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# Source apportionment of the carbonaceous aerosol in Norway – quantitative estimates based on <sup>14</sup>C, thermal-optical and organic tracer analysis

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aerosol from biogenic precursors (OC<sub>BSOA</sub>).

In the present study, source apportionment of the ambient summer and winter time particulate carbonaceous matter (PCM) in aerosol particles (PM $_1$  and PM $_{10}$ ) has been conducted for the Norwegian urban and rural background environment. Statistical treatment of data from thermal-optical,  $^{14}$ C and organic tracer analysis using Latin Hypercube Sampling has allowed for quantitative estimates of seven different sources contributing to the ambient carbonaceous aerosol. These are: elemental carbon from combustion of biomass (EC $_{bb}$ ) and fossil fuel (EC $_{ff}$ ), organic carbon from combustion of biomass (OC $_{bb}$ ), fossil fuel (OC $_{ff}$ ), primary biological aerosol particles (OC $_{PBAP}$ , which includes plant debris, OC $_{pbc}$ , and fungal spores, OC $_{pbs}$ ), and secondary organic

Our results show that emissions from natural sources were particularly abundant in summer, and with a more pronounced influence at the rural compared to the urban background site. 80% of total carbon (TC $_p$ , corrected for the positive artefact) in PM $_{10}$  and 70% of TC $_p$  in PM $_1$  could be attributed to natural sources at the rural background site in summer. Natural sources account for about 50% of TC $_p$  in PM $_{10}$  at the urban background site as well. The natural source contribution was always dominated by OC $_{BSOA}$ , regardless of season, site and size fraction. During winter anthropogenic sources totally dominated the carbonaceous aerosol (83–90%). Combustion of biomass contributed slightly more than fossil-fuel sources in winter, whereas emissions from fossil-fuel sources were more abundant in summer.

Mass closure calculations show that PCM likely dominated the mass concentration of the ambient PM regardless of size fraction, season, and site. A larger fraction of PM<sub>1</sub>(64–69%) was accounted for by carbonaceous matter compared to PM<sub>10</sub> (51–67%), but only by a small margin. In general, there were no pronounced differences in the relative contribution of carbonaceous matter to PM with respect to season or between the two sites.

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#### Introduction

Particulate carbonaceous matter (PCM) is found to constitute 10-40% (mean 30%) of PM<sub>10</sub> levels at rural and natural background sites in Europe (Yttri et al., 2007a; Putaud et al., 2004). PCM is believed to have important impacts on global climate (Novakov and Penner, 1993; Kanakidou et al., 2005), and on human health (McDonald et al., 2004; Bell et al., 2009). PCM consists largely of organic matter (OM), and usually smaller quanities of so-called elemental or black carbon (EC or BC). OM is dominated by its carbon content (OC), but with associated oxygen, hydrogen, nitrogen, sulphur. The sum of EC and OC is referred to as total carbon (TC). (Table 1 summarizes the notation used in the present study.) Recent reviews have highlighted the complexity of the carbonaceous aerosol both in terms composition and formation mechanisms (Baltensperger et al., 2005; Donahue et al., 2005, 2009; Kanakidou et al., 2005; Kroll and Seinfeld, 2008; Pöschl, 2005; Hallquist et al., 2009).

Over the last few years a number of studies have become available which shed light on the important sources of PCM in Europe. Szidat et al. (2006) attributed the ambient aerosol carbon content to either fossil carbon (from combustion of coal, oil) or modern carbon (from recent vegetation, either by combustion, emissions, or formation using <sup>14</sup>C-analysis). They found that fossil-fuel combustion accounted for only 30% of OC throughout the year, even in the city center of Zurich (Switzerland). Biomass burning in wintertime and SOA in summertime seemed to account for the majority of the remaining OC. Follow-up studies in Switzerland using a variety of techniques have confirmed the basic source patterns (Szidat et al., 2007; Lanz et al., 2007, 2008).

The EU CARBOSOL project (CARBOnaceous AeroSOLs over Europe, Legrand and Puxbaum, 2007) provided the first long-term assessment (2002-2004) of PCM in Europe. By using the results of <sup>14</sup>C-, thermal-optical-, and organic tracer analysis as input, Gelencsér et al. (2007) made use of a novel statistical method to calculate the likely contributions of different sources of the PCM at six sites in Central/Southern Europe. Wintertime residential-wood burning was found to be a significant contributor to

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measured PCM levels at all the sites, whereas in summertime, biogenic SOA (BSOA) was the dominant contributor. These data enabled for the first time an evaluation of the components (primary, secondary, anthropogenic, biogenic) of chemical transport model simulations (Simpson et al., 2007). Previously, only EC or OC has been available for model comparison, and as shown by Simpson et al. (2007), this has been wholly inadequate for understanding the role of e.g. local biomass burning versus long range transport, or of primary organic aereosol (POA) versus secondary (SOA).

In Northern Europe, studies addressing the sources of PCM have been limited. In a pioneering study, Currie et al. (1986) showed that residential wood burning was the major contributor to the carbonaceous aerosol in winter in Norway. A number of recent studies have confirmed the importance of wood-burning emissions to ambient PM levels in Nordic areas in wintertime (Glasius et al., 2006; Hedberg et al., 2006; Saarikoski et al., 2008; Szidat et al., 2009; Yttri et al., 2005, 2009). Scandinavia is a particularly interesting region with respect to natural sources of PCM. Large forested areas, emitting vast amounts of VOCs during the long days of the Scandinavian summer, create a basis for BSOA formation. Using a network of Nordic sites measuring aerosol number size distributions, combined with extensive trajectory analysis, Tunved et al. (2006) has shown a very good relationship between accumulated biogenic VOC (BVOC) emissions and particle mass changes, suggesting that, in clean-air conditions at least, the buildup of PM can be interpreted almost entirely in terms of biogenic precursors and presumably SOA formation. However, this study described very clean air conditions, and it is unclear how far this result can be generalized to typical Nordic conditions. Nevertheless, a substantial 64% contribution of SOA to OC was reported for Helsinki, Finland, during summertime by Saarikoski et al. (2008), of which the major fraction of SOA was likely to have a biogenic origin. Szidat et al. (2009) found that the contribution of non-fossil sources (likely BSOA) (53%) and fossil sources (47%) to TC almost equaled each other at a site in Central Gothenburg in June 2006.

Yttri et al. (2007a) showed that levels of OC in PM<sub>10</sub> were higher in summer compared to winter at rural background sites in Scandinavia, as compared to rural

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background sites in Continental Europe. Based on results from the Norwegian site Birkenes this was attributed to coarse OC, which constituted approximately 50% of OC in PM<sub>10</sub> on a monthly basis during summer. Further evidence that this coarse OC could be attributed to primary biological aerosol particles (OC<sub>PRAP</sub>) stems from 5 seasonal and size distribution measurements of sugars and sugar-alcohols, which are tracers of OC<sub>PBAP</sub> (Yttri et al., 2007b).

Model studies have also suggested that BSOA formation can make a large contribution to PCM levels in Northern Europe (Andersson-Sköld and Simpson, 2001; Chung and Seinfeld, 2002; Tsigaridis and Kanakidou, 2003; Simpson et al., 2007). The study of Simpson et al. (2007) noted the possibility that BSOA formation may even be a significant contributor to PCM levels in wintertime at Nordic sites, despite low emission rates of precursor species, as a result of the higher condensation rates applicable in cold Nordic conditions. This study also stressed that a major problem in all modeling of PCM is that too many steps in the calculation process are highly uncertain, and there are too few constraints. The uncertainties cover emissions from both anthropogenic and biogenic sources, atmospheric chemistry, and the fate of the various semi volatile products involved in SOA formation. Clearly, there is a strong need for measurements which are able to test the model's abilities to predict all major components of PCM.

In the present study we present concurrent measurements of TC, EC, OC, monosaccharide anhydrides, sugars and sugar-alcohols, and cellulose, conducted at an urban background site (Oslo) and a rural background site (Hurdal), as part of the Norwegian SORGA (Secondary ORGanic Aerosols in Urban Areas) project. These data are used to apportion the measured TC concentrations into primary, secondary, fossil-fuel, and non-fossil- fuel related sources by the aid of <sup>14</sup>C-analysis. Further, the carbonaceous aerosol emitted from these sources has been examined with respect to season, size fraction and time of day.

#### Sampling sites

Aerosol filter samples were collected at an urban background site in Oslo, Norway, (Veterinary Institute, 59°56′ N, 10°44′ E, 77 m a.s.l.) and at a rural background site (Hurdal, 60°22′ N, 11°04′ E, 300 m a.s.l.) situated 70 km north east of Oslo, for a summer period (19 June-15 July 2006) and winter period (1-8 March 2007).

