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**Carbonaceous
aerosols at urban
influenced sites in
Norway**

K. E. Yttri et al.

Carbonaceous aerosols at urban influenced sites in Norway

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Received: 9 July 2008 – Accepted: 17 September 2008 – Published: 17 November 2008

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Little is known regarding levels and source strength of carbonaceous aerosols in Scandinavia. In the present study, ambient aerosol (PM_{10} and $PM_{2.5}$) concentrations of elemental carbon (EC), organic carbon (OC), water-insoluble organic carbon (WINSOC), and water-soluble organic carbon (WSOC) are reported for a curbside site, an urban background site, and a suburban site in Norway in order to investigate their spatial and seasonal variations. Aerosol filter samples were collected using tandem filter sampling to correct for the positive sampling artefact introduced by semi volatile OC. Analyses were performed using the thermal optical transmission (TOT) instrument from Sunset Lab Inc., which corrects for charring during analysis. Finally, we estimated the relative contribution of OC from wood burning based on the samples content of levoglucosan.

Levels of EC varied by more than one order of magnitude between sites, likely due to the higher impact of vehicular traffic at the curbside and the urban background sites. In winter, the level of particulate organic carbon (OC_p) at the suburban site was equal to (for PM_{10}) or even higher (for $PM_{2.5}$) than the levels observed at the curbside and the urban background sites. This finding was attributed to the impact of residential wood burning at the suburban site in winter, which was confirmed by a high mean concentration of levoglucosan (407 ng m^{-3}). This finding indicates that exposure to primary combustion derived OC_p could be equally high in residential areas as in a city center. It is demonstrated that OC_p from wood burning (OC_{wood}) accounted for almost all OC_p at the suburban site in winter, allowing a new estimate of the ratio $TC_p/\text{levoglucosan}$ for both PM_{10} and $PM_{2.5}$. Particulate carbonaceous material (PCM = Organic matter + Elemental matter) accounted for 46–83% of PM_{10} at the sites studied when considering the positive artefact of semi volatile OC, thus being the major contributor to PM.

ACPD

8, 19487–19525, 2008

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1 Introduction

During the last 10–15 years the carbonaceous aerosol has become one of the most intensely studied fields within the atmospheric sciences. This can be attributed to its postulated impacts on the global climate (Novakov and Penner, 1993; Kanakidou et al., 2005) and on human health (McDonald et al., 2004; Metzger et al., 2004). There are a vast number of organic molecules associated with respirable aerosols, which impact on human health has not yet been clarified. Furthermore, studies seem to point towards combustion particles, which are enriched in carbonaceous material, when ascribing the negative health effects on humans following exposure to ambient aerosols (Künzli et al., 2000; Hoek, et al., 2002; Forsberg et al., 2005; WHO, 2003). Likewise, organic molecules are predicted to play a role in cloud formation, once thought to be affected only by the inorganic constituents of the aerosol (Novakov and Corrigan, 1996; Cruz and Pandis, 1997). Along with black carbon, which is the principal light absorbing species in the atmosphere, the carbonaceous aerosol influences the Earth's radiative balance (IPCC, 2001). When studying aerosol impact on climate, the largest uncertainties by far are associated with the effects of the carbonaceous aerosol.

For the reasons mentioned, effort is needed to identify the constituents of the carbonaceous aerosol along with their physical and chemical properties. Further, their origin needs to be clarified in order to differentiate between the anthropogenic and the natural contributions (e.g. Gelencsér et al., 2007). Quantification of the components of OC also allows for a much better evaluation of atmospheric models than simple comparison with total OC allows. For example, Simpson et al. (2007) clearly showed that increased knowledge of the wood-burning (and other) contributions to measured OC levels could explain many apparent discrepancies between modelled and measured wintertime OC. While quantification of single organic molecules can be useful for the purpose of source apportionment, this approach becomes an insurmountable task when it comes to mass closure studies due to the great number of organic molecules associated with the carbonaceous aerosol. Thus, implementation of operational defini-

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tions of bulk carbonaceous material, such as EC (elemental carbon) and OC (organic carbon) has been necessary. Although these definitions have certain advantages, the quality of such data is affected by the use of sampling procedures and analytical techniques that fail to account for significant artefacts.

5 Only a few studies have addressed the ambient aerosols levels of EC and OC in Scandinavia. Zappoli et al. (1999), Ricard et al. (2002) and Yttri et al. (2007a) all presented concentrations observed at rural background sites, while the studies by Viidanoja et al. (2002), Sillanpää et al. (2006), Glasius et al. (2006) and Saarikoski et al. (2007) reported observation from the urban environment. Of these studies, only
10 three accounted for the positive sampling artefacts of OC. Furthermore, various analytical instruments and temperature programs have been used for quantification, making it difficult to compare the results. Thus, additional high quality measurements of EC and OC, covering a broad range of site categories and all seasons, are needed for a better understanding of the carbonaceous aerosol in Scandinavia.

15 The objective of the present study was to quantify the ambient aerosol concentration of EC, OC, WINSOC, and WSOC in PM₁₀ and PM_{2.5}, and to estimate the concentration of OC from residential wood burning based on levoglucosan measurements. Samples were collected at three different sites in Norway, and at different times of the year to reflect different sources and to address the spatial and seasonal variation of the
20 carbonaceous sub-fractions.

2 Experimental

2.1 Site description and measurement period

25 Sampling campaigns were conducted at three sites in Norway; at an urban background site and at a curbside site in the city of Oslo (59°56' N, 10°44' E), and at a suburban site in the small town of Elverum (60°53' N, 11°34' E), 120 km north-east of Oslo. The urban background site is situated in a park in the central part of Oslo, at which sampling

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was performed on weekdays during the period 21 November–14 December 2001. The curbside site, Helsfyr, is located next to one of the most heavily trafficked roads in Oslo (50 000 vehicles per 24 h); here sampling was performed during the period 9 September–3 October 2002. The suburban site in Elverum is located on the outskirts of a residential area. At this site, sampling was conducted both for a winter period (30 January–15 March 2002) and a summer period (22 March–28 June 2002). All sampling inlets were installed approximately 4 m above ground level.

2.2 Aerosol sampling

2.2.1 Aerosol sampling for determination of the carbonaceous content in PM₁₀ and PM_{2.5}

Two different inlets were used for collection of PM₁₀. At the two sites in Oslo and during the winter campaign at the suburban site, a NILU filter-unit with a NILU PM₁₀ inlet was used. For the summer campaign at the suburban site, a NILU filter-unit with a Gent PM₁₀ inlet was used. Both samplers have been proven equal to the CEN 12341 standard (Marsteen and Schaug, 2007).

Two different samplers were used to collect PM_{2.5}. Low volume samplers from Derenda were used at the curbside site and at the suburban site during the summer campaign, whereas a NILU stacked-filter unit (SFU) with a NILU PM₁₀ inlet was used at the urban background site and at the suburban site during the winter campaign. When using a flow rate of 10 l min⁻¹, the NILU-SFU acts as a dichotomous sampler, as the flow through the initial filter (8 μm pore, 47 mm Nucleopore) will result in the collection of particles with an equivalent aerodynamic diameter (EAD) of 2.5 μm on the second filter (Hopke et al., 1997). An overview of the different samplers, including their flow rate and filter face velocity, is shown in Table 1. The sampling time for each sampler was 23 h.

