

Response to Anonymous Referee #2

The referee's comments are in italics, our responses in plain font.

This manuscript presents results from long-term measurements of PM mass, EC, OC, and WSOC concentrations in PM10 filter samples collected at the Zotino Tall Tower (ZOTTO) in Siberia over the period of 5 years (2010-2014). These measurements are also complemented with CO measurements. The manuscript is well written and data has been adequately discussed. However, there are a few shortcomings which needs to be addressed before considering this manuscript for publication.

We thank the Referee #2 for the constructive criticism and suggestions for improvement that were taken into account upon manuscript revision. Responses to individual comments are given below.

Specific comments:

(1) Abstract is too long and shall be shortened by 30 or 40%.

Done

(2) Page 8, line 5: how the uncertainty on PM mass determination was assessed?

Include this information in the text.

We re-checked the uncertainty of the PM determination using a Mettler-Toledo micro balance model XP6. The Table below shows typical weighing results from four aerosol-loaded quartz filters. The same measurement uncertainty (even better) was observed for the back filters. Based on the gravimetric measurements the PM uncertainty was estimated at 10 µg. Thus, we replaced the initial value of 3.5 µg by 10 µg.

Table. Weighing protocol (mg)

112.386	110.787	111.968	116.159	
112.381	110.786	111.948	116.167	
112.397	110.798	111.964	116.174	
112.388	110.790	111.960	116.167	Aver
0.0067	0.0054	0.0086	0.0061	Stdev

(3) WSOC was measured indirectly, which can be a source of significant uncertainty.

The method used here may overestimate WSOC because 12 h soaking of filter in deionized water would remove water-soluble fraction of OC, however, at the same time some insoluble OC may also come out of the filter, which will be estimated as WSOC.

How the reliability of this method was tested? This information shall be included in the text.

The reliability of the method for WSOC analysis was tested by comparison with a TOC analyzer (TOC-V_{CPH}, Shimadzu), which allow to determine WSOC in the aqueous extracts directly. For this method aqueous extracts were prepared by manual shaking of the filter punches during 5 min, after which it was allowed to stand for 30 min. Figure 1 shows that both methods are in agreement, in spite of the fact that extraction time was different (12 h versus 30 min). A good

consistency between TOT and TOC- V_{CPH} methods were also obtained by Timonen et al. (Boreal Environ. Res. 13, 335-346, 2008; Fig. 2).

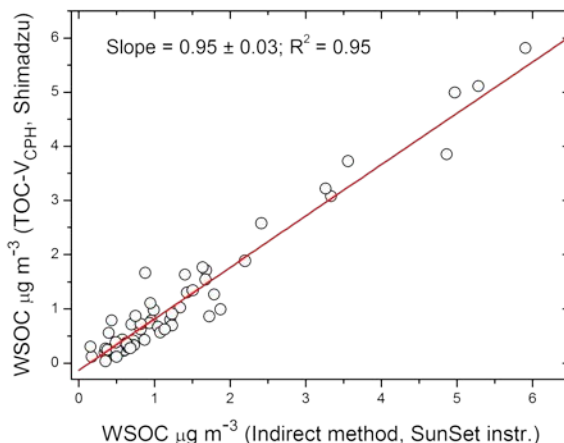


Figure 1. Comparison of measured WSOC concentrations between TOT and TOC- V_{CPH} method.

The following clarifying text has been added:

In addition to the TOT method, a TOC- V_{CPH} analyzer (5000 A, Shimadzu) was also used for WSOC analysis. A two-step procedure consisting of measurements of water-soluble total carbon (WSTC) and water soluble inorganic carbon (WSIC) was applied. WSOC is then calculated as a difference between WSTC and WSIC (Chi et al., 2009). The TOT and TOC- V_{CPH} measurements of WSOC concentrations cover the date range from April 2010 to December 2011. In general, the agreement between the two methods during this time period was within 10%. Due to fatal technical problems with the TOC- V_{CPH} , after December 2011 WSOC was measured only by the Sunset instrument. The estimated error of the WSOC concentrations using the TOT method is 10% - 15%, depending on the filter loading, which results in a 12-17% error for the WSOC/OC ratio.

(4) Uncertainty in WSOC measurements will also affect the WSOC/OC ratio reported in this study, which shall be discussed in the text.

This information has been added to the text (see previous response).

(5) To convert OC into OM, a conversion factor of 1.8 has been used, which is another source of uncertainty in estimated TCA. This conversion factor is likely not uniform throughout the study period. It may vary from 1.4 to 2.2 (Turpin and Lim, AS&T, 2001). This fact shall be mentioned in the text while discussing TCA or TCA/PM data.

The following text has been added:

Organic matter (OM) was estimated as $1.8 \cdot \text{OC}$. The same OC-to-OM conversion factor of 1.8 had been used in the SMEARII (Finland) (Maenhaut et al., 2011a) and K-puszt (Hungary) (Maenhaut et al., 2008) remote coniferous forest sites, providing the best agreement in the aerosol chemical mass closure calculations. As a result, the total carbonaceous matter (TCM) was calculated as $\text{TCM} = 1.8 \cdot \text{OC} + \text{EC}$. It should be noted that there is considerable variability in reported OM/OC ratios for organic compounds depending on the relative contribution of primary and secondary organic aerosol sources, with reported values ranging from 1.2–2.4 (Turpin and Lim, 2001). In this study OM and TCM are estimated and used mainly to illustrate their temporal variability. However, as will be shown below, the obtained estimates of the TCM/PM₁₀ ratio are reasonably consistent with published values for the sources of the pollution plumes.

(6) Page 10, line 25: WSOC/OC ratios are given in %, which looks odd. Ratio should be written in fraction form e.g. 65% should be written as 0.65.

Done

(7) At many places in the manuscript, sometime abbreviations are used and sometime the full words are used. Use abbreviations only after defining them when they appear for the first time.

Done

(8) Fig. 3: y-axis is on log scale. Start y-axis from 1 rather than 0.1 so that EnF are prominently visible.

Done

(9) Interpretation of Fig. 15 doesn't look very convincing as there is no significant effect of temperature on monoterpene emissions up to 15 °C or so, whereas OC/PM ratio shows the observed trend for the temperature ranging from 0 to 15 °C.

We do not agree. Our interpretation of the observed near exponential growth of OC/PM ratio due to biogenic activity seems quite reasonable.