#### Aerosol sampling

#### PM<sub>10</sub>, PM<sub>1</sub>, EC, OC, TC, monosaccharide anhydrides, sugars, 2.2.1 sugar-alcohols

Aerosol (PM<sub>10</sub> and PM<sub>1</sub>) filter samples for subsequent analysis of mass concentration, EC, OC, TC, monosaccharide anhydrides, sugars and sugar-alcohols were collected on prefired (850°C, 3h) quartz fibre filters (Whatman QM-A, 47 mm). Tandem filter setups (McDow and Huntzicker, 1990) operating according to the QBQ-approach (quartzfibre filter behind quartz fiber filter) were applied to account for the positive sampling artefact of OC. All quartz fibre filters were obtained from the same batch number to minimize differences in the adsorptive capacity, which otherwise would have biased the estimate of the positive artefact (Kirchstetter, 2001). After sampling, the front filters were conditioned at a temperature of  $20 \pm 2$  °C and at a relative humidity of  $50 \pm 5$ % for 48 h for gravimetric determination of the mass concentration. Thereafter the filters were placed in petrislides, packed in airtight plastic bags, and stored at -20°C. The backup filters were stored immediately after sampling. Leckel LVS 3.1 samplers, operating at a flowrate of 381 min<sup>-1</sup>, were used to collect aerosol filter samples both at the urbanand the rural background site. Two 12 h samples were collected pr. 24 h, approximating day (9 p.m.-9 a.m.) and night (9 a.m.-9 p.m.), respectively.

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#### 2.2.2 <sup>14</sup>C, cellulose

Aerosol filter samples (PM<sub>10</sub> and PM<sub>1</sub>) were collected at both sites for subsequent <sup>14</sup>C and cellulose analysis. The aerosol were collected on prefired (850°C, 3h) quartz fibre filters (Whatman QM-A, 47 mm). At the rural background site, the time resolution for this sampling was 336 (14  $\times$  24) hours during the summer campaign and 168 (7  $\times$  24) hours during the winter campaign. The time resolution for the urban background site was 168 h during the summer campaign and 84 h during the winter campaign, as sampling was separated into day and night, respectively. A Leckel LVS 3.1 samplers, operating at a flow rate of 381 min<sup>-1</sup> were used to collect PM<sub>1</sub>, whereas a NILU (Norwegian Institute for Air Research) filter holder with an IVL (Swedish Environmental Research Institute) inlet, operating at a flowrate of 16.71 min<sup>-1</sup>, was used to collect PM<sub>10</sub>.

The different sampling procedures (averaging times and filter fac velocity) gave somewhat different estimates of OCp, by 12% of average, presumably due to differences in homogeneity and unavoidable differences in sampling artefacts between the systems. We have thus scaled the sampled cellulose by the OCp ratios in order to generate compatible values of all components.

#### Thermal-optical transmission analysis

The aerosol filter samples content of EC, OC, and TC was quantified using a thermaloptical transmittance (TOT) instrument from Sunset laboratories Inc., operating according to a NIOSH derived temperature program. The "8785 Air Particulate Matter On Filter Media" reference material from The National Institute of Standards and Technology (NIST) was used to test the performance of the temperature program. For further details about the temperature program and its performance see Yttri et al. (2007a).

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#### 2.4.1 Monosaccharide anhydrides

Levoglucosan, mannosan, and galactosan (monosaccharide anhydrides, MAs) were quantified according to the method described by Dye and Yttri (2005), described briefly here. One punch (1.5 cm<sup>2</sup>) from the quartz fibre filter was soaked in tetrahydrofuran (2 ml) and subjected to ultrasonic agitation (30 min). The extract was then filtered through a syringe filter (0.45 µm) to remove PM and filter parts. The filter punch was extracted twice and the extracted volumes were pooled and evaporated to 1 ml by N<sub>2</sub>. Before analysis the sample solvent elution strength was adapted to the mobile phase by adding Milli-Q water (0.8 ml). Concentrations of the individual monosaccharide anhydrides were determined using a High-Performance Liquid Chromatography (HPLC, Agilent model 1100) instrument combined with High-Resolution Mass-Spectrometry - time-of-flight (HRMS-TOF, Micromass model LCT) operated in the negative electrospray mode. The compounds were identified by comparison of retention time and mass spectra with authentic standards. The compound separation was performed with two series-connected reversed-phase C18 columns (Atlantis dC18, Waters). The limit of quantification for the method at a signal to noise ratio of ten is approximately 20-40 ng injected of the individual MAs. An isotope labeled standard of mannosan, purchased from Cambridge Isotope Laboratories, was used as internal standard for quantification of the monosaccharide anhydrides.

#### Sugars and sugar-alcohols 2.4.2

Sugars (fructose, glucose, sucrose, trehalose), and sugar-alcohols (arabitol, erythritol, inositol, mannitol), were quantified according to the method described by Yttri et al. (2007b). Briefly, one punch (1.5 cm<sup>2</sup>) from the quartz fibre filter was soaked in Milli-Q water (1.5 ml) and subjected to ultra sonic agitation (30 min). Each filter was extracted twice. The extracted volumes were pooled, filtered through a syringe filter (0.45 µm)

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to remove particulates and filter debris, and evaporated to dryness under an N<sub>2</sub> atmosphere. The dry extract was redissolved in Milli-Q water (100 µl) and isopropanol (300 µl) prior to analysis. The analysis was performed by HPLC/HRMS-TOF analysis operated in the negative electrospray mode. The compounds were identified by 5 comparison of retention time obtained by separation on an amino column (Asahipak NH2-2D, 2.0 mm id × 150 mm). The limit of quantification of the method at a signal to noise ratio of ten is approximately 30 ng injected of the individual sugars and sugaralcohols. An isotope-labeled standard of glucose, purchased from Cambridge Isotope Laboratories, was used as internal standard for quantification of the sugars and sugaralcohols.

#### 2.4.3 Cellulose

Free cellulose was determined based on the enzymatic method described by Kunit and Puxbaum (1996). Free cellulose was assumed to be 72% of total cellulose (Ibid.). Filter aliquots of 2 cm<sup>2</sup> were eluted with 2.2 ml of 0.05 M citric acid solution (pH of 4.8), containing 0.05% of Thymol to prevent bacterial growth, during 45 min under ultrasonic agitation. The cellulose was then saccharified by two enzymes (Trichoderma reesei cellulase 80 µl 10% and Aspergillus Niger cellobiase 80 µl 1%) during 24 h at 45 °C. After stopping the saccharification by denaturizing the enzymes at 80 °C and centrifuging, the supernatant was collected in order to determine D-glucose using a modified test-combination (D-Glucose-HK, Megazyme, Ireland). Glucose was phosphorylated to glucose-6-phosphate, and subsequently oxidized by Nicotinamide adenine dinucleotide phosphate (NADP) to gluconate-6-phosphate (G-6-P), with the formation of NADPH, stoichiometrically proportional to the amount of D-glucose originally present. NADPH was quantified by UV photometry at 340 nm, read against distilled water. Free glucose was subtracted from the result. The limit of detection is 160 ng cm<sup>-2</sup>.

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#### Radiocarbon analysis

Prior to the <sup>14</sup>C analysis, the carbon was extracted from filters (about 75 µg of carbon per filter). This was achieved by first combusting the sample to CO<sub>2</sub> in presence of pre-cleaned CuO (powder and needles) and Cu (needles) in a vacuum system. Elemental carbon was then produced reducing the CO<sub>2</sub> to elemental carbon using H<sub>2</sub> and a heated metal catalyst (about 2 mg of Fe powder, Merck, pro analysis reduced, diameter 10 µm, at 450 °C), and Mg(ClO<sub>4</sub>)<sub>2</sub> (Merck, diameter 1–4 mm) as a drying agent. The time for complete reduction of each sample was less than 3h. After complete graphitization the carbon and iron catalyst were pressed into Al sample holders and put on a 40 position sample wheel together with graphitized standards (Oxl, IAEA-C6 and IAEA-C7) and blanks (anthracite).

The <sup>14</sup>C analysis was performed by accelerator mass spectrometry at Lund University, using a compact, high capacity machine dedicated to radiocarbon dating, as well as to biomedical and environmental research (Skog, 2007; Skog et al., 2010).

#### Source-apportionment methodology

The measurements of EC, OC, TC, <sup>14</sup>C, levoglucosan, cellulose, sugars and sugaralcohols in this study provide a set of tracers which provide information on the sources of the observed carbonaceous aerosol (c.f. Table 1). Levoglucosan is used as a tracer of TC from woodburning (TC<sub>bb</sub>), whereas measurements of cellulose and selected sugar-alcohols (here: mannitol) can be used to determine the contribution of various primary biological aerosol particles (OC<sub>PBAP</sub>). Measurements of <sup>14</sup>C help to determine the percentage of modern carbon versus carbon from fossil-fuel sources.

Here we perform a very similar analysis as that of Gelencsér et al. (2007), in an effort to calculate the relative contributions of the main anthropogenic and natural sources of the carbonaceous aerosol. The methodology is updated with newer emissions ratios and information on the contribution of OC from fungal-spores provided by the inclusion

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of the sugar-alcohol mannitol. We also make an attempt to account for the negative as well as the positive artifact associated with sampling of OC. The approach used here is summarized in Eqs. (1)–(12), in Table 2, and the terms discussed in Sects. 3.1–3.9.