Aerosol sampling was performed using tandem filter set-ups (McDow and Huntzicker, 1990) in order to correct for the positive sampling artefact introduced by semi

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volatile OC. The QBT (quartz fibre filter behind Teflon filter) approach was used for all samplers except for PM₁₀ at the urban background site where the QBQ (quartz fibre filter behind quartz fibre filter) approach was applied. Both the QBT and QBQ approach were used to quantify the positive sampling artefact of OC for PM_{2.5} at the urban background site.

The Teflon filters were purchased from Gelman (2 μm pore, 47 mm Zefluor Teflon, Gelman P5PJ047), whereas quartz fibre filters were purchased from Whatman (Whatman QM-A, 47 mm). All quartz fibre filters used were from the same batch number in order to minimize differences in adsorptive capacity (Kirchstetter et al., 2001). The quartz fibre filters were pre-baked at 850°C for 3.5 h and stored at –30°C until exposed. After sampling the filters were transported back to the laboratory in a mobile freezer (–5°C) and stored in sealed Petri dishes at –30°C to prevent post-collection evaporation. Field blanks were assigned to each fourth day of sampling and were treated in exactly the same manner regarding preparation, handling, transport and storage, as the filters being exposed.

2.2.2 Aerosol sampling for determination of mass concentration

Mass concentrations of PM₁₀ were obtained using a high volume sampler (Andersen Instruments Inc.) collecting aerosols on quartz fibre filters (Whatman QM-A, 8'' × 10''). The instrument was operated at a flow rate of 1.11 m³ min⁻¹. The filters were conditioned at 20°C and 50% RH (relative humidity) for 48 h prior to and after exposure. The sampling time was 23 h.

2.2.3 Aerosol sampling for determination of Potassium

Aerosol filter samples for analysis of water soluble Potassium was collected using a three stage filter pack with a Teflon (2 μm Zefluor) particle front filter followed by a potassium hydroxide impregnated cellulose filter (Whatman 40) and finally an oxalic acid impregnated cellulose filter (Whatman 40) (EMEP, 1995). Potassium was obtained

from the front Teflon filter. The filter pack was operated at flow rate of 16 l min^{-1} . The sampling time was 23 h.

2.3 Thermal optical transmission analysis of EC, OC, TC, WINSOC and WSOC

TC, EC, OC, WSOC and WINSOC, were quantified using the thermal optical transmittance (TOT) instrument from Sunset laboratories Inc., which corrects for charring of OC during analysis (Birch and Cary, 1996). Details concerning the temperature program (quartz.par) used, and its performance when analyzing the “8785 Air Particulate Matter On Filter Media”, is reported by Yttri et al. (2007a) and will not be described in detail here. In brief, an EC/TC ratio of $22.7 \pm 2.1\%$ (Mean ± 2 SD) ($n=3$) was found for the NIOSH derived quartz.par temperature program when analysing the reference material from NIST. This is slightly lower than the certified value for NIOSH STN ($27.9 \pm 2.3\%$) (Mean ± 2 SD), and approximately a factor of two lower than the certified value for IMPROVE ($49 \pm 5\%$) (Mean ± 2 SD) (Klouda et al., 2005).

2.4 Sample preparation for quantification of WSOC

The WSOC fraction was quantified according to equation (1) after extracting a 1.5 cm^2 filter-punch with Milli-Q water (100 ml).

$$\text{WSOC} = \text{OC} - \text{WINSOC} \quad (1)$$

The equipment used for extraction of WSOC is illustrated in Fig. 1. The extraction is based on gravity feed of water through the exposed filter-punch. To prevent loss of carbonaceous material that is not water-soluble and to keep the EC uniformly distributed, a supporting filter punch (1.5 cm^2) is placed on top of the exposed punch during extraction. After extraction, and prior to analysis, the filter punches are left to dry in a desiccator. The reproducibility of the WSOC measurements, including aliquot preparation, extraction procedure and instrument variability, was found to be satisfac-

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tory (RSD<8%). The reproducibility was calculated based on analyses of three aliquots from each of two different filter samples collected at the curbside site.

2.5 HPLC/HRMS-TOF analysis of monosaccharide anhydrides

Analysis of levoglucosan, mannosan and galactosan in PM₁₀ aerosol filter samples was performed using High Performance Liquid Chromatography (HPLC) in combination with HRMS-TOF (High Resolution Mass Spectrometry Time-of-Flight) and according to the method described by Dye and Yttri (2005). The method holds the combined merits of short preparation, high sensitivity and complete separation of levoglucosan from its isomeric compounds, as evaluated by Schkolnik and Rudich (2006). The methodology has successfully been applied on filter samples containing levoglucosan concentrations ranging from background (Stohl et al., 2007) to urban levels (Yttri et al., 2005).

2.6 Ion chromatography analysis of Potassium

Prior to ion chromatography analysis, the Zefluor Teflon filters were soaked in Milli-Q water (10 ml) and subjected to ultrasonic agitation (30 min). The extracts were analyzed on a Dionex120DX ion chromatograph, using a Dionex cation exchange CS12A column (4 mm×200 mm), and a conductivity detector. The sample was eluted using sulphuric acid at a flowrate of 1 ml min⁻¹.

3 Results and discussion

3.1 Estimating the positive sampling artefact of OC

Table 2 shows the OC_B/OC_{FF} ratios for the various sites. OC_B is gaseous OC present on the backup filter and OC_{FF} is the sum of gaseous and particulate OC on the front filter. This ratio provides an estimate of the magnitude of the positive sampling artefact

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of OC when using tandem filter sampling. When subtracting OC_B from OC_{FF} positive-artefact-corrected particulate organic carbon (OC_p) is obtained.

The positive artefact of OC ranged from 12–37% for PM_{10} , whereas the corresponding range for $PM_{2.5}$ was 27–44%. This shows that OC_p could be severely overestimated if the positive artefact is not accounted for. Performing parallel sampling of $PM_{2.5}$ at the urban background site, the QBT approach was found to provide a consistently higher estimate (44%) of the positive artefact of OC compared to the QBQ approach (19%). This finding is in accordance with what has been reported by others (McDow and Huntzicker, 1990; Turpin et al., 1994; Subramanian et al., 2004).

To align the positive artefact of OC in PM_{10} at the urban background site (using the QBQ-approach) with the other sites, which employed the QBT approach, we used the OC_B concentration of the $PM_{2.5}$ sampler operated in parallel and at the same filter face velocity as the PM_{10} sampler (Table 2). This exercise raised the estimate of the positive artefact of OC from 12% to 29%.

Although the QBT approach provides a higher estimate of the positive artefact than the QBQ approach, any general recommendation concerning which sampling train is the better one should be considered with caution, due to various atmospheric and meteorological conditions encountered at different sampling sites. Further, Subramanian et al. (2004) reported an overcorrection of 16–20% for the QBT approach compared to that of denuder-based sampling. It should be noted that the dual filter approach provides a conservative estimate of OC_p , as the negative artefact (evaporation of semi-volatile OC from the front filter) is not accounted for.

