Atmos. Chem. Phys. Discuss., 13, C4733–C4737, 2013
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Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Primary and secondary organic aerosol origin by combined gas-particle phase source apportionment" *by* M. Crippa et al.

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Received and published: 12 July 2013

Answers to Referee 2

- as shown in Figures 6 and SI 1.2, the SVOOA components (day and night) are not clearly differentiated for some days (i.e 27th July), peaking both components simultaneously. Is any explanation for this?

As mentioned at page 8557 (lines 9-15) during some events these SOA components showed a similar trend. We attribute this to the role of meteorology. As depicted in Fig. 6, during Atlantic polluted periods both the secondary sources and the HOA factor showed an increased mass concentration, meaning that during these events all factors were partially enhanced due to the contribution of polluted air masses. This

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interpretation is therefore applicable to the event happening on the 27th of July.

-Although the methodology proposed permits a better identification of sources of organic aerosols there are some aspects still unclear. Thus, inclusion of PTRMS did not permit to better identify the cooking source.

The Reviewer's comment is relevant; the PTRMS only measured a selection of fragments which unfortunately did not include characteristic markers for cooking emissions, preventing the identification of gas-phase compounds typical of this source. This is mentioned at page 8552 line 23-26 of the manuscript:

"The lack of a dominant contribution from a particular VOC in Table 1 also indicates that the selected subset of PTR-MS masses does not contain a good cooking marker. Future high resolution PTR-MS measurements would indeed provide a more resolved and comprehensive information on the gas-phase composition, which may aid the identification of specific cooking emission markers.

-Methodology: some information is needed. Indicate in this section instruments and sampling locations used during each campaign. Please, specify height about ground level. Provide some information about meteorology during campaigns; this would help for interpretation.

Details about the instrumentation and meteorological conditions were added in the methodology section as following:

A PM10 inlet was located at $\sim$ 6 meters above ground level and a comprehensive suite of particle and gas-phase instrumentation was deployed at the site. Details about the instruments used at the LHVP site during the summer 2009 and winter 2010 campaigns can be found in Freutel et al. (2013) and Crippa et al. (2013a), respectively. Here we primarily discuss particle composition measurements from an Aerodyne aerosol mass spectrometer (AMS) and VOC measurements from an Ionicon proton transfer reaction mass spectrometer (PTR-MS, Ionicon Analytik, Innsbruck, Austria). Meteorological

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conditions were significantly different during the two campaigns. Marine air masses influenced the continent during the summer period resulting in very low PM concentrations, while continental air masses strongly affected the air pollution in Paris during wintertime (especially in the middle and at the end of the campaign).

- Page 13 L 40-please, delete (SV-OOA day)

SV-OOA day was removed as suggested by the Reviewer.

- Page 13 L 40-46 - Isoprene may be also emitted by anthropogenic sources such as oil and wood combustion, gasoline, tobacco etc, (Adam T, et al, 2006, Chem Res Toxicol 19(4), 511-520).

While we recognize that some combustion sources may emit isoprene, it is well established that during summer time isoprene is overwhelmingly related with biogenic emissions. More clarifications are added in the corrected version of the manuscript.

"SV-OOAday exhibited a correlation with ambient temperature (SV-OOAday). Additional correlations with isoprene and its oxidation products suggest that this factor arises in large part from locally-formed biogenic SOA (although isoprene may be also emitted by anthropogenic sources such as oil and wood combustion, gasoline, tobacco etc. (Adam et al., 2006)). Isoprene (m/z 69), MVK/methacrolein (m/z 71) and monoterpenes (m/z 137) were the characteristic VOCs of this factor, in agreement with results reported by Jordan et al. (2009). Isoprene and monoterpenes are in fact emitted by plants during daytime and with increasing temperatures, while MVK and methacrolein are the major isoprene oxidation products. These results are in agreement with 14C measurements performed during the MEGAPOLI summer campaign which show summertime SOA being primarily non-fossil (80%) (Beekmann et al., 2013)."

- Section 3.2.2 It is too descriptive. It should be better explained the differences and similarities between the two methods. It is not clearly stated which AMS database is used for the PMFPTRMS-AMS analysis

Section 3.2.2 describes the comparisons between different source apportionment methods which are previously discussed in the methodological section 2.3. Therefore the reader should refer to section 2.3 for the description of the methods, while in section 3.2.2 only results from different source apportionment procedures are reported. Concerning the PMFPTRMS-AMS solution at page 8560 (line15) it is now clearly stated which results are presented for the PMFPTRMS-AMS: "(PMFAMS-PTRMS (six sources, present study))". The AMS datasets used for the combined source apportionment are the unit mass resolution MS at LHVP, as discussed in section 2. More clarifications on the limitations of the approaches used are added in the corrected manuscript, following the comments of Referee 1 too.

"However, a combined gas-particle phase source apportionment is a critical technique since involves species with different lifetimes and several dynamic processes. This approach is suitable for a clearer identification of primary sources, where particulate and gaseous pollutants are co-emitted. On the other hand, secondary gas and particle phase species form and decay at different timescales; hence their covariance does not allow discrimination between different secondary sources, but may be rather used to infer the formation timescales and lifetimes of OOA species. Such methodology has been successfully used on several works (Slowik et al., 2010;El Haddad et al., 2012), which gave valuable insights into the formation and aging processes of OOA."

- Page 16-L18. As far as I understand, the SVOOA night component was identified during the summer period but not during the winter period. However, a contribution of 18% was estimated for this source in winter. It is not clear to me how this value was calculated.

During summertime two SV-OOA components were separated, as described in the manuscript:

"By adding the gas phase species into the PMFAMS-PTRMS analysis, these processes were decoupled yielding two SV-OOA factors: SV-OOAday and SV-OOAnight. On average, SV-OOAday mass builds steadily during the day, despite the development of the boundary layer, and significantly correlates with ozone and methacrolein + methyl vinyl ketone (m/z 71), short-lived early generation products of isoprene oxidation. This factor can be interpreted as stemming from the production of short-lived secondary organic compounds during peak photochemistry. By contrast, SV-OOAnight contribution is enhanced during nighttime with the temperature decrease and the increase of relative humidity. This suggests that similar to nitrate this factor may be related to the partitioning of semi-volatile SOA into the particle phase."

During wintertime it was possible to separate only one SV-OOA component, correlating with nitrate and therefore having a similar behavior to the SV-OOAnight identified during summer. The word "night" was attributed only to the summer case to distinguish between the two SV-OOA components identified: SV-OOAnight and SV-OOAday.

- Figure 5. Please, improve quality of this figure (y-axis and resolution)

The quality of Fig.5 was improved as suggested by the Reviewer.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C4733/2013/acpd-13-C4733-2013-supplement.zip

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