Each step of this analysis has a level of uncertainty, but a major advantage of the present study compared to those presented in e.g., Gelencsér et al. (2007) and Szidat et al. (2006, 2009) is that almost all parameters are derived from self-consistent studies from Oslo and surrounding areas. In particular, Yttri et al. (2009) reported very high correlations (*R* > ~ 0.9) between levoglucosan and OC in the small town of Elverum, 120 km east of Oslo, during wintertime sampling, and that the wood-burning contribution to OC/EC/TC was close to 100%. Further, related studies by Yttri et al. (2007b, 2005) and Dye and Yttri (2005) all provide a comprehensive background to the nature of the carbonaceous aerosol in Norway, and on the contribution of wood-burning and PBAP. These data have a number of advantages for our analysis:

Location: emission sources at Elverum (type of wood, appliance, etc.) are expected to be similar as those around the sampling sites in the current study.

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- Consistency of analytical methodology: the analytical methodology used to quantify the samples content of EC/OC and levoglucosan in the present study were identical with that reported by Yttri et al. (2009), thus excluding many of the problems associated with differences in analytical methods, e.g. the EC/OC split. Consequently, one does not require any assumptions about the equivalence of the ambient data with those of an emission inventory.
- Consistency of sampling: in the present study the corrections for the positive sampling artifact of OC are consistent with the procedures of Yttri et al. (2009).

Uncertainties still remain of course, and following Gelencsér et al. (2007) we define both a central best-estimate value for each factor and a plausible range of uncertainty. In order to tackle the multitude of possible combinations of uncertain parameters, we have made use of an effective statistical approach known as Latin-hypercube sampling (LHS) (Iman et al., 1981; Beachkofski and Grandhi, 2002). LHS approaches

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more effective way of sampling the data, and for our purposes provides essentially the same results as a full Monte-Carlo analysis. We used the software available at http://people.scs.fsu.edu/~burkardt/f\_src/ihs/ihs.html for this purpose. Due to the paucity of information on the input factors, parameters are (unless otherwise noted below) assigned equally between the low limit and the central value and between the central value and the high limit. All valid combinations of parameters (i.e. excluding those producing negative contributions) are condensed in frequency distributions of possible solutions. Table 3 describes the equations that are used for the source-apportionment analysis, and Table 3 summarizes the best-estimates and range of uncertainty for each factor. The rationale for each value is provided in Sects. 3.1–3.9.

are somewhat similar to Monte Carlo calculations, and allow vast numbers of com-

binations of input variables to be computed. A Monte-Carlo simulation would involve testing all possible combinations of input parameters. LHS provides a much

#### 3.1 Estimation of EC, c.f. Eq. (1)

As discussed extensively by Schmid et al. (2001), different analytical methods give rather similar values for TC, but very different values for EC; i.e. more than a factor of 2 differences for EC concentrations measured by various techniques. Ideally, the same analytical method should be used when establishing the emission ratios including EC (e.g. (OC/EC)<sub>bb</sub>) as that used for analysis of the ambient aerosol content of EC. Since thermal-optical analysis, used in the present study, is the most accepted and widely used technique for EC measurements and also used for the emission ratio assumptions in the present study, we have adopted a narrower range of uncertainty,  $\Phi_{FC}$ , than that found by Schmid et al. (2001), with low, central and best estimates of 0.75, 1.0 and 1.25.

## 3.2 Estimation of OC, TC, c.f. Eqs. (2), (3)

Collection of ambient aerosol filter samples for analysis of the particulate phase OC content is associated with both positive and neagative sampling artefacts. As

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discussed by Chow et al. (2010), the positive artifact is usually bigger than the negative one. We assume this to be true for the Nordic countries as well, thus, the true OC should lie between the measured front and back filter values, i.e. between OC<sub>F</sub> and  $OC_B$ . Hence, we make use of the  $\Phi_{NA}$  term in Eq. (2), and have assigned the low,  $_{5}$  central, and high value of OC to be  $OC_{p}$ ,  $OC_{p} + 0.2 \times (OC_{F} - OC_{B})$ ,  $OC_{F}$ .

Given EC and OC, TC is derived simply as EC + OC (Eq. 3).

#### 3.3 Estimation of TC<sub>bb</sub>, c.f. Eq. (4)

On a mass basis, the largest source of biomass burning emission in Norway, and neighbouring countries, is wood burning in stoves and fireplaces used for residential heating. Wildfires may occassionally impact PCM in Norway (Tsyro et al., 2007; Yttri et al., 2007a), but the contribution seems to be low in general. During the summertime measurement period in the current study, no nearby fire-activity was detected by MODIS (http://modis-fire.umd.edu).

Levoglucosan (LG) is emitted in high concentrations from wood burning and is regarded as a highly useful tracer of wood burning emissions (e.g. Schauer et al., 2001; Simoneit, 2002). A large number of studies concerning the emission ratio [(OC/LG)<sub>bb</sub> or (TC/LG) bb] is available, and their suitability with respect to European conditions has been discussed by Gelencsér et al. (2007); Puxbaum et al. (2007) and Simpson et al. (2007).

Here we make use of the emission ratios estimated by Yttri et al. (2009) for Norwegian urban areas, reporting a best-fit (TC/LG)<sub>bh</sub> ratio of 20 for PM<sub>10</sub> and 16 for PM<sub>2.5</sub>, based upon a quartz-behind-teflon (QBT) sampling protocol. In the present study, levoglucosan was collected on a quartz-behind-quartz fiber filter setup (QBQ), which have been shown to retain some levoglucosan compared to Teflon filters (Dye and Yttri, 2005). Based upon Dye and Yttri (2005), we have applied a factor of 0.76 for the ratios to account for this. Our best-estimate values and corresponding ranges of  $(TC/LG)_{hh}$  are thus 15 (11–17) for PM<sub>10</sub> and 12 (7.6–14) for PM<sub>2.5</sub>. Allowing the lower  $PM_{2.5}$  limit to apply for  $PM_{10}$  also, the range for  $PM_{10}$  is 7.6–17 and 7.6–14 for  $PM_{1}$ .

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It is hard to prove if modern carbon is due to biomass burning or other sources of modern carbon, e.g. BSOA. Thus, we regard the TC<sub>bb</sub> values derived here from levoglucosan as an "operational" definition. As noted in Szidat et al. (2009), OC<sub>bb</sub> when derived from ambient (OC/LG)<sub>bb</sub> measurements may be higher when compared to data derived from ratios from laboratory emission studies, as the data derived from ambient measurements include some condensed and secondary OC. Indeed, Grieshop et al. (2009b,a) demonstrated in smog chamber experiments that wood-burning SOA actually may reach up to similar levels as POA. The ratios used in the present study were derived from wintertime ambient measurements (Yttri et al., 2009) and may also include condensed vapors.

#### 3.4 Estimation of $OC_{bb}$ , $EC_{bb}$ , $EC_{ff}$ , c.f. Eqs. (5)–(7)

Analysis of the Yttri et al. (2009) data suggests best values for (OC/TC)<sub>bb</sub> of 0.78 for PM<sub>10</sub> and 0.71 for PM<sub>2.5</sub>. In general the range of variation of this ratio was small, with the uncertainty range given in Table 3 encompassing most points. Given TC<sub>bb</sub> and  $OC_{bb}$ ,  $EC_{bb}$  is the difference, Eqs. (5), (6).

EC<sub>ff</sub> is derived simply as the difference between EC and EC<sub>hh</sub> (Eq. 7).

#### Estimation of OC<sub>pbc</sub> from plant debris, c.f. Eq. (8)

Similar to Gelencsér et al. (2007) and Sánchez-Ochoa et al. (2007) the contribution of plant debris to the carbonaceous aerosol is estimated from the measured amount of free cellulose. Free cellulose forms on average about 72% of total cellulose. Further, for the mass balance the amount of plant debris has to be expressed in carbon equivalent, which is about 57% of the total mass. Plant debris is assumed to be about two times the concentration of total cellulose (Puxbaum and Tenze-Kunit, 2003). Thus, measured cellulose concentrations were multiplied by a factor of  $1.6 (= 0.57 \times 2/0.72)$  as a bestestimate of the contribution of OC<sub>phc</sub> on a carbon equivalent basis. As in Gelencsér et al. (2007), a factor of two is used in the uncertainty estimates.

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#### 3.6 Estimation of OC<sub>pbs</sub> from fungal spores, c.f. Eq. (9)

OC<sub>pbs</sub> is calculated from mannitol. Bauer et al. (2008a) found that mannitol accounted for 1.2–2.5 pg spore<sup>-1</sup> and that the spores OC content was 13 pg OC spore<sup>-1</sup> (Bauer et al., 2002). This indicates that OC<sub>pbs</sub> to mannitol ratios are ranging from 5.2–10.8.

A mean mannitol-to-arabitol ratio of  $1.5 \pm 0.5$  can be deduced from the study of Bauer et al. (2008a). The 24 h mean mannitol-to-arabitol ratios observed for PM<sub>10</sub> in summer (Oslo:  $1.4 \pm 0.3$ ; Hurdal:  $1.3 \pm 0.5$ ) in the present study (Table 5) were within the range reported by Bauer et al. (2008a), thus justifying our use of their findings. During winter the ratio was slightly lower (Oslo:  $1.1 \pm 0.3$ ; Hurdal:  $1.3 \pm 0.5$ ) and the variation greater, for unknown reasons. Further, the mannitol-to-arabitol ratio had a diurnal variation in summer, with a higher ratio during night compared to the day. This could indicate influence from various genera of fungi.