3.2 Ambient concentrations of EC and OC_p

Concentrations of elemental carbon (EC), positive-artefact-corrected particulate organic carbon (OC_p), organic carbon on backup filters (OC_B), particulate total carbon (TC_p), and EC/ TC_p ratios for PM_{10} and $PM_{2.5}$ are presented in Table 3. Notably, the mean concentrations of OC_p reported for PM_{10} ($6.1 \mu\text{g m}^{-3}$) and for $PM_{2.5}$ ($4.8 \mu\text{g m}^{-3}$) at the

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suburban site in winter, were equal to, or even higher, than the concentrations reported for the curbside site and the urban background site. The mean concentration of EC in PM_{10} were up to one order of magnitude higher at the curbside site ($5.7 \mu\text{g m}^{-3}$) and at the urban background site ($4.6 \mu\text{g m}^{-3}$) compared to the suburban site ($0.4 \mu\text{g m}^{-3}$ in summer and $1.7 \mu\text{g m}^{-3}$ in winter), likely due to the impact of vehicular traffic. The higher concentrations of EC at the curbside site and the urban background site are also reflected by high EC/TC_p ratios, e.g. a mean EC/TC_p ratio of 66% ($PM_{2.5}$) was observed at the curbside site, possibly indicating a high influence of vehicles running on diesel. A pronounced seasonal variation was observed for EC and OC_p at the suburban site, both for PM_{10} and $PM_{2.5}$. In winter, the mean concentration of OC_p was 2.5–3 times higher compared to summer, whereas the corresponding range for EC was 4–6. Concurrent measurements of levoglucosan (See Table 3) suggest that this seasonal variation is attributed to residential wood burning. The influence of residential wood burning is discussed in detail in Sect. 3.3. OC_p and EC were mainly associated with fine aerosols (Table 4), which are to be expected, as emissions from combustion of fossil fuel and biomass tend to be the dominant sources of carbonaceous aerosols in urban influenced areas. The association with fine aerosols was not as pronounced for OC_p as for EC. While EC stems from combustion solely, primary biological aerosol particles (PBAP), which predominantly reside in the coarse fraction of PM_{10} , could make a contribution to OC_p . Indeed, sugars and sugar-alcohols, which are tracers of PBAPs, were reported for all sites examined in the present study by Yttri et al. (2007b). Condensation of semi volatile organic vapours might also contribute to OC_p in the coarse mode. The highest daily concentrations of EC and OC_p in PM_{10} were observed at the urban background site for a period of three consecutive days (10–13 November 2001) (Fig. 2), where EC ranged from $11\text{--}14 \mu\text{g m}^{-3}$ and OC_p between $16\text{--}21 \mu\text{g m}^{-3}$. Surprisingly, the majority (68%) of EC and OC_p were associated with the coarse fraction during these days. We suggest that this could be attributed to agglomeration of fine carbonaceous matter and coarse mineral dust on the ground, which then is resuspended. In winter, resuspension of road dust, generated from the extensive use

of studded tires, is frequently seen in Oslo during favorable meteorological conditions, giving rise to concentrations of $PM_{10-2.5}$, which could exceed $100 \mu\text{g m}^{-3}$ on a 24 h basis (Hagen and Haugsbakk, 2000; Haugsbakk, 1994). It is also likely that the bitumen content of the paved road dust contribute mainly to OC in $PM_{10-2.5}$.

The wide range of analytical methods used to quantify EC and OC in different studies generate data that are hard to compare. The only study that to our knowledge has reported an extended time series of EC and OC concentrations for an urban area in Scandinavia using thermal optical analysis is that of Viidanoja et al. (2002), which reported monthly mean concentrations of OC_p and EC for PM_{10} and $PM_{2.5}$ for a traffic-influenced urban site in Helsinki (Finland).

The mean OC_p concentrations reported for the curbside and the urban background sites in the present study are within ($PM_{2.5}$) and in the upper (PM_{10}) range of the monthly means reported for Helsinki. For EC, the mean concentrations observed at the curbside and the urban background sites are higher than the maximum monthly mean reported for Helsinki, typically by a factor of 2–3. The aerosol content of TC is not subject to uncertainties related to the split between EC and OC. Hence, this more robust parameter can be used to confirm some of the findings deduced from the EC and OC data. Here we find that the mean concentrations of TC_p at the curbside and the urban background site are 1.2–1.5 times higher than the highest monthly mean reported for Helsinki.

3.3 Impact of OC from wood burning

3.3.1 Levels of levoglucosan

Ambient concentrations of the monosaccharide anhydrides levoglucosan, mannosan and galactosan in PM_{10} are reported in Table 3. Levoglucosan was the most abundant isomer accounting for 72–78% of the sum of the three isomers. The mean concentration of levoglucosan ranged from 20 ng m^{-3} at the suburban site during summer to 407 ng m^{-3} in winter. Intermediate levels were observed for the curbside site

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(45 ng m⁻³) and the urban background site (166 ng m⁻³) where sampling was performed in early fall and late fall, respectively. The mean concentration of levoglucosan observed in winter at the suburban site was more than 20 times higher compared to summer. Such pronounced seasonal variations have previously been reported by Zdráhal et al. (2001) for an urban background site in Ghent (Belgium), and by Puxbaum et al. (2007) for a semi urban site in Aveiro (Portugal) and the rural site K-Puzta in Hungary. The mean relative contribution of levoglucosan C to OC_p ranged from 0.3±0.6% at the suburban site in summer to 3.2±0.9% in winter (Table 5). This range corresponds to that reported by Puxbaum et al. (2007) for six sites of various site categories in Central and Southern Europe.

3.3.2 Estimates of OC from wood burning

A wide range of levoglucosan-to-OC emission ratios has been reported for various types of softwood and hardwood (Fine et al., 2001, 2002a, b, 2004; Mazzolini et al., 2007; Simoneit et al., 1999), typically for US tree types, using US appliances. For example, Fine et al. (2002b) found a levoglucosan-to-OC emission ratio of 14±2% (PM_{2.5}) for ten regions covering the entire US, when normalizing with respect to local tree types and appliances used. Emission ratios for European tree types are scarce; Puxbaum et al. (2007) reported a levoglucosan-to-OC-ratio (PM₁₀) of 14%, based on test burns of beech (7.9%) and spruce (20.6%) indigenous to Austria, Hedberg et al. (2006) found a levoglucosan to PM₁₀ ratio of 0.3% to 22% for wood and pellet boilers in Sweden, and Schmidl et al. (2008) found ratios between 8% and 30% (PM₁₀) for Austrian wood stoves. Studies with single appliances show a large variability, partly caused by different combustion conditions (e.g. Schmidl et al., 2008), different technologies or type of wood used. Studies using ambient data provide a good method of averaging for the various complications, but results will also vary from region to region. For example, Sandradewi et al. (2008) found levoglucosan/OC ratios ranging from 5–20%, and showed that this ratio varied for different parts of Switzerland, likely due to different

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usage of hard and soft woods. Hedberg et al. (2006) found a levoglucosan to PM_{wood} ratio of 18% on average for a town in Northern Sweden, which would suggest a very high Levoglucosan/OC ratio (over 40%) (Simpson et al., 2007). However, the source-reconciliation used by Hedberg et al. (2006) had a missing mass of $2.8 \mu g m^{-3}$, almost comparable with the identified PM_{wood} of $3.01 \mu g m^{-3}$, so the real ratio may be closer to 20%.

