

Interactive comment on “Carbonaceous aerosols at urban influenced sites in Norway” by K. E. Yttri et al.

K. E. Yttri et al.

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Reply to referee 1

We would like to thank the reviewer for the positive comments made to our manuscript and for the effort made to clarify and improve its content.

General comments made by the referee:

The particle samples of this study have been collected in series at three sampling sites (curbside/Oslo: Sept 9 to Oct 3, 2002; urban background/Oslo: Nov 21 to Dec 14, 2001; suburban/Elverum: Jan 30 to Mar 15, 2002 and Mar 22 to June 28, 2002). This rise up a few questions: How long was the gap between sampling and analysis? May the chemical composition of PM sample have changed during the long storage?

Answer: The samples were analyzed within one to three months after collection, which

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is an acceptable lag time as long as the filters have been stored properly. Thus, there is no reason to suspect changes in the chemical composition due to long term storage, as this has not taken place. This additional information has been added to the section 2.2.1:

All samples were analyzed within 1 - 3 months after collection.

One objective of the study is to address the spatial and seasonal variation of the carbonaceous sub fractions. How is it possible to investigate the spatial variation on a basis of this dataset, since the sampling campaigns have been carried out sequentially, not simultaneously in different sampling sites? In addition, the seasonal variation can only be investigated at the suburban site (Elverum). These things should be made clear in the paper. How well do the selected sampling periods represent the actual season (winter Jan 30 to Mar 15, summer Mar 22 to June 28)? See also early fall and late fall (p. 19498, r 2).

Answer 1: To use the term spatial resolution does not require that measurements are performed simultaneously at the various sites. However, in order to avoid any confusion regarding this we have rephrased the sentence:

Samples were collected at three different sites in Norway, and at different times of the year to reflect the influence of various sources of carbonaceous particulate matter.

Answer 2: The mean ambient temperature observed during the various measurement periods were within 1 standard deviation of the mean ambient temperature reported for the nearest meteorological station. Thus, the sampling periods seem to reflect that of the actual season rather good. For both sites the average temperature was calculated for the period 1961 -2008.

We have included the following sentence to clarify this:

The mean ambient temperatures observed during the various measurement periods were within one standard deviation of the mean ambient temperature reported for the

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nearest meteorological station, thus reflecting the seasonal climatology rather well.

Specific comments (p and r indicates the page number and row of the ACPD paper):

P 19490, r 8: One reference should be added in urban studies carried out in Scandinavia: Sillanpää M. et al. (2005) Organic, elemental and inorganic carbon in particulate matter of six urban environments in Europe. Atmos. Chem. Phys. 5, 28698211;2879, 2005.

Answer: The reference to Sillanpää et al. (2005) has been included:

Sillanpää M., Frey, A., Hillamo, R., Pennanen, A.S., Salonen, R.O.: Organic, elemental and inorganic carbon in particulate matter of six urban environments in Europe. Atmos. Chem. Phys., 5, 2869-2879, 2005.

P 19491, r 11-15: Unfortunately, the reviewer is not familiar with the inlets used in this study and he had no access to the report made by Marsteen and Schaug (2007). For the interpretation of the results, it is interesting to know how much the PM10 concentrations based on NILU-PM10 and GENT- PM10 inlets deviate from that of the reference method (defined in CEN 12341).

Answer: The EN12341 standard requires that the calculated reference equivalence function is bounded within the limits of the $y = x \pm 10$ acceptance envelope when compared with the reference data (x), and that the variance coefficient R2 is > 0.95 .

When comparing the NILU sampler and the Gent sampler with that of the reference sampler, we find that they both meet the criteria and thus are equivalent to the reference sampler for PM10 at levels lower than $100 \mu\text{g m}^{-3}$.

Orthogonal regression of the NILU and Gent samplers data with the reference data set

$\text{NILU} = a * \text{Reference} + b$

$a = 1.04$; $\text{CL}_{95\text{low}} = 1.02$; $\text{CL}_{95\text{high}} = 1.07$; $b = -0.31$; $\text{CL}_{95\text{low}} = -0.85$; $\text{CL}_{95\text{high}} = 0.23$; $R^2 = 0.98$

Gent= a* Reference + b

a = 1,07; CL95low = 1.04; CL95high = 1.10; b = -0.56; CL95low = -1.19; CL95high = 0.07; R2 = 0.98

Based on the request of the referee we have extended the following sentence in section 2.2.1:

Both samplers have proven equal to the CEN 12341 standard (Marsteen and Schaug, 2007), which requires that the calculated reference equivalence function is bounded within the limits of the $y = x \pm 10$ acceptance envelope when compared with the reference data (x), and that the variance coefficient R2 is > 0.95 .