# 3.7 Estimation of $F^{14}C$ , $F^{14}C_{bb}$ , $F^{14}C_{spores}$ Eq. (10)

F<sup>14</sup>C is the so-called modern fraction of <sup>14</sup>C, indicating of the amount of contemporary carbon in the aerosol. Fossil-fuels have F<sup>14</sup>C values of zero, and recently grown vegetation has F<sup>14</sup>C values close to 1. A F<sup>14</sup>C value of 1.0 approximately refers to conditions before the atomic bomb tests of the 1950s (Currie et al., 2002). The average <sup>14</sup>C signature of atmospheric CO<sub>2</sub> for the two years (2006, 2007) during which the sampling campaigns of the present study was conducted gives F<sup>14</sup>C = 1.055 (Szidat et al., 2009). The measured value of F<sup>14</sup>C in an aerosol sample, [F<sup>14</sup>C], is one of the most accurately measured quantities in these analyses, but still has a small uncertainty due to both measurement errors and heterogeneity on the filters. We allow for a small error, of  $\pm$  5%, and use an uncertainty factor  $\Phi_{\text{F}^{14}\text{C}}$  (0.95–1.05) and symmetrical beta (2,2) distribution (NIST/SEMATECH, 2010) to assign probabilities around the central value. The <sup>14</sup>C signature of biomass burning emissions (F<sup>14</sup>C<sub>bb</sub>) is generally uncertain, ranging from 1.055 for burning of agricultural residues to a maximum of 1.25 from

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burning of trees (Lewis et al., 2004; Szidat et al., 2006). For plant debris, we have assumed a recent biological origin,  $F^{14}C_{debris} = 1.055$ .

Asco- and Basidiomycota (ABM) are heterotrophic organisms, i.e. they require carbon for growth and energy but cannot fix carbon themselves. ABM can acquire organic molecules from dead organic material, (e.g. dead plant leaves, logs, etc.), so-called decomposers, or as symbionts. Formation of mycorrhiza, which is a symbiotic association between quite a few ABM and the root systems of plants, makes the fungus benefit from photosynthetic products produced by the tree, which has a recent origin; i.e. F<sup>14</sup>C similar to plant debris (1.055). Decomposers may live on old as well as recent substrate, thus in order to account for this we allow for a wider range of F<sup>14</sup>C<sub>spores</sub> (1.055–1.25) than for plant debris.

#### 3.8 Estimation of OC<sub>BSOA</sub>, c.f. Eq. (11)

 $OC_{BSOA}$  is calculated from the radiocarbon balance equation (Eq. 11), assuming modern origin (F<sup>14</sup>C<sub>bio</sub> = 1.055) for this aerosol component. As BSOA is thought to be generated mainy from isoprene and monoterpenes (e.g. Hallquist et al., 2009), and these originate from recent foliage, the radiocarbon signature should be well defined.

#### 3.9 Estimation of $OC_{ff}$ , c.f. Eq. (12)

Finally,  $OC_{ff}$  is defined as the difference between OC and the sum of  $OC_{BSOA}$ ,  $OC_{pbs}$ ,  $OC_{pbc}$  and  $OC_{bb}$ . Unlike Gelencsér et al. (2007) we have not separated  $OC_{ff}$  into primary and secondary as the partitioning theory presented by Robinson et al. (2007) and Donahue et al. (2006) suggests that these two components are extremely difficult to distinguish in principal. In addition, calculations at the end of the chain of equations given above (Eqs. 1–12) are the most uncertain.

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#### 4.1 PM<sub>10</sub> and PM<sub>1</sub> mass concentrations

Mean concentrations of  $PM_{10}$  and  $PM_1$  are presented in Table 4. These values are similar to annual mean concentration ranges of  $PM_{10}$  and  $PM_{2.5}/PM_1$  previously reported for Norwegian urban and rural environments (Yttri, 2007). The rather short duration of the two sampling periods conducted in the present study could influence their representativeness; e.g. summer time concentrations are found to be higher than or equal to the concentrations observed during winter, which is opposite of that commonly seen (Yttri, 2007).  $PM_1$  accounted for a larger fraction of  $PM_{10}$  at the rural background site compared to the urban background site. Also, the  $PM_1$ -to- $PM_{10}$  ratio was higher in winter compared to summer.

#### 4.2 EC and OC concentrations

The carbonaceous aerosol (here: EC,  $OC_p$ ,  $TC_p$ ) concentrations (see Table 4) observed at the urban background site is in the lower range of that previously reported for Norwegian urban areas (Yttri et al., 2005, 2009), whereas it is in the upper range of what has been observed for the rural environment (Yttri et al., 2007a). The levels of  $OC_p$  and  $TC_p$  should be considered as conservative estimates, as the QBQ sampling approach account for the positive but not the negative sampling artefact of OC (see Sect. 2.2). The mean positive artifact of OC ranged from  $18 \pm 8.6\%$  for the urban background site in winter to  $50 \pm 14\%$  at the rural background site in winter.

The difference between the urban- and the rural background site with respect to the  $OC_p$  level is relatively small in summer (25–30% higher at the urban background site), whereas the difference was substantially larger (a factor 2.4–2.8 higher at the urban background site) in winter. EC had a similar pattern, with urban background levels being 50% higher compared to the rural background site in summer and a factor of approximately 3 higher in winter.

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Levels of OC<sub>n</sub> in PM<sub>10</sub> were higher in summer than during winter at the urban background site, whereas it is opposite for PM<sub>1</sub>. For EC, levels were higher in winter for both size fractions. At the rural background site, levels of OC<sub>p</sub> were substantially higher in summer compared to winter. This seasonal variation was more pronounced for PM<sub>10</sub> than for PM<sub>1</sub>. For EC, a similar pattern as for OC<sub>p</sub> was observed for PM<sub>10</sub>, whereas for PM₁ the levels remained unchanged.

Only minor differences in the EC/TC<sub>p</sub> ratio were observed between the two sites, regardless of season and size fraction. The EC/TC<sub>p</sub> ratio was found to be substantially higher in winter (27-31%) compared to summer (16-21%) at both sites and for both size fraction.

#### **Organic tracers concentrations**

Ambient mean concentrations of the wood burning tracer levoglucosan are reported in Table 4. The mean concentration observed at the urban background site in winter (130 ng m<sup>-3</sup>) was lower than that previously reported by Yttri et al. (2005) for Norwegian urban areas during winter time sampling (i.e. 166-407 ng m<sup>-3</sup>). At the rural background site, the winter time mean levoglucosan concentration (40 ng m<sup>-3</sup>) was 2-3 times higher than that reported by Yttri et al. (2007b) for the rural background site Birkenes in Southern Norway. The levoglucosan concentration was 3-5 times higher in winter compared to summer.

Levels of sugars and further sugar alcohols (see Table S1 of the Supplementary material) were within the range previously reported for Scandinavian and continental rural (Carvalho et al., 2003; Ion et al., 2005; Yttri et al., 2007b) and urban background (Yttri et al., 2007b) environments in Europe. Higher concentrations in summer compared to winter were observed for all species. In summer, the highest concentrations were seen at the rural background site (except for ribose), whereas it was the opposite way around in winter. With the exception of ribose and fructose, close to 90% of the sugars and sugar-alcohols observed in summer could be attributed to the coarse fraction of

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 $PM_{10}$ . A similar calculation could not be performed for the samples collected during the winter campaign.

The content of cellulose in the ambient aerosol has been reported in a very few studies only (Kunit and Puxbaum, 1996; Puxbaum and Tenze-Kunit, 2003; Sánchez-Ochoa et al., 2007). The cellulose concentrations observed in the present study, ranging from 5–130 ng m<sup>-3</sup>, are comparable to those observed for the European rural background environment by Sánchez-Ochoa et al. (2007), as is the seasonal variation with increased concentrations in summer.

# 4.4 Radiocarbon, F<sup>14</sup>C

The F<sup>14</sup>C values are reported in Table 4. We find that F<sup>14</sup>C was higher at the rural background site compared to the urban background site, regardless of season and size fraction. In addition, F<sup>14</sup>C was typically higher in summer compared to winter at both sites. The F<sup>14</sup>C values show no diurnal variation, except for PM<sub>1</sub> at the urban background site in winter; i.e. F<sup>14</sup>C was 0.70 during the night and 0.59 during the day. F<sup>14</sup>C appears to be in the upper range of, or higher than, values reported for European and US urban and rural areas by Glasius et al. (2011).

#### 4.5 Source apportionment analysis

The best estimate concentration, the 10th and the 90th percentile of the carbonaceous fractions in  $PM_{10}$  and  $PM_{1}$ , as calculated by Latin Hypercube Sampling (LHS) are presented in Fig. 1 and Tables S2–S5 of the supplementary material, whereas the relative contributions of these concentrations to  $TC_p$  are shown in Tables 6–9. These results are discussed in detail below.

The results of the source-apportionment analysis, presented in Fig. 1 and Tables 6–9 (also Tables S2–S4) show broad scale features similar to those found in previous studies: large contributions of wood-burning in winter time and large contributions of BSOA in summertime. We discuss the major components in Sects. 5.1–5.5 and in Sect. 5.6 we present an estimate of the contribution of the organic aerosol components to the total PM<sub>1</sub> and PM<sub>10</sub> mass concentrations.