Here we argue that wintertime samples from the suburban site Elverum serve as very good indicator of emissions from wood-burning relevant to conditions in this part of Norway, as observed in the ambient atmosphere. The wintertime measurements taken at this site spanned a wide range of temperatures (daily means from $+1$ to $-19^{\circ}C$, daily minimum from -2 to $-26^{\circ}C$), with colder temperatures leading to markedly higher levels of TC_p (Fig. 3) and other pollutants. By itself, the change in concentrations level says little about any change in emissions, since meteorology also plays a strong role, but an examination of the ratios between pollutants contains valuable information. Over such a temperature range the emissions from residential wood-burning are expected to change drastically as households increasingly supplement any other forms of heating. Increased wood-burning leads to higher levoglucosan and Potassium levels as well as increased EC, OC_p and TC_p , as the temperature decreases. On the other hand, emissions from road-vehicles or other sources such as from cooking are not expected to be as strongly affected by temperature.

Figure 4 shows the variation of levoglucosan, $WSOC_p$, Potassium, EC, and OC_p as a fraction of TC_p as a function of the daily minimum temperature, T_{min} , for both PM_{10} and $PM_{2.5}$. Results for daily mean temperature, not shown, are similar. For both size fractions, the relative share of the various components remains remarkably constant at all temperatures. The slopes of the regression lines are close to zero and only significant ($p < 0.05$) for the case of Potassium against T_{min} for $PM_{2.5}$. Even here the slope is very small (0.00033). The almost constant ratios among pollutants found in these samples strongly implies that dispersion rather than changing sources of emissions is responsible for the increase of TC_p with lowering temperature.

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The correlation between OC_p and TC_p is remarkably good for these data-sets for both size fractions (Table 5). The slope is 0.824 for PM_{10} and 0.74 for $PM_{2.5}$, implying OC_p/EC ratios of 4.26 for PM_{10} and 2.8 for $PM_{2.5}$. These OC_p/EC ratios are much higher than those found at the curbside site, where OC_p/EC was found to be 0.78 for PM_{10} and 0.54 for $PM_{2.5}$. As it seems likely that the curbside site would give if anything an overestimate of the OC_p/EC ratio from traffic emissions, the amount of OC_p we can attribute to traffic at Elverum is small even if all EC was assumed to be from traffic.

Figure 5 and Table 5 show a rather good correlation between levoglucosan and TC_p for both size fractions. The intercept of the least-square fit is slightly negative on the TC_p axis, implying that the changes in levoglucosan alone are more than sufficient to explain the variations in TC_p . The very good relationship for the PM_{10} fraction is somewhat surprising, since Yttri et al. (2005) have shown previously that about 95% of levoglucosan is found in the $PM_{2.5}$ fraction. These results would suggest that some of the factors mentioned in Sect. 3.1 (agglomeration of fine OC followed by resuspension, or condensation of OC to the coarse mode) could play a role in associating coarse OC with wood-burning.

In conclusion, the high OC_p/EC ratio, the lack of variation of OC_p/TC_p or levoglucosan (or K^+)/ TC_p with decreasing temperature, and the very good linear correlation between levoglucosan (or K^+) and TC_p all suggest that residential wood-burning emissions are by far the major source of OC_p and TC_p at the Elverum site for both $PM_{2.5}$ and PM_{10} . These results are fully consistent with emission inventory estimates for Elverum, which states that over 99% of the PM_{10} in this town arises from wood combustion (Trond Sandmo, Statistics Norway pers. comm.). These findings suggests that these Elverum data may be used to derive ratios of levoglucosan to TC_p , OC_p and EC, which are likely to be representative of wintertime emissions in southern Norway, at least for areas where similar wood types are burnt. As shown in Figure 5a, a simple relation that $TC_p = 19.63 \cdot \text{levoglucosan}$ fits the PM_{10} data very well. A rounded value $TC_p/\text{levoglucosan} = 20$ may be used as our best-estimate of the $TC_p/\text{levoglucosan}$ ratio for Elverum wood burning emissions. We have also estimated some reasonable uncer-

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tainty limits for these ratios, which for PM_{10} range between 14 and 23, as indicated in Fig. 5a. For $PM_{2.5}$ the equivalent best estimate ratio (rounded) is 15, with a likely range of between 10 and 18 (Fig. 5b). The ratio OC_p/TC_p was found to be rather constant, with mean values of 0.775 for the PM_{10} samples, and 0.723 for the $PM_{2.5}$ samples. Thus, we derive OC_p /levoglucosan ratios of 15 (range 11–18) for PM_{10} and 11 (range 7.8–14) for $PM_{2.5}$. Alternatively, levoglucosan accounts for 6.7% (range 5.6–9.1%) of OC_p in PM_{10} and 8.9% (range 7.1–12%) of OC_p in $PM_{2.5}$. As an additional complication, Dye and Yttri (2005) found that approximately 25% more levoglucosan was extracted from Teflon filters than quartz, based upon concurrent sampling at Elverum in wintertime. This would suggest that the “true” emission ratios, of OC_p /levoglucosan, might be lower than those found here by around 25%, and that the choice of factor used depends on the type of filter available. However, this is not a problem with the current study as both emission-ratios and source allocation are done consistently.

Table 6 summarizes the contribution of OC_{wood} to OC_p levels for the PM_{10} fraction at other sites, assuming a levoglucosan-to- OC_p emission ratio of 6.7% (range 5.6–9.1%) as determined above. These estimates suggest that wood-burning is contributing significantly to OC_p also within the city.

At the kerbside site we find that OC_{wood} accounted for 11–18% of OC_p in PM_{10} , whereas the corresponding range for the urban background site was 22–36%. The least influence of residential wood burning emissions was experienced at the suburban site during summertime sampling, i.e. OC_{wood} accounted for only 8–13% of OC_p . This low level could partly be explained by the fact that levoglucosan was present in only 6 of 22 samples. For those days when levoglucosan was present in the aerosol filter samples, the relative contribution of OC_{wood} to OC_p was substantially higher; i.e. 29–48%. Caution should be used however when extrapolating levoglucosan-to- OC_p ratios from wintertime conditions to summer, as the latter will likely include many diverse sources, such as open agricultural burning and forest fires. Still, this finding shows that the contribution of OC_{wood} to OC_p in urban areas could be considerable even in summer. The relative contribution of OC_{wood} to OC_p was estimated to be between 73%

and 100% at the suburban site during winter time sampling. Although the estimated range is rather wide, our conclusion that residential wood burning is the major source of organic carbon at this site is solid.

3.3.3 Ratios of K^+ -, EC^- -, OC_p^- -, and $WSOC_p^-$ -to-levoglucosan

5 Ratios of K^+ -to-levoglucosan, EC^- -to-levoglucosan, OC_p^- -to-levoglucosan, and $WSOC_p^-$ -to-levoglucosan was calculated for samples collected at the suburban site in winter because of its pronounced residential wood burning profile for comparison with corresponding ratios reported in the literature.