P 19491, r 21-22: The NILU-SFU dichotomous sampler collects the coarse particles (PM_{2.5-10}) on the initial filter and the fine particles (PM_{2.5}) on the second filter. If the reviewer understood correctly, the fine particles go through the initial filter, which cause the upper cutoff size of the fine particles. If this is the case, the cutoff size must be sensitive for the loading of initial filter. Could the filter loading explain partly the surprising finding described in section 3.2 (page 19496, rows 23-29)?

Answer: Filter with pores straight-through (the filter) has size segregating features and can be used to separate particles of different sizes. A rule of thumb (See Hopke et al., 1997) using the NILU-SFU dichotomous sampler is that if the sum of the particulate mass collected on the two filters $< 800 - 1000 \mu\text{g}$, then the flow rates remain sufficiently high so that the sampler performance is reliable. When the total mass is larger, then the flow rate in the SFU will start to decrease and its size segregating feature will start to decrease.

In the present study, three of the collected samples exceeded $800 \mu\text{g}$, whereas none exceeded $1000 \mu\text{g}$. However, only one of the samples collected during the period in question (10-13.12.2001) exceeded $800 \mu\text{g}$; i.e. 12-13.12.2001. For the two other samples with mass loadings exceeding $800 \mu\text{g}$, no high levels of OC and EC in the coarse

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fraction was observed. Based on these data, it cannot be concluded that the high levels of EC and OC in the coarse fraction of PM₁₀ for the period 10-13.12.2001 are due to sampling artefacts. On the other hand, we cannot totally exclude this possibility either, thus we have included the following sentence:

The sampling technique (see section 2.2.1.) used to separate the coarse and the fine fraction of PM₁₀ at the urban background site is subject to uncertainties at particularly high mass loadings, however the samples collected are within the suggested safe range reported in the literature. Thus, we can only speculate about the potential influence of sampling artefacts on our findings.

P 19491-2, section 2.2.1: The number of samples collected during each sampling campaign should be given either in text or in one of the tables. Were the samples collected also during weekends? (According to Figure 2, 2 to 4 samplings were carried out in a week).

Answer: Sampling was performed on weekdays only. The samples was collected according to the following scheme: Monday - Tuesday, Tuesday - Wednesday, Wednesday - Thursday, and Thursday - Friday. The number of samples collected at each site has been included in the the text in section 2.1.:

Sampling was performed on weekdays only. The number of days subjected to sampling at each of the sites was: 14 for the urban background site, 15 for the curbside site, 22 for the suburban site during the summer campaign, and 25 for the winter campaign at the suburban site.

P 19491-2, section 2.2.1: Since the sampling methods vary to some extent between both the sampling campaigns and different analysis, the uncertainties of presented results should be estimated in the paper.

Answer: A comparison of the inlets used for collection of PM₁₀ to that of the reference method was already requested by the referee (See answer given to P19491, r 11-15).

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This comparison shows that the two PM10 samplers used for collection of aerosol filter samples for subsequent thermal optical analysis is within the limits of the $y = x \pm 10$ acceptance envelope (red lines in Figure 1) when compared with the reference data (x), and that the variance coefficient R^2 is > 0.95 . At the time this study was performed no reference method for PM2.5 approved by CEN was established, thus any such comparison does not exist for PM2.5. However, both samplers used for collection of PM2.5 are widely used and commonly reported in the scientific literature.

Two types of tandem filter sampling was used to account for the positive artefact of OC; namely the QBQ-approach and the QBT-approach. The QBQ approach was applied for one of the sites (Urban background site, PM10), whereas the QBT approach was used for the others. At the urban background site, parallel sampling using both approaches was performed for PM2.5. As stated in section 3.1 (page 19494 and 19495, and in particular line 5 - 9) with reference to the scientific literature (e.g. Subramanian et al. 2004) on this topic and to the present study, the QBT approach provides a higher estimate of the positive artefact than that of the QBQ-approach. For PM2.5 at the urban background this difference has been quantified (See Page 14945, line 7 and 8). Further we have aligned the positive artefact of OC in PM10 at the urban background site (using the QBQ-approach) with the other sites, which employed the QBT approach, by using the OCB concentration of the PM2.5 sampler operated in parallel and at the same filter face velocity as the PM10 sampler (See Table 2 in the manuscript). This exercise raised the estimate of the positive artefact of OC from 12 % to 29 %.

P 19492, r 1: The QBT approach should be described in more detail. Teflon filter were hardly analyzed with the TOT.

