) and ACTRIS (Aerosols, Clouds, and trace gases Research Infrastructure, www.actris.eu). Coordinated experiments were performed to assess the positive and negative artefacts which affect particulate OC sampling during different seasons at several regional background sites across Europe. A sampling train (Fig. S1), which minimizes positive sampling artefacts without significantly increasing negative artefacts was also tested and validated. The comparability of the analyses performed by all the laboratories which produced the data discussed in the current study was also assessed on the basis of annual inter-laboratory comparisons.

Combining our knowledge of site-dependent sampling artefacts and laboratory-dependent possible analytical discrepancies allowed us to construct the most robust data set on particulate carbonaceous aerosol available for Europe so far. We can thus discuss with a level of confidence previously not available the similarities and differences in carbonaceous aerosol concentration, its contribution to PM mass, and its composition in terms of OC and EC, among 10 regional background sites across Europe. Seasonal variations are also examined, which can provide information on carbonaceous aerosol sources at some of these sites.

2. Experimental

The data we discuss here were obtained between 2008 and 2011 as a result of the collaboration among research institutes running 10 atmospheric observatories at regional background sites located across Europe (Fig. 1): Aspvreten (APT), Birkenes (BIR), Vavihil (VAV), Harwell (HRL), Melpitz (MEL), Kosetice (KOS), Ispra (IPR), Puy de Dôme (PUY), Montseny (MSY), and Finokalia (FIK). Specific experiments related to sampling artefacts were also performed at Hurdal (HUR), Mace Head (MHD), and K-puszta (KPS).

2.1. Mass and carbonaceous aerosol concentration measurements

2.1.1. Sampling

Sampling was performed using quartz fibre filters of different types for periods between 24 and 168 h at face velocities ranging 20–53 cm/s (Table 1). Denuders (P/Nr 55-008923-002, Air Monitors, UK) were continuously used for daily measurements for at least one size fraction at APT, VAV, and IPR, as well as in KOS from Sep. 2011. Quartz fibre back up filters were used for daily measurements at KOS, and at 7 more sites to assess positive sampling artefacts during specific experiments (Table 1). At the remaining 4 sites, bare quartz fibre filters only were used.

2.1.2. Analysis

PM_{10} and $PM_{2.5}$ mass concentrations were determined by gravimetric analyses of the quartz fibre filters used for OC and EC measurements at 4 sites, by gravimetric analyses of Teflon™ and Emfab™ filters collected simultaneously at KOS and HRL,

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