10 The K^+ /levoglucosan ratio (0.33 ± 0.10) is in the upper range of that reported by Fine et al. (2001, 2002a, b, 2004), Schauer et al. (2001) and Schmidl (2008) for test burns of various tree types, ranging from 0.02–0.9, but equal to ambient observations reported for Christchurch (0.3) and Auckland (0.3) (New Zealand), which both are cities considerably influenced by residential wood burning, as reported by Wang et al. (2006). A K^+ /levoglucosan ratio of 0.3 was also reported during wild fire events in the Amazonas (Graham et al., 2002).

15 The levoglucosan content of $WSOC$ is rarely reported. Here we find that levoglucosan accounted for $9.7 \pm 3.6\%$ of $WSOC_p^-$, which is higher than that reported for Christchurch (5.7%) and Auckland (5.2%), but lower than observed during wild fire events in the Amazonas (14.2%) (Graham et al., 2002). No $WSOC$ -to-levoglucosan ratio has been reported for test burns of wood that we are aware of.

20 Data on EC^- /levoglucosan ratios are scarce, but could be calculated from the test burn studies of Fine et al. (2001, 2002a, b, 2004), Schauer et al. (2001) and Schmidl et al. (2008) and from ambient samples collected during forest fire events (Graham et al., 2002). Compared to the abovementioned studies, the EC^- /levoglucosan ratio of 4.3 \pm 1.1 reported here is in the upper range.

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3.4 Ambient concentrations of WSOC_p

The mean concentration of WSOC_p in PM₁₀ ranged from 1.2 μg m⁻³ at the curbside site to 4.8 μg m⁻³ at the suburban site in winter (Table 3). The relative contribution of WSOC_p to OC_p was particularly high at the suburban site, accounting for 79% in winter and 67% in summer. This finding contradicts the general assumption that WSOC is higher in summer compared to winter due to secondary organic aerosol (SOA) formation in summer (Castro et al., 1999). During winter the high WSOC_p to OC_p ratio was consistent with wood burning being the major source, emitting a large number of partly and completely water-soluble compounds, which are lignin, cellulose, and hemi-cellulose breakdown products (Nolte et al., 2001; Schauer et al., 2001; Fine et al., 2002b; Simoneit, 2002). In summer, the low concentration of levoglucosan (20 ng m⁻³) excluded residential wood burning as a major contributor of WSOC_p. Concurrent measurements of selected sugars and sugar-alcohols, which are completely soluble in water, verified concentrations 5 times higher than that of levoglucosan (Yttri et al., 2007b), demonstrating that primary biological aerosol particles (PBAP) are important contributors of water-soluble organic compounds in summer. Other possible sources of WSOC could be SOA and long-range transported compounds from distant polluted regions (e.g. Simpson et al., 2007), however, these sources cannot be quantified with the available data.

The relative contribution of WSOC_p to OC_p at the curbside site (28%) and the urban background site (39%) was substantially lower than at the suburban site. Possible explanations could be low levels of polar oxygenated compounds in vehicular exhaust and the short distance from this source to the sampling sites, which leaves little time for oxidation of aerosol constituents and precursor compounds. Still, diacids present in petrol and diesel motor exhaust (Kawamura and Kaplan, 1987), and diacid formation following photochemical oxidation of anthropogenic gaseous precursors such as benzene and toluene, (Kawamura and Ikushima, 1993) are suggested to be important contributors to the WSOC fraction in the urban atmosphere. On the other hand, the

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WSOC_p to OC_p ratios for the curbside and urban background sites (Table 3) are substantially higher than those reported for other traffic influenced sites by Ruellan and Cachier (2001) (12.5%) and Mader et al. (2004) (14–26%). This difference could partly be attributed to the influence of residential wood burning, as confirmed by the samples content of levoglucosan (Table 3), and to the presence of sugars and sugars-alcohols associated with PBAP (Yttri et al., 2007b). Different extraction procedures and different analytical instrumentation could also add to the observed difference.

The concentration of WSOC_p in PM_{2.5} was only quantified in samples from the urban background site (1.2 μg m⁻³). Based on days with concurrent measurements of WSOC_p in PM₁₀ and PM_{2.5}, 55% of WSOC_p was associated with the coarse fraction of PM₁₀ (PM_{10-2.5}). Indeed, the concentration of seven sugars and sugar-alcohols, which typically are associated with PM_{10-2.5}, was higher than that of the monosaccharide anhydrides at the suburban site, suggesting that PBAP may well be an important source of WSOC even in winter (Yttri et al., 2007b).

For PM_{2.5}, the relative contribution of WSOC_p to OC_p was 32% (Table 3), which is somewhat less than observed for PM₁₀ (Table 3).

3.5 Contribution of carbonaceous matter to the ambient concentration of PM₁₀

Conversion of OC to OM (Organic matter) is a critical factor of uncertainty in mass closure calculations. A conversion factor of 1.4 has commonly been applied both in urban and rural areas (Gray et al., 1986; Malm et al., 2004; Putaud et al., 2004), but Turpin and Lim (2001) concluded that 1.6±0.2 was a better estimate for urban areas when reviewing conversion factors reported in the literature. Furthermore, 1.9–2.3 was suggested for aged aerosols, and 2.2–2.6 for aerosols originating from biomass burning (Turpin and Lim, 2001). Thus, using one factor for all sites is likely to introduce uncertainties, as the source contribution varies from site to site. In the present study we attempt to reduce some of this uncertainty by using conversion factors for WSOC and WINSOC. As shown in Table 3, the relative contribution of WSOC_p and WINSOC_p to OC_p vary considerably between the sites investigated. WSOC contains molecules

that have a high molecular weight pr. carbon weigh (1.5–3.8), while the corresponding range for the water-insoluble fraction is much lower (1.1–2.1) (Turpin and Lim, 2001). Still, experimentally derived conversion factors for WSOC and WINSOC for selected sites are scarce and the lack of such could still bias the results.

5 An experimental derived factor of 2.0 (Kiss et al., 2002) was used to convert WSOC to WSOM (Water-soluble organic material), except from samples collected at the suburban site in winter where a factor of 2.3 was used. The presence of highly oxygenated organic compounds, such as monosaccharide anhydrides, sugars and sugar-alcohols, which have theoretical conversion factors from 2.3–2.5, have been demonstrated at all
10 sites investigated in the present study (Table 3, Yttri et al., 2007b), somewhat justifying the conversion factor used for WSOC. The higher conversion factor (Turpin et al., 2000; Turpin and Lim, 2001; Kiss et al., 2002) used at the suburban site in winter could be justified by the impact of residential wood burning, as substantiated by the very high levels of levoglucosan. For WINSOC a conversion factor of 1.3 was applied, whereas
15 a conversion factor of 1.1 was used to convert EC to EM (Elemental matter) (Kiss et al., 2002).

The relative contribution of particulate carbonaceous matter [$PCM = (WSOC \times 2.0 - 2.3) + (WINSOC \times 1.3) + (EC \times 1.1)$] to PM_{10} ranged from 46–83%, thus being the major fraction at the sites investigated. The very high PCM reported for the suburban site in
20 winter (83%) is consistent with emissions from residential wood burning, which have a high carbon content.