The following sentence has been included to avoid the potential misunderstanding that Teflon filters in a QBT set up is subjected to TOA-analysis:

The QBT approach requires two identical samplers operated in parallel, of which one sampler is loaded with a single quartz fiber filter and the other with a Teflon filter in front

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of a quartz fiber filter.

P 19492, section 2.2.3: What was the upper cut-off size in sampling of potassium analysis? Answer: The filter pack used to collect filter samples for subsequent analysis of potassium had an open filter face. According to EMEP (1995) the cut off of this filter pack is just in excess of 10 μm EAD. Parallell sampling of PM10 and TSP at the suburban site in winter showed no difference in mass concentration between the two size fractions, thus it is unlikely that the open filter face filter pack would be collecting potassium- containing particles larger than that of PM10.

The following sentence has been included:

According to EMEP (1995) the cut off of the open face filter pack is just in excess of 10 μm EAD. Parallel sampling of PM10 and TSP at the suburban site in winte showed no difference in mass concentration between the two size fractions, thus it is unlikely that the open filter face filter pack would be collecting potassium- containing particles larger than that of PM10.

P 19498-19501, section 3.3.2: The authors are recommended to compare their results to those published recently by Saarikoski et al. (2008) Sources of organic carbon in fine particulate matter in northern European urban air. Atmos. Chem. Phys., 8, 6281-6295, 2008

The following reference to Saarikoski et al. (2008) have been made in section 3.3.2.:

The range observed at the urban background site correspond with that reported by Saarikoski et al. (2008) for an urban background site in Helsinki for PM1, where OC-wood constituted 41 +- 15 % of OC in winter and 20 +- 4 % in fall.

The following reference to Saarikoski et al. (2008) have been made in section 3.4.:

Indeed, Saarikoski et al. (2008) reported that SOA constituted 78 +- 8 % of WSOC in PM1 during summer at an urban background site in Helsinki.

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P 19503-19504, section 3.4: The authors are recommended to compare their results to those published recently by Timonen et al. (2008) Size distributions, sources and source areas of water-soluble organic carbon in urban background air. Atmos. Chem. Phys., 8, 5635-5647, 2008

The following reference to Timonen et al. (2008) has been made in section 3.4.:

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; Timonen et al., 2008).

P 19516, Table 3: How have the particulate WINSOC and WSOC been determined? Are they intentionally labeled particulate indicating the positive-artefact-corrected results? However, it is assumed that the WINSOC and WSOC have not been corrected for positive sampling artefact, and therefore particulate and subscript P should be removed from these two OC fractions.

Answer: The p - index of WSOC (WSOC_p) and WINSOC (WINSOC_p) is used deliberately. Five backup filters from each of the campaigns were subjected to water extraction in order to enable an estimate of particulate WSOC and WINSOC. For each campaign the mean WSOC/OC and WINSOC/OC ratios for the selected backup filters was established. These ratios were subsequently used to calculate the WSOC and the WINSOC content of OC on all the backup filters from each of the campaigns. Hence, the calculated WSOC and WINSOC content of the backup filter were subtracted from that of the front filter to provide WSOC_p and WINSOC_p. We have included the following sentence in the paper to clarify this:

Five backup filters from each of the campaigns were subjected to water extraction in order to enable an estimate of particulate WSOC (WSOC_p) and WINSOC (WINSOC_p). For each campaign the mean WSOC/OC and WINSOC/OC ratios for the selected backup filters were established. These ratios were used to calculate the WSOC and the WINSOC content of OC on all the backup filters. Hence, the calculated WSOC and

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WINSOC content of the backup filters were subsequently subtracted from that of the front filter to provide WSOCp and WINSOCp.

P 19518 and P 19524: Are Table 5 and Figure 4 necessary for the paper?

Answer: In our opinion Table 5 and Figure 4 should be kept. We argue that the regression data from Table 5 and Figure 4 are important to the paper and we think it is important to show the data points as well, since it both helps the reader visualize the arguments given in the text, and provides much more information on the validity of the regression analysis than a simple correlation coefficient and a slope alone can provide.

Technical corrections (P and r indicates the page and row number of the ACPD paper)

P 19489, r 5-7: The sentence is unclear. Check and revise it.

Revised sentence:

The carbonaceous aerosol contains a large number of organic species, but the majority remains yet to be identified. The presence of well-known toxics has been reported, but the scientific community is still grappling with what causes the ambient aerosol toxicity. However, studies seem to point towards combustion particles, which are enriched in carbonaceous material, when ascribing the adverse health effects on humans following exposure to ambient aerosols (Künzli et al., 2000; Hoek, et al., 2002; Forsberg et al., 2005; WHO, 2003).