#### 5.1 Carbonaceous aerosol from fossil-fuel sources and biomass burning

OC originating from fossil-fuel sources ( $OC_{ff}$ ) was more abundant than OC from wood burning ( $OC_{bb}$ ) during summer, whereas it was the other way around in winter (see Tables S2–S5). Further, there was an urban background increment with respect to both  $OC_{ff}$  and  $OC_{bb}$  compared to the rural background site: i.e. the mean concentration of  $OC_{ff}$  and  $OC_{bb}$  was 1.8–3.5 times higher at the urban background site than at the rural background site, both seasons and size fractions included.

We find that there is almost no difference in the relative contribution of  $OC_{ff}$  (26–29%) and  $OC_{bb}$  (33–38%) to  $TC_p$  in winter between the two sites, both size fractions included, and that wood burning was the major source. It might be that the rather high ambient temperature observed during the winter campaign (i.e. 2.2°C compared to the long term mean of -1.5°C at the urban background site) had an influence on the levoglucosan concentration, which was relatively low compared to previously reported studies (Yttri et al., 2005). In summer, the relative contribution of  $OC_{ff}$  to  $TC_p$  was higher than that of  $OC_{bb}$  to  $TC_p$  at both the urban ( $OC_{ff}$  to  $TC_p = 20-27\%$ ;  $OC_{bb}$  to  $TC_p = 12-16\%$ ) and the rural background site ( $OC_{ff}$  to  $TC_p = 7-12\%$ ;  $OC_{bb}$  to  $TC_p = 4-5\%$ ).

Combustion of fossil fuel was the major source of EC regardless of site, season, size fraction and time of the day, accounting for 9–18% of  $TC_p$ . Further, there was an urban background increment for the EC<sub>ff</sub> and EC<sub>bb</sub> concentration compared to the rural

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background site, ranging from 1.3–3.2. As expected, the concentrations of EC<sub>bb</sub> grew larger in winter due to increased emissions from residential wood burning, accounting for 30-40% of EC compared to 10-20% in summer. A similar seasonal variation was reported by Szidat et al. (2006) for an urban background site in Zurich, using a combination of <sup>14</sup>C and organic tracer analysis, however the EC<sub>bb</sub> contribution to EC was slightly less in Zurich than is Oslo, i.e.  $6 \pm 2\%$  in summer and  $25 \pm 5\%$  in winter.

EC<sub>bb</sub> and OC<sub>bb</sub> concentrations typically increased somewhat during night, which in the present study was defined from 9 a.m.-9 p.m. Thus, the night only partly reflects the time period when people usually use their wood stoves for heating, i.e. between 5 a.m. and 11 a.m. and to some extent between 6 p.m. and 9 p.m. (Haakonsen and Kvingedal, 2001). This may explain why the diurnal variation was not more pronounced. Somewhat surprising the diurnal variation was most evident during summertime sampling. It is not obvious what caused this, but reasons might include larger day/night changes in dispersion conditions, or shifts in the timing of residential or barbeque-related woodburning emissions.

# 5.2 Biogenic secondary organic aerosol, OC<sub>BSOA</sub>

OC<sub>BSOA</sub> was a major contributor to the carbonaceous aerosol in summer, accounting for 56% of  $TC_p$  in the  $PM_{10}$  fraction at the rural background site and 36% at the urban background site. For PM<sub>1</sub>, OC<sub>BSOA</sub> was even more pronounced, constituting 68% of TC<sub>p</sub> at the rural background site and 38% at the urban background site. Summer time OC<sub>BSOA</sub> concentrations ranging from 1.6–2.3 µg C m<sup>-3</sup> was observed for PM<sub>10</sub>, whereas the range was 1.1–2.0 μg C m<sup>-3</sup> for PM<sub>1</sub>. A profound diurnal and seasonal variation was observed, with OC<sub>BSOA</sub> concentrations being 1.4-1.5 times higher during daytime sampling compared to nighttime sampling, and concentrations being 1.4-1.8 times higher at the rural background site compared to the urban background site. In winter, no more than 10% of TC<sub>n</sub> could be attributed to OC<sub>RSOA</sub>, and the concentrations were substantially lower (4-11 times) than those observed during summer. The diurnal variation was also evident in winter, while OC<sub>RSOA</sub> concentrations were found

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to be somewhat higher at the urban background site than at the rural background site in winter. The latter effect is presumably due to the increased condensational fraction of BSOA compounds that can arise in areas of high total OA concentrations (e.g. Donahue et al., 2009).

#### 5 5.3 Primary biological aerosol particles OC<sub>PBAP</sub>

Fungal spores and plant debris are likely the major sources of OC<sub>PBAP</sub>. In the present study, OC associated with fungal spores (OC<sub>pbs</sub>) and plant debris (OC<sub>pbc</sub>) was calculated based on the aerosol filter samples content of mannitol and cellulose. respectively. The results show that  $OC_{PBAP}$  (here:  $OC_{PBAP} = OC_{pbs} + OC_{pbc}$ ) comprised on average 16% of TC<sub>p</sub> in PM<sub>10</sub> at the urban background site and 24% of TC<sub>p</sub> in PM<sub>10</sub> at the rural background site in summer.  $OC_{pbs}$  and  $OC_{pbc}$  contributed approximately equally to TC<sub>p</sub> at the urban background site, whereas the fungal spore contribution was almost three times higher than that of plant debris at the rural background site. In winter, the OC<sub>PBAP</sub> contribution to TC<sub>p</sub> was substantially lower than in summer. A higher relative contribution of OC<sub>PBAP</sub> to TC<sub>p</sub> at the rural site (7.8%) compared to the urban site (2.6%) was consistent with the pattern observed in summer. One major difference observed during winter compared to summer was that plant debris totally dominated OC<sub>PBAP</sub> at the rural background site. For PM<sub>1</sub>, the relative contribution of OC<sub>PBAP</sub> to TC<sub>p</sub> did not exceed 2% regardless of season and site. From the results presented in Table 4 we find that  $OC_{PBAP}$  constituted 72% of  $TC_{p,PM_{10-1}}$  (1.3 µg C m<sup>-3</sup>) at the rural background site in summer, which we consider to be a very high share.  $OC_{PBAP}$  comprised a considerable contribution (44%) to  $TC_{p,PM_{10-1}}$  (1.6  $\mu$ g C m<sup>-3</sup>) at the urban background site as well. The abundance of  $OC_{PBAP}$  in the Scandinavian rural background environment, its pronounced seasonal variation, as well as its presence in the coarse fraction of PM<sub>10</sub> have previously been addressed and reported by Yttri et al. (2007a,b) and Bauer et al. (2008b). However, this is the first time a quantitative measurement-based estimate of the OC<sub>PBAP</sub> fraction has been made for this region.

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Our results confirm that OC<sub>PBAP</sub> is an important source of the carbonaceous aerosol both in the rural and the urban background environment.

As OC<sub>PRAP</sub> typically reside in the coarse fraction of PM<sub>10</sub>, local sources can have a relatively large impact on concentrations. The apparent similarity of PBAP seen for Oslo and Hurdal is misleading: there are great differences in the relative composition of OC<sub>PBAP</sub> with respect to OC<sub>pbs</sub> and OC<sub>pbc</sub> at the two sites (see above), implying that different source types contribute to  $OC_{PBAP}$  at the two sites. This is further supported by the diurnal and seasonal variation of the individual sugars and sugar-alcohols (Table S1). For example, arabitol, mannitol and trehalose in PM<sub>10</sub> all peak during the night at the rural background site in summer, which likely reflects nocturnal discharge of fungal spores (Graham et al., 2003; Ion et al., 2005), while this is not observed at the urban background site. We also find that the individual concentrations of all sugars and sugar-alcohols are higher at the urban background site than for the rural background site in winter.

#### 5.4 Natural versus anthropogenic sources of the carbonaceous aerosol

Carbonaceous aerosols arising from fossil-fuel sources and biomass combustion (EC<sub>bb</sub>, EC<sub>ff</sub>, OC<sub>bb</sub> and OC<sub>ff</sub>) are here defined as anthropogenic emissions. Wild fire emissions can obviously be categorized as natural in cases when ignited by lightening, but most incidences are due to human activity (Achard et al., 2008; Winiwarter et al., 1999). However, as noted in Sect. 3.1 no nearby fire-activity was detected by MODIS for this summertime measurement period. OC<sub>RSOA</sub> and OC<sub>PBAP</sub> are defined as natural sources, although anthropogenic emissions facilitate the transformation of biogenic VOCs to OC<sub>BSOA</sub> (Tsigaridis and Kanakidou, 2003; Carlton et al., 2010; Donahue et al., 2009) and increased concentrations of OCPRAP has been found in association with sewage plants and are associated with other anthropogenic activities as well, e.g. house demolition and agricultural activities (Matthias-Maser, 1998, and references therein).