Disentangling PCM into particulate organic matter (OM_p) and EM showed that OM_p accounted for 27–72% of PM_{10} , whereas EM accounted for 5–25% of PM_{10} . The relative contribution of OM_p to PM_{10} at the curbside (27%) and the urban background
25 (38%) sites correspond with the monthly mean based range (29–38%) reported by Viidanoja et al., (2002) for an urban traffic-influenced site in Helsinki, using a factor of 1.6 to convert OC into OM. For EM, the relative contribution to PM_{10} is substantially higher at the curbside (25%) and the urban background site (19%) than for the Helsinki site (4–12%).

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It is shown that particulate carbonaceous material (PCM) is the major (46–83% of PM_{10}) contributor to PM_{10} in the urban environment regardless of site classification and despite that the calculations are based on conservative estimates of particulate organic carbon (OC_p). Levels of EC varied by more than one order of magnitude between sites, likely due to the higher impact of vehicular traffic at the curbside and the urban background sites. In winter, the level of OC_p at the suburban site was equal to (for PM_{10}) or even higher (for $PM_{2.5}$) than the levels observed at the curbside and the urban background sites. This finding was attributed to the impact of residential wood burning at the suburban site in winter, which was confirmed by a high mean concentration of levoglucosan (407 ng m^{-3}). This finding shows that exposure to primary combustion derived OC_p could be equally high in residential areas as in a city center. It was estimated that OC from wood burning (OC_{wood}) constituted 73–100% to OC_p at the suburban site in winter, whereas the corresponding percentage for summer was 8–11%. Thus, seasonal variations in emissions from residential wood burning, i.e. concentrations of levoglucosan being >20 times higher in winter compared to summer, could explain the great seasonal variation seen for the concentration of the carbonaceous aerosol at this site. It is demonstrated that OC from wood burning (OC_{wood}) accounted for almost all OC_p at the suburban site in winter, allowing a new estimate of the ratio $OC_p/\text{levoglucosan}$ for both PM_{10} and $PM_{2.5}$. These ratios were found to be 15 (11–18) for PM_{10} and 11 (7.8–14) for $PM_{2.5}$. This corresponds to 6.7% for PM_{10} and 8.9% for $PM_{2.5}$.

Acknowledgements. This work was supported by VISTA (The Norwegian Academy of Science and Letters and Statoil) project number 6143, the Norwegian Research Council project number 171691/S30 (SORGA), the EU project number 036833-2 (EUCAARI) within the 6th Framework Programme, and UNECE (the EMEP project).

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Table 1. Volumetric flow rate (l min^{-1}) and filter face velocity (cm s^{-1}) for the aerosol samplers operating according to tandem filter set ups.

Size fraction	Oslo Urb. backgr. (Nov–Dec, 2001)	Oslo Curbside (Sept–Oct, 2002)	Elverum Suburban (Jan–Mar, 2002)	Elverum Suburban (May–Jun, 2002)
PM ₁₀	NILU Filter Unit/ NILU (10 l min ⁻¹) (13 cm s ⁻¹) PM ₁₀ inlet	NILU Filter Unit/ NILU (10 l min ⁻¹) (13 cm s ⁻¹) PM ₁₀ inlet	NILU Filter Unit/ NILU (10 l min ⁻¹) (13 cm s ⁻¹) PM ₁₀ inlet	NILU Filter Unit/ Gent (16.7 l min ⁻¹) (22 cm s ⁻¹) PM ₁₀ inlet
PM _{2.5}	NILU-SFU PM _{2.5} (10 l min ⁻¹) (13 cm s ⁻¹)	Derenda PM _{2.5} (38 l min ⁻¹) (53 cm s ⁻¹)	NILU-SFU PM _{2.5} (10 l min ⁻¹) (13 cm s ⁻¹)	Derenda PM _{2.5} (38 l min ⁻¹) (53 cm s ⁻¹)

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Table 2. Mean (min and max) OC_B/OC_{FF} ratios for PM_{10} and $PM_{2.5}$ using QBT tandem filter set ups (%).

Site	$OC_B/OC_{FF}(PM_{10})$	$OC_B/OC_{FF}(PM_{2.5})$
Urban background (Nov–Dec) ²⁾	12 (5–25) ¹⁾ 29 (18–52) ³⁾	19 (9–35) ¹⁾ 44 (23–60) ²⁾
Curbside (Sep–Oct)	26 (15–38)	27 (19–42)
Suburban (Jan–Mar)	31 (14–59)	36 (24–53)
Suburban (May–Jun)	37 (23–44)	38 (20–73)

1) QBQ is used to correct for the positive artifact of OC

2) Both QBQ and QBT were used to estimate the positive artifact of OC in $PM_{2.5}$ at the urban background site.

3) A QBT-based estimate of the positive artifact was based on OC_B from the $PM_{2.5}$ sampler operated in parallel and at the same filter phase velocity.

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Table 3. Mean (min and max) weekday concentrations of OC_P, OC_B, EC, TC_P, WSOC_P, WINSOC_P and monosaccharide anhydrides in PM₁₀ and PM_{2.5} and relative contributions of EC-to-TC_P, WSOC_P-to-OC_P and WINSOC_P-to-OC_P in PM₁₀ and PM_{2.5}.

Site	Size	OC _P μg m ⁻³	OC _B μg m ⁻³	EC μg m ⁻³	TC _P μg m ⁻³	EC/TC _P %	WINSOC _P μg m ⁻³	WSOC _P μg m ⁻³	(WINSOC/OC) _P %	(WSOC/OC) _P %	Levoglucosan ng m ⁻³	Mannosan ng m ⁻³	Galactosan ng m ⁻³	SUM MA ng m ⁻³
Urban background (Nov–Dec)	PM ₁₀ ⁽¹⁾	7.6 (1.6–21)	0.8 (0.2–1.3)	4.6 (0.9–14)	12 (3.1–35)	35 (20–46)	4.8 (1.6–12)	3.7 (0.4–8.9)	61 (42–88)	39 (12–58)	166 (nd–475)	41 (nd–155)	3 (nd–17)	211 (nd–616)
	PM _{2.5} ^(2,3,4)	6.4 (1.0–18)	2.0 (0.9–4.3)		11 (2.4–32)	41 (23–59)								
	PM ₁₀ ⁽¹⁾	3.9 (1.1–9.3)	0.9 (0.4–2.1)	2.2 (0.7–5.1)	6.1 (2.0–14)	36 (22–46)	1.9 (0.9–3.4)	1.2 (nd–4.5)	68 (43–83)	32 (17–57)				
	PM _{2.5} ^(2,3)	2.8 (0.8–7.9)	2.0 (0.9–4.3)		5.0 (1.7–13)	45 (28–60)								
Curbside (Sep–Oct)	PM ₁₀ ⁽¹⁾	4.5 (2.6–6.5)	1.6 (1.1–2.6)	5.7 (3.3–8.2)	10 (6.0–14)	49 (45–57)	3.2 (2.0–4.2)	1.3 (0.1–2.9)	72 (55–95)	28 (5–45)	45 (9.4–109)	8.5 (nd–21)	3.5 (nd–7.3)	57 (11–134)
	PM _{2.5} ^(2,3)	2.9 (1.2–5.0)	1.0 (0.7–1.2)	5.4 (3.1–7.1)	8.3 (4.3–12)	66 (58–73)								
Suburban (Jan–Mar)	PM ₁₀ ⁽¹⁾	6.1 (1.2–17)	2.3 (1.1–3.9)	1.7 (0.5–3.5)	7.8 (1.7–21)	23 (16–32)	1.3 (0.1–4.6)	4.8 (1.1–13)	13 (8–45)	79 (55–92)	407 (134–971)	116 (34–286)	2 (1–7)	526 (187–1240)
	PM _{2.5} ^(2,3)	4.8 (1.2–11)	2.3 (1.3–3.6)	1.7 (0.5–4.1)	6.5 (1.6–16)	28 (21–31)								
Suburban (May–Jun)	PM ₁₀ ⁽¹⁾	2.1 (0.8–4.3)	1.1 (0.2–2.2)	0.4 (0.1–1.0)	2.5 (0.9–5.0)	17 (11–26)	0.6 (0.1–1.6)	1.4 (0.3–2.7)	33 (5–75)	67 (25–95)	20 (nd–151)	6 (nd–42.3)	1 (nd–7)	25 (nd–201)
	PM _{2.5} ^(2,3)	1.9 (0.2–3.7)	0.9 (0.6–2.0)	0.3 (0.1–0.7)	2.2 (0.3–4.3)	18 (9–39)								