P 19489, r 9: negative should be adverse.

See previous correction

P 19489, r 11: predicted should be assumed.

predicted has been changed to assumed

P 19489, r 13-15: Along with black carbon ... the carbonaceous aerosol ... rephrase the sentence.

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The original sentence has been removed and replaced by the following:

When studying aerosol impact on climate, the largest uncertainties by far are associated with the effects of the carbonaceous aerosol. This is mainly attributed to the black carbon part of the carbonaceous aerosol, which absorbs solar radiation in the atmosphere. According to Ramanathan and Carmichael (2008), this feature has made black carbon the second most important contributor to global warming after carbon dioxide. However, the climate effect of black carbon is uncertain and debated (Forster et al., 2007).

P 19490, r 19: the spatial and seasonal variation see the general comment 1 and revise accordingly.

Revised sentence: Samples were collected at three different sites in Norway, and at different times of the year to reflect the influence of various sources of carbonaceous particulate matter.

P 19491, r 27: semi volatile should be volatile and semivolatile

The requested changes have been performed.

P 19496, r 9: Start new paragraph from A pronounced seasonal variation....

The requested change has been performed.

P 19496, r 22: semi volatile should be semivolatile

The requested changes have been performed.

P 19497, r 1: during favorable could be for example under certain favorable has been changed to stagnant

P 19497, r 15-16: Rephrase the sentence The aerosol content...

The sentence has been rephrased.

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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. E.g. the mean concentrations of TCp at the curbside and the urban background site are 1.2 - 1.5 times higher than the highest monthly mean reported for Helsinki.

P 19497, r 18: Rephrase here we find.

See the previous correction.

P 19499, r 5: wood should be written as a subscript (compare row 1 above)

The requested change has been performed.

P 19499, r 12-13: Rephrase the change in concentrations level says little about any change in emissions

The sentence has been rephrased:

Changes in the ambient concentration levels do not necessarily reflect a change in the emissions, as meteorology also has a strong influence. However, an examination of the ratios between pollutants contains valuable information.

P 19499, r 23: Rephrase Results for daily mean temperature, not shown, are similar

The sentence referred to has been removed from the manuscript. The content of the removed sentence is valid, though.

P 19499, r 26: Rephrase ... for the case of Potassium against T_{min} for PM_{2.5}. Even here ...

The sentence has been rephrased: The slopes of the regression lines are close to zero and only significant ($p < 0.05$) for the case of Potassium/TCp versus T_{min} for PM_{2.5}., but the slope is very small (0.0003).

P 19499, r 24 and P 19500, r 2: both size fractions should be both PM size fractions

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The sentences have been changed according to the suggestion made by the referee.

P 19500, r 1 and P 19500, r 11: Replace good

Good has been replaced with high.

P 19500, r 10: Rephrase ... implying that the changes in levoglucosan alone are more than sufficient to explain the variations in TCp.

The sentence has been rephrased:

The intercept of the least-square fit (TCp versus LG) is slightly negative on the TCp axis, implying that the changes in levoglucosan alone are more than sufficient to explain the variations in TCp.

P 19500, r 22-23: over 99

The sentence has been changes to:

These results are fully consistent with emission inventory estimates for Elverum, which states that 99 % of the PM10 in this town arises from wood combustion.

P 19501, r 25: Rephrase caution should be used

They sentence has been changed:

However, caution should be made when extrapolating levoglocosan-to-OCp ratios from wintertime conditions to summer, as levoglucosan is likely to originate from diverse sources in summer, such as open agricultural waste burning and forest fires.

P 19501, r 27, P 19503, r 24 and P 19505, r 3: Replace Still

P 19501, r 27: Still has been replaced by Nevertheless.

P 19503, r 24: Still has been replaced by Yet.

P 19505, r 3: Still has been replaced by However.

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P 19502, r 6: was should be were
was has been replaced by were

P 19502, r 12: revise which both

The sentence as been rephrased:

The K+/levoglucosan ratio 0.32 is in the upper range of that reported by Fine et al. (2001; 2002a; 2002b; 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 are cities considerably influenced by residential wood burning, as reported by Wang et al. (2006).

P 19514, P 19516: Modify Table 1 and 3 so that they are more legible.

We agree that the content in tables 1 and 3 are small in the printed version. However, comments were made in the first round, which allows for technical corrections only, that some tables ought to be merged. In the final version of the paper it might be an option to present these tables, in particular Table 3, in landscape format. However, we leave this for the editorial/production office to decide.

P 19518: Give the values of slope and intercept in two significant numbers.

The slope and the intercept will be presented with two significant numbers according to the request made by the referee.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 19487, 2008.

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