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As expected, emissions from natural sources were found to be particularly abundant in summer, and with a more pronounced influence of natural sources at the ruralcompared to the urban background site. At the rural background site, 80% of TC<sub>n</sub> in PM<sub>10</sub> and 70% of TC<sub>p</sub> in PM<sub>1</sub> could be attributed to natural sources. The slightly lower percentage seen for PM<sub>1</sub> is due to OC<sub>PBAP</sub> primarily residing in the coarse fraction of  $PM_{10}$ . Natural sources accounted for about half (51%) of  $TC_p$  in  $PM_{10}$  at the urban background site as well. As for the rural background site, the relative contribution of natural sources to TC<sub>p</sub> in PM<sub>1</sub> (39%) at the urban background site was slightly less than for PM<sub>10</sub>, but still quite substantial taken into account that it is an urban site. The natural source contribution was always dominated by OC<sub>BSOA</sub>, regardless of season, site and size fraction. During winter the picture was reversed, as anthropogenic sources totally dominated the carbonaceous aerosol (83–90%), whereas no more than 10–17% could be explained by natural sources. Combustion of biomass contributed slightly more than combustion of fossil fuel in winter, whereas emissions from fossil fuel combustion were more abundant in summer.

#### Consistency of results

The challenges associated with splitting OC<sub>PBAP</sub>, OC<sub>bb</sub> and OC<sub>BSOA</sub> have already been mentioned (see Sect. 3.3). As we have calculated the two major contributors to PBAP, i.e. OC associated with plant debris and fungal spores, the dominating uncertainty factors for OC<sub>BSOA</sub> have been accounted for. On the other hand, the vast number of sources contributing to this highly heterogeneous group still makes the split between OC<sub>BSOA</sub> and OC<sub>PBAP</sub> somewhat uncertain. For example, Ceburnis et al. (2011) recently demonstrated a major impact of fine mode PBAP derived from plankton from the marine environment at Mace Head on the West coast of Ireland, which is not accounted for by the PBAP tracers used in the present study. Tire debris is also a source of coarse mode contemporary carbon not accounted for in our source apportionment. Although, natural rubber only constitutes 0.5–3.5% of the tire's tread (Edeskär, 2004) a certain contribution of contemporary carbon should be expected.

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Emissions from cooking could make a significant contribution to contemporary carbon at the urban background site, while we find this less likely at the rural background site. Analysis of cholesterol from the filters could have provided valuable information on this matter, but such measurements have not been undertaken.

Nevertheless, the seasonality, the diurnal variation, and the regional character observed for  $OC_{BSOA}$  strongly support that BSOA has been quite successfully separated from other sources of contemporary carbon.

#### 5.6 Mass closure of PM

Converting the ambient aerosol OC fraction to organic matter (OM) with a high level of accuracy requires detailed speciation of OC at a level which currently is not available. Further, using one fixed conversion factor will inevitably introduce an unknown level of uncertainty, as the source contribution varies between sites and seasons. By segregating OC into sub-fractions, and by using conversion factors for each of these, more accurate estimates of the aerosol OM fraction may be obtained. However, experimentally derived conversion factors are still scarce and would be needed for a wider range of environments to reduce the uncertainty further.

Reviewing theoretic and historic experimental conversion factors, Turpin and Lim (2001) concluded that a range of 1.9–2.3 would cover the aged aerosol, whereas 2.2–2.6 would be representative for the aerosol originating from biomass burning. A factor of 1.2–1.5 was suggested for water insoluble carbonaceous material. Their estimate for the aged aerosol has been supported by the experimentally derived conversion factor for the rural background site K-puszta in Hungary, ranging from 1.9–2.0 (Kiss et al., 2002), whereas a conversion factor of 1.1 was derived for EC. Recently, OC:OM ratios derived from aerosol mass spectrometer (AMS) measurements have become available, which have confirmed conversion factors around 2 for the aged aerosol and the biomass burning aerosol (e.g. Lanz et al., 2008). AMS measurements have also reported OC:OM ratios of 1.2 for the hydrogenated organic aerosol (HOA) fraction likely to originate from traffic (Lanz et al., 2008). In the present study we have used

a conversion factor of 2.0 for OC from wood burning ( $OC_{bb}$ ) and for SOA. Although we have preferred not to split  $OC_{ff}$  into primary and ASOA contributions through this text, such a split does exist in the LHS processing, with  $OC_{ASOA}$  estimated to contribute between 41 and 69% of  $OC_{ff}$ . We accordingly use a factor 2 for the ASOA fraction, and a factor 1.2 for the primary fraction of  $OC_{ff}$ . A conversion factor of 1.75 was used for  $OC_{pbs}$  and  $OC_{pbs}$ , and 1.1 for  $EC_{bb}$  and  $EC_{ff}$ .

Figures 2 and 3 illustrate the results of this mass closure estimates. These show that that particulate carbonaceous matter, i.e. OM + EC, likely dominated the mass concentration of the ambient PM regardless of size fraction, season, and site category. In general, there were no pronounced differences in the relative contribution of carbonaceous matter to PM with respect to season or between the two sites. A somewhat larger fraction of  $PM_1$  (64–69%) was accounted for by carbonaceous matter compared to  $PM_{10}$  (51–67%). The likely explanation for this similarity is simply that Oslo is a relatively small city (ca. 0.6 million inhabitants). Both sites are strongly influenced by long-range transport, providing a common background, and local activities such as wood-burning for heating take place inside Oslo as well as in the countryside affecting Hurdal.

The substantial contribution of carbonaceous aerosol from natural sources to the PM loading in summer should be emphasized. We found that about 30% of the urban background PM<sub>1</sub> and PM<sub>10</sub> could be attributed to natural sources, whereas for the rural background site the contribution was 52–57%. In, winter no more than 7–11% of the PM loading could be attributed to natural sources. Due to the rather low PM experienced during this wintertime sampling at the urban background site (mean PM<sub>10</sub> mass concentration of 10.3  $\mu$ g m<sup>-3</sup>, compared to 19.1 ± 4.0  $\mu$ g m<sup>-3</sup> for all Norwegian urban background sites for 2009–2010), we assume that the contribution of natural sources to PM is in the upper range of what can be expected in winter.

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Source apportionment of the ambient summer- and winter time carbonaceous aerosol (PM<sub>10</sub> and PM<sub>1</sub>) has been conducted for one urban and one rural background environment site in Norway. Statistical treatment of data from thermal optical, <sup>14</sup>C and organic tracer analysis using Latin Hypercube Sampling has allowed for quantitative estimates of seven different sources contributing to the ambient carbonaceous aerosol. These are: elemental carbon from combustion of biomass (EC<sub>bb</sub>) and fossil fuel (EC<sub>ff</sub>), organic carbon from combustion of biomass (OChh) and fossil fuel (OCff, which includes both primary and secondary components), primary biological aerosol particles (OC<sub>PBAP</sub>) from plant debris (OC<sub>pbc</sub>) and fungal spores (OC<sub>pbs</sub>), and secondary organic aerosol from biogenic precursors (OC<sub>BSOA</sub>). Hence, this approach makes it possible to separate between natural and anthropogenic sources, which is highly important in order to sort out abatement strategies for reducing man-made emissions.

The results show that particulate carbonaceous matter (PCM) dominated (51–69%) the ambient PM loading at these Norwegian urban and rural background sites, regardless of season (summer/winter) and size fraction (PM<sub>10</sub>/PM<sub>1</sub>).

Natural sources (here: BSOA and PBAP) contributed substantially to the rural (70-80%) and the urban background (39–51%) of total carbon (TC<sub>n</sub>) loading during summer for both size fractions. The natural contribution was dominated by BSOA both for the rural (56–68% of TC<sub>n</sub>) and the urban background environment (36–38% of TC<sub>n</sub>), but also primary biological aerosol particles (PBAP) were found to make a noticeable contribution, (e.g. PBAP accounted for 24% of TC<sub>p</sub> in PM<sub>10</sub> at the rural background site).

Anthropogenic sources (EC<sub>ff</sub>, EC<sub>bb</sub>, OC<sub>ff</sub>, OC<sub>bb</sub>) contributed the most (> 80%) to TC<sub>b</sub> during winter, regardless of size fraction and environment. Fossil-fuel derived sources were the major anthropogenic contribution to TC<sub>p</sub> in summer, whereas there was a shift to wood burning in winter.

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The results reported in the present study are highly complementary to those of Gelencsér et al. (2007) and Szidat et al. (2009), applying the same software/methodology, but updated for Norwegian conditions. Together they generate a picture of BSOA being the major contributor to the rural background carbonaceous aerosol during summer, not only for Continental Europe, but also for its northern parts. In winter, BSOA levels are low, and as in parts of Central Europe wood-burning makes the largest contribution. The suggested abundance of BSOA in Scandinavian winter time aerosol needs further investigation, in particular with respect to the potential confounding factors associated with compounds from wood burning, and to what extent long range transport and/or increased condensation of BSOA can contribute to the observed sources of modern carbon.

Finally, we would emphasize that use of source specific organic tracers, <sup>14</sup>C and elemental analysis, combined with source apportionment and/or mass-balance approaches provide a much firmer basis for model evaluation than is possible using EC and/or OC measurements alone. The extra information enables evaluation of each component of the model system (emissions, separation of wood-burning from fossilfuel sources, SOA formation, etc.). Such data and model evaluation are urgently needed before we can claim to understand the carbonaceous aerosol in the atmosphere. These aerosol phase measurements should be supported by simultaneous measurements of the likely biogenic and anthropogenic gas-phase precursors of the secondary carbonaceous aerosol, including their formation and degradation products.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/11/7375/2011/ acpd-11-7375-2011-supplement.pdf.