- (1) OC_P and OC_B is based on QBQ.
- (2) OC_P and OC_B is based on QBT.
- (3) OC_P and TC_P are calculated from OC_B (PM_{2.5}) obtained from the PM_{2.5} sampler operated at the same site according to the QBT approach, and at an identical filter face velocity.
- (4) The figures for levoglucosan, mannosan, galactosan and SUM MA are previously presented in Yttri et al. (2005), 55, 1169–1177.
- (5) Monosaccharide anhydrides were detected in 6 of 22 samples.

Footnote: Explanation of abbreviations: OC_P=particulate organic carbon, OC_B=organic carbon on backup filter, EC=elemental carbon, and TC_P=particulate total carbon. WINSOC_P=water-insoluble particulate organic carbon, WSOC_P=water-soluble particulate organic carbon.

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Table 4. Mean weekday $PM_{2.5}/PM_{10}$ ratios for OC_p , EC and TC_p (%).

Site	$(PM_{2.5}/PM_{10})_{OCp}$	$(PM_{2.5}/PM_{10})_{EC}$	$(PM_{2.5}/PM_{10})_{TCP}$
Urban background (Nov–Dec)	64	65	64
Curbside (Sept–Oct)	61	94	79
Suburban (Jan–Mar)	73	95	85
Suburban (May–Jun)	77	80	77

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Table 5. Results of regression analysis from the suburban site (Elverum), wintertime data. For each pair of pollutants (x,y), we give the mean value, slope, intercept and R^2 of regression $y=mx+c$. LG denotes levoglucosan (always from PM₁₀ samples), other data as in text. All data from QBT samples only.

Size	y	x	Mean y/x	Slope	Intercept	R^2
PM ₁₀	OC _p	TC _p	0.79	0.824	-0.305	1.00
	PM ₁₀	LG	43.0	37.178	+2.35	0.81
	TC _p	LG	19.6	20.486	-0.35	0.87
	OC _p	LG	15.4	16.889	-0.598	0.87
	EC	LG	4.2	3.597	+0.248	0.80
	WSOC _p	LG	11.8	12.814	-0.401	0.84
	K ⁺	LG	0.32	0.260	-0.023	0.72
	TC _p	K ⁺	62.3	65.293	-0.391	0.82
	OC _p	K ⁺	49.0	53.538	-0.599	0.81
PM _{2.5}	OC _p	TC _p	0.72	0.74	-0.099	1.00
	TC _p	LG	15.5	16.690	-0.509	0.88
	OC _p	LG	11.2	12.414	-0.505	0.89
	EC	LG	4.3	4.275	-0.004	0.84

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Table 6. Estimated contributions of levoglucosan C and OC_{wood} to OC_p at different sites. Only data from PM_{10} (QBT) sampling are presented.

Site	Levoglucosan C/ OC_p (%) ¹	OC_{wood} ($\mu\text{g m}^{-3}$) ²	OC_{wood}/OC_p (%) ²
Urban background (Nov–Dec)	0.9±0.5	2.3 (1.7–2.8)	30 (22–36)
Curbside (Sep–Oct)	0.5±0.4	0.6 (0.5–0.8)	15 (11–18)
Suburban (Jan–Mar)	3.2±0.9	6.1 (4.5–6.1) ³	100 (73–100) ³
Suburban (May–Jun)	0.3±0.6	0.3 (0.2–0.3)	11 (8–13)

(1) Uncertainty range represents variability (std. dev.) in measured ratios;

(2) Here we give the best estimate contribution derived from levoglucosan, with the min and max estimate in parenthesis, see text;

(3) Max. contribution limited to 100% of OC_p .

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Table 7. Mean concentration of PM₁₀ and PCM, and the relative contribution of PCM-to-PM₁₀.

	PM ₁₀ (μg m ⁻³)	PCM (μg m ⁻³)	PCM/PM ₁₀ (%)	OM _p /PM ₁₀ (%)	EM/PM ₁₀ (%)
Urban background (Nov–Dec)	26.4	17.3 ¹ /15.2 ²	66 ¹ /58 ²	46 ¹ /38 ²	19
Curbside (Sep–Oct)	24.8	13.0	53	27	25
Suburban (Jan–Mar)	17.5	14.5	83	72	11
Suburban (May–Jun)	8.8	4.0	46	41	5

(1) Correction of the positive artifact of organic carbon is performed by the QBQ-approach, while QBT is used for the others. (2) A recalculation has been performed using the OC concentration from the backup filter of the PM_{2.5} sampler operated at the same site according to the QBT approach, and at an identical filter face velocity.

Footnote: Explanation of abbreviations (PCM=Particulate carbonaceous matter corrected for the positive artifact of OM (OM=OC×conversion factor), OM_p=organic material corrected for the artifact of OM, EM=elemental matter (EC×conversion factor)).

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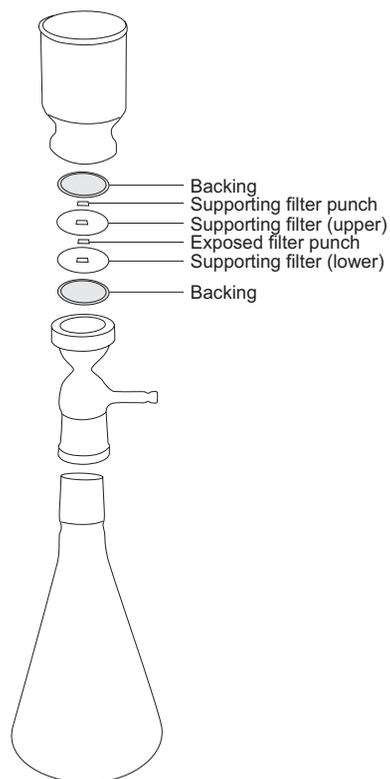


Fig. 1. Set up for extraction of the water-soluble organic carbon in ambient aerosols.

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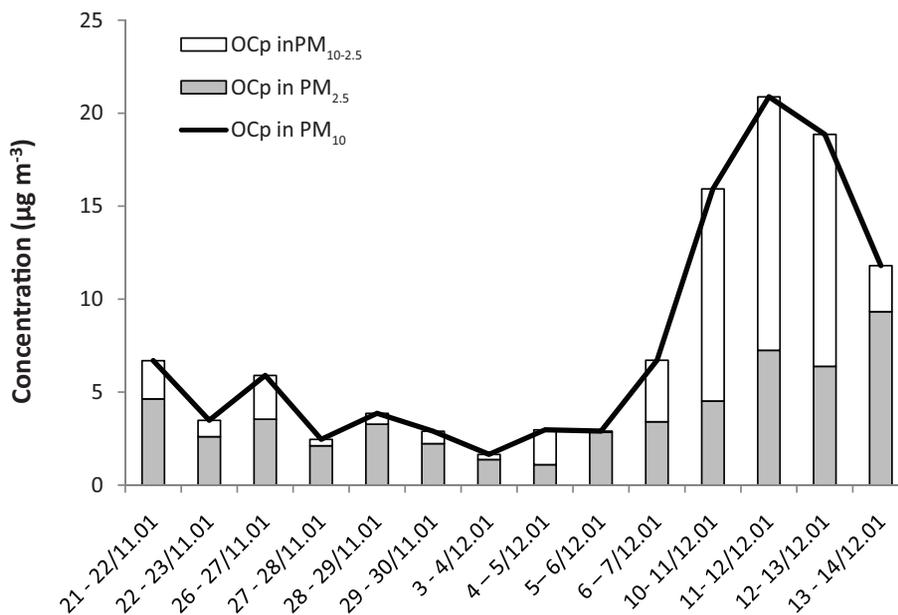


Fig. 2. Time series of OC_p in PM₁₀, PM_{10-2.5} and PM_{2.5}. Urban background site Nov–Dec, 2001.

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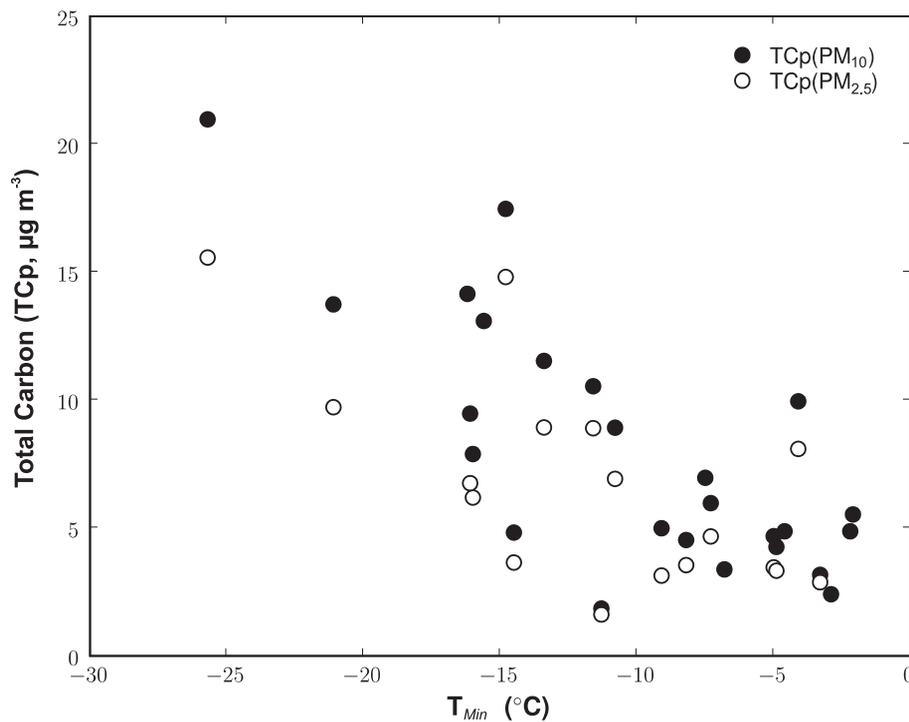


Fig. 3. Total carbon (TC_p) versus daily minimum temperature. Suburban site, Jan–Mar, 2002.

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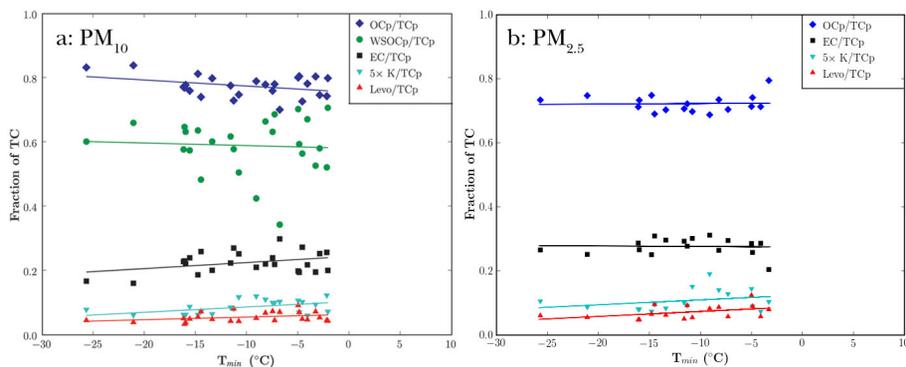


Fig. 4. Variation of ratio X/TC_p versus temperature for various pollutants for PM_{10} (a) and $PM_{2.5}$ (b).

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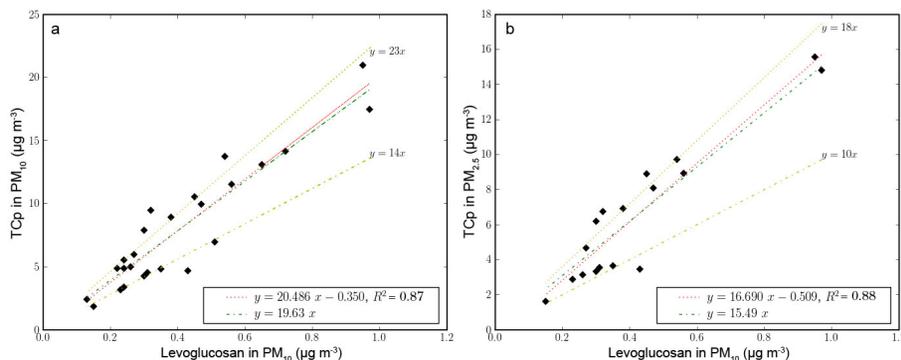


Fig. 5. Total carbon (TC_p) in PM₁₀ (a) and PM_{2.5} (b) versus levoglucosan, as well as regression lines, a central line (no intercept) is given, and curves to capture the likely range of TC_p/levoglucosan ratios indicated.

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