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**Table 1.** Notation used in this study.

$OC_F$	Measured organic carbon on front filter
OC <sub>B</sub>	Measured organic carbon on back filter
$OC_p$	Organic carbon after correction for positive artefact, $OC_p = OC_F - OC_B$
TC <sub>p</sub>	Total carbon after correction for positive artefact, $TC_p = OC_p + EC$
LG	Levoglucosan
OC	Organic carbon
OA	Organic aeorosol (includes other elements, eg O, H, N)
POA	OA arising from primary emissions
ASOA	OA arising from secondary oxidation of anthropogenic precursors
BSOA	OA arising from secondary oxidation of biogenic precursors (mainly terpenes, isoprene)
PBAP	Primary biological aerosol particles, includes fungii, plant debris, virus, etc.
Cel.	Free cellulose, as measured
PCM	Particulate carbonaceous matter
Subscript bb	Associated with biomass burning
Subscript ff	Associated with fossil-fuels (includes POA and ASOA for OC)
Subscript pbs	PBAP associated with sugars, eg fungii, etc.
Subscript pbc	PBAP associated with cellulose, eg plant debris
Subscript ASOA, BSOA	Associated with ASOA, BSOA
F <sup>14</sup> C	Fraction of modern carbon relative to a reference standard (Reimer et al., 2004)
	(without subscript, F <sup>14</sup> C refers to whole TC sample, otherwise subscript indicates origin)
$\Phi_{FC}$	Uncertainty factor for EC observations
Φ <sub>NA</sub>	Uncertainty factor to account for negative sampling artefact of OC
Φ <sub>F14</sub> C	Uncertainty factor for F <sup>14</sup> C observations

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**Table 2.** Basis for source-apportionment.

$EC = [EC] \times \Phi_{EC}$	(1)	
$OC = [OC_p] + \Phi_{NA} \times (OC_F - OC_B)$	(2)	
TC = OC + EC	(3)	
$TC_{bb} = [LG] \times (TC/LG)_{bb}$	(4)	
$OC_{bb} = TC_{bb} \times (OC/TC)_{bb}$	(5)	
$EC_{bb} = TC_{bb} - OC_{bb}$	(6)	
$EC_{ff} = EC - EC_{bb}$	(7)	
$OC_{pbc} = [Cel.] \times (OC_{pbs}/Cel.)$	(8)	
$OC_{pbs} = [Mannitol] \times (OC_{pbs}/Mannitol)$	(9)	
$F^{14}C = [F^{14}C] \times \Phi_{F^{14}C}$	(10)	
$OC_{\rm BSOA} = ([TC] \times F^{14}C - TC_{\rm bb} \times F^{14}C_{\rm bb} - OC_{\rm pbs} \times F^{14}C_{\rm spores} - OC_{\rm pbc} \times F^{14}C_{\rm debris})/F^{14}C_{\rm bio}$	(11)	
$OC_{ff} = OC - (OC_{bb} + OC_{pbs} + OC_{pbc} + OC_{BSOA})$	(12)	
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Notes: Square brackets denote measured quantities. Parentheses denote emission ratios, see Table 3. See also Table 1 for notation.

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**Table 3.** Summary of uncertainty factors used in the LHS analysis. Low, central and high values are listed.

Parameters	Low	Central <sup>a</sup>	High	Equation	Comment
Φ <sub>EC</sub>	0.75	1.0	1.25	(1)	Uncertainty in EC
$\Phi_{NA}$	0.0	0.2	1.0	(2)	Artefact uncertainty, Sect. 3.2
(TC/LG) <sub>bb</sub> , PM <sub>10</sub>	11	15	17	(4)	Ratio in emissions
$(TC/LG)_{bb}$ , $PM_{2.5}$	7.6	12	14		Ratio in emissions
$(OC/TC)_{bb}$ , $PM_{10}$	0.73	0.78	0.82	(5)	Ratio in emissions
$(OC/TC)_{bb}$ , $PM_{2.5}$	0.66	0.71	0.76		Ratio in emissions
OC <sub>pbc</sub> /Cel.	8.0	1.6	3.2		See Sect. 3.5
OC <sub>pbs</sub> /Mannitol	5.2	_	10.8	(9)	See Sect. 3.6
Ф <sub>F<sup>14</sup>C</sub>	0.95	1.0	1.05	(10)	Uncertainty in F <sup>14</sup> C, Sect. 3.7
$F^{14}C_{bb}$	1.055	_	1.25	(11)	See Sect. 3.7
F <sup>14</sup> C <sub>spores</sub>	1.055	-	1.25	(11)	See Sect. 3.7
F <sup>14</sup> C <sub>debris</sub>		1.055		(11)	See Sect. 3.7

<sup>&</sup>lt;sup>a</sup> Where no central value is given, the value used is simply the mean of the low and high values.

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Table 4. Measured concentrations of aerosol components.

Size	Site	Season	Period	PM μg m <sup>-3</sup>	ΟC μg C m <sup>-3</sup>	$\mathrm{OC_p}\ \mu\mathrm{g}\mathrm{C}\mathrm{m}^{-3}$	EC μg C m <sup>-3</sup>	$TC_p$ $\mu g  C  m^{-3}$	F <sup>14</sup> C frac	Levo. ng m <sup>-3</sup>	Mannitol ng m <sup>-3</sup>	Cellulose µg m <sup>-3</sup>
PM <sub>10</sub>	U	S	Day	14.8	4.04	3.15	0.68	3.83	0.73	29	24	0.18
PM <sub>10</sub>	U	S	Night	16.1	3.56	2.82	0.74	3.56	0.72	53	26	0.069
PM <sub>10</sub>	U	S	24h	15.4	3.8	2.98	0.71	3.70	0.73	40	25	0.13
PM <sub>10</sub>	R	S	Day	11.4	3.76	2.36	0.45	2.81	_	8.3	28	-
PM <sub>10</sub>	R	S	Night	11.3	3.64	2.54	0.49	3.03	_	12	64	_
PM <sub>10</sub>	R	S	24h	11.3	3.70	2.45	0.47	2.92	0.93	10	45	0.066
PM <sub>10</sub>	U	W	Day	10.8	3.18	2.53	0.99	3.52	0.59	118	3.2	0.026
$PM_{10}$	U	W	Night	9.88	2.38	1.98	0.86	2.85	0.63	142	2.6	0.034
$PM_{10}$	U	W	24 h	10.3	2.78	2.26	0.93	3.18	0.61	130	2.9	0.03
$PM_{10}$	R	W	Day	5.53	1.58	1.02	0.30	1.32	_	51	0.78	-
$PM_{10}$	R	W	Night	4.46	1.09	0.69	0.29	0.99	-	53	0.53	-
$PM_{10}$	R	W	24 h	4.99	1.34	0.86	0.30	1.15	0.68	52	0.65	0.043
$PM_1$	U	S	Day	7.31	2.67	1.83	0.34	2.17	0.65	29	(a)	0.006
PM₁	U	S	Night	7.87	2.33	1.72	0.58	2.29	0.66	53	(a)	0.005
PM₁	Ū	S	24 h	7.60	2.50	1.77	0.46	2.23	0.65	40	1.5	0.0055
PM₁	R	S	Day	7.67	2.84	1.44	0.28	1.72	_	8.3	_	_
PM₁	R	S	Night	6.83	2.36	1.36	0.26	1.62	_	12	_	_
PM₁	R	S	24 h	7.66	2.60	1.40	0.27	1.67	0.83	10	2.1	0.005
PM₁	U	W	Day	8.04	2.33	2.02	0.81	2.83	0.59	118	(a)	0.01
PM₁	Ū	W	Night	7.48	2.04	1.67	0.76	2.43	0.70	142	(a)	0.002
PM <sub>1</sub>	Ü	W	24 h	7.76	2.19	1.84	0.78	2.63	0.64	130	1.3	0.006
PM₁	Ř	W	Day	4.50	1.67	0.79	0.23	1.04	_	51	_	_
PM₁	R	W	Night	4.14	0.97	0.76	0.30	1.06	_	53	_	_
PM <sub>1</sub>	R	W	24 h	4.32	1.32	0.77	0.27	1.05	0.70	52	1.8	0.012

Notes: Sites are (U)rban: Oslo, (R)ural: Hurdal; Seasons are (S)ummer = 19 Jun-15 Jul 2006, (W)inter = 1 Mar-8 Mar 2007.

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<sup>(</sup>a) Day and night mannitol not measured, but assumed equal to 24 h value for LHS analysis. (Values are very small, so have little impact on the analysis.)

**Table 5.** Mannitol-to-arabitol ratios for PM<sub>10</sub>.

	24 h	Day	Night
Oslo Summer	1.4 ± 0.3	1.3 ± 0.2	1.5 ± 0.4
Winter Hurdal	$1.3 \pm 0.5$	$1.3 \pm 0.5$	$1.3 \pm 0.5$
Summer Winter	$1.5 \pm 0.3$ $1.1 \pm 0.3$	$1.2 \pm 0.2$ $1.1 \pm 0.2$	$1.7 \pm 0.3$ $1.1 \pm 0.5$

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**Table 6.** Calculated pecentage contributions to total carbon from LHS analysis, PM<sub>10</sub>, Summer.

	Hur-19 Jun-15 Jul-24 h		Osl-19 Jun-15 Jul-24 h		Osl-19 Jun-15 Jul-Day		Osl-19 Jun-15 Jul-Night	
	B.E.	Range	B.E.	Range	B.E.	Range	B.E.	Range
EC <sub>bb</sub>	0.90	(0-2)	2.80	(1–4)	1.90	(0-3)	3.90	(1–6)
$EC_{ff}$	8.90	(6–11)	14.20	(8–19)	13.80	(8–18)	14.50	(8–20)
$OC_{bb}$	3.60	(2-4)	11.70	(7–15)	8.20	(5-10)	16.20	(10-20)
$OC_{\rm ff}$	7.00	(4–9)	19.90	(14-25)	19.70	(14-25)	21.10	(14–27)
$OC_{BSOA}$	55.90	(48-62)	35.60	(28-42)	40.50	(33-46)	29.60	(22-37)
$OC_{PBAP}$	23.70	(17–29)	15.80	(11–20)	15.90	(11–21)	14.70	(10–18)
$OC_{obs}$	18.00	(12-22)	7.40	(5–9)	5.90	(4–7)	9.20	(6-11)
$OC_{pbc}$	5.60	(3–9)	8.40	(4–13)	10.00	(5–16)	5.50	(2-8)

B.E. is best estimate (50th percentile), range is 10th-90th percentiles of LHS results.

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**Table 7.** Calculated pecentage contributions to total carbon from LHS analysis, PM<sub>10</sub>, Winter.

	Hur-1 Mar-8 Mar-24 h		Osl-1 Mar-8 Mar-24 h		Osl-1 Mar-8 Mar-Day		Osl-1 Mar-8 Mar-Nigh	
	B.E.	Range	B.E.	Range	B.E.	Range	B.E.	Range
EC <sub>bb</sub>	8.10	(3–13)	7.80	(3–12)	7.20	(2–12)	8.80	(3–13)
EC <sub>ff</sub>	14.60	(5-22)	18.10	(8-27)	17.80	(8-26)	17.80	(7-27)
$OC_{bb}$	34.40	(26-42)	32.80	(25-40)	30.70	(22-39)	35.80	(29-42)
$OC_{ff}^{ss}$	25.70	(17–34)	28.60	(19–38)	30.40	(21–40)	27.20	(17–37)
OC <sub>BSOA</sub>	9.50	(1–18)	10.10	(1–18)	11.60	(1–22)	7.50	(1–14)
OC <sub>PBAP</sub>	7.80	(4-12)	2.60	(1–3)	2.40	(1–3)	2.80	(1–4)
OC <sub>pbs</sub>	(a)	(a)	(a)	(a)	(a)	(a)	(a)	(a)
$OC_{pbc}$	7.20	(4–11)	(a)	(a)	(a)	(a)	2.10	(1–3)

B.E. is best estimate (50th percentile), range is 10th-90th percentiles of LHS results.

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 $<sup>^{\</sup>rm (a)}$  Individual values for  ${\rm OC_{pbs}}$ ,  ${\rm OC_{pbc}}$  not shown when < 2%, since LHS precision too low, see text.

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**Table 8.** Calculated pecentage contributions to total carbon from LHS analysis, PM<sub>1</sub>, Summer.

	Hur-19 Jun-15 Jul-24 h		Osl-19 Jun-15 Jul-24 h		Osl-19 Jun-15 Jul-Day		Osl-19 Jun-15 Jul-Nigh	
	B.E.	Range	B.E.	Range	B.E.	Range	B.E.	Range
EC <sub>bb</sub>	1.20	(0-2)	3.80	(1–6)	2.80	(1–4)	5.10	(1–8)
$EC_{ff}$	11.50	(7–15)	14.70	(8-20)	11.40	(6-15)	17.00	(9-24)
OC <sub>bb</sub>	5.10	(3–6)	16.10	(11–20)	11.90	(8-14)	20.80	(15–26)
$OC_{ff}$	12.00	(7–16)	26.60	(20-33)	29.40	(24-34)	23.90	(16–31)
OC <sub>BSOA</sub>	68.40	(66–70)	37.90	(33–43)	43.30	(39–47)	32.50	(26–39)
OC <sub>PBAP</sub>	1.70	(1–3)	0.80	(0–2)	1.30	(0–2)	0.50	(0-2)
OC <sub>pbs</sub>	(a)	(a)	(a)	(a)	(a)	(a)	(a)	(a)
OC <sub>pbc</sub>	(a)	(a)	(a)	(a)	(a)	(a)	(a)	(a)

B.E. is best estimate (50th percentile), range is 10th-90th percentiles of LHS results.

 $<sup>^{\</sup>rm (a)}$  Individual values for  ${\rm OC_{pbs}}$ ,  ${\rm OC_{pbc}}$  not shown when < 2%, since LHS precision too low, see text.

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**Table 9.** Calculated pecentage contributions to total carbon from LHS analysis, PM<sub>1</sub>, Winter.

	Hur-1 Ma	ar–8 Mar-24 h	Osl-1 Mar-8 Mar-24 h		Osl-1 Ma	ar–8 Mar-Day	Osl-1 Mar-8 Mar-Night	
	B.E.	Range	B.E.	Range	B.E.	Range	B.E.	Range
EC <sub>bb</sub>	9.00	(3–14)	8.90	(3–14)	7.90	(3–12)	10.60	(4–16)
EC <sub>ff</sub>	13.30	(4-21)	17.20	(7-26)	17.80	(7-27)	15.80	(6-24)
OC <sub>pb</sub>	38.40	(29-47)	37.00	(29-44)	33.40	(25-41)	41.50	(33–49)
$OC_{ff}$	25.50	(17-34)	27.10	(17-37)	30.60	(21-40)	23.40	(14-33)
OC <sub>BSOA</sub>	10.70	(1–20)	9.00	(1–16)	9.10	(1–17)	8.20	(1–15)
OC <sub>PBAP</sub>	3.20	(2-4)	0.80	(0–2)	1.30	(0-2)	0.50	(0-2)
OC <sub>pbs</sub>	(a)	(a)	(a)	(a)	(a)	(a)	(a)	(a)
OC <sub>pbc</sub>	(a)	(a)	(a)	(a)	(a)	(a)	(a)	(a)

B.E. is best estimate (50th percentile), range is 10th-90th percentiles of LHS results.

 $<sup>^{\</sup>rm (a)}$  Individual values for  ${\rm OC_{pbs}}$ ,  ${\rm OC_{pbc}}$  not shown when < 2%, since LHS precision too low, see text.

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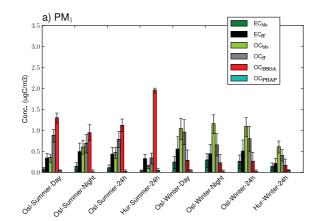
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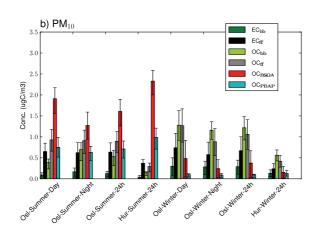
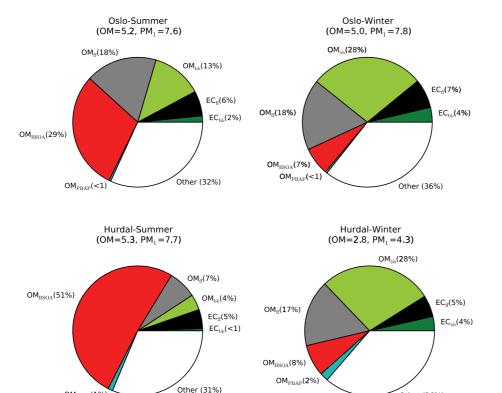


Fig. 1. Best estimate concentrations (μg C/m³) of different carbonaceous particle fractions for the SORGA samples. Uncertainty bars represent 10 and 90 percentiles from LHS calculations.

Other (36%)



**Fig. 2.** Estimated contribution of carbonaceous matter components to  $PM_1$ . Estimates use central values from LHS analysis. Numbers in parenthesis give total aerosol concentrations and OM in  $\mu g \, m^{-3}$ . See Sect. 5.6 for assumed OM: OC ratios.

OM<sub>PBAP</sub>(1%)

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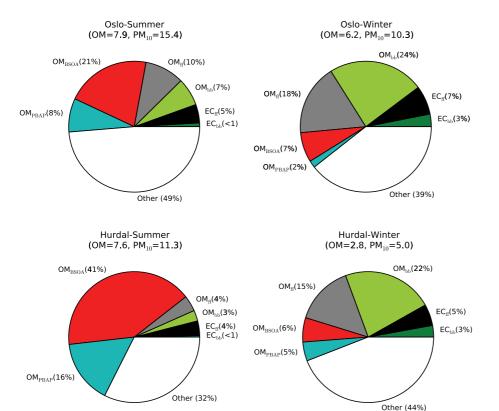




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**Fig. 3.** Estimated contribution of carbonaceous matter components to  $PM_{10}$ . Estimates use central values from LHS analysis. Numbers in parenthesis give total aerosol concentrations and OM in  $\mu g \, m^{-3}$ . See Sect. 5.6 for assumed OM: OC ratios.

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