Atmos. Chem. Phys. Discuss., 9, C12106–C12123, 2010 www.atmos-chem-phys-discuss.net/9/C12106/2010/ © Author(s) 2010. This work is distributed under the Creative Commons Attribute 3.0 License.



Interactive comment on "Organic aerosol components observed in worldwide datasets from aerosol mass spectrometry" by N. L. Ng et al.

N. L. Ng et al.

ng@aerodyne.com

Received and published: 14 April 2010

We thank the reviewers for their comments. We have made changes to the text to address these comments. There are three main concerns raised by both reviewers:

1. Abstract/Introduction should emphasize the main idea/uniqueness of the paper more clearly, as many of the datasets have been used in other papers.

This is a good point. In this study we compile and present factor analysis results from 43 ambient AMS datasets. Of these datasets 27 are reanalyzed as part of this work and include a majority of the sites in Zhang et al. (2007) and Jimenez et al. (2009). In this work we focus mainly on the OOA component and the reanalysis allows for further deconvolution of the total OOA component reported by Zhang et al. (2007) into LV-OOA

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



and SV-OOA components. The OA components obtained from this work were used by Jimenez et al. (2009) to form the basis of a modeling framework that links oxidation and volatility to capture the evolution of OA in the atmosphere. In this manuscript we integrate the factor analysis results from the ambient datasets with each other as well as with laboratory data to obtain a holistic view of how the AMS component mass spectra change with atmospheric aging. The components, when taken together, show a continuum of OOA properties in ambient aerosol. The changes in f44 (ratio of m/z 44 to total signal in the component mass spectrum) and f43 (ratio of m/z 43 to total signal in the component mass spectrum) and f43 (ratio of m/z 43 to total signal in the component mass spectrum) in particular, are used to develop a new mass spectral diagnostic (the f44-f43 space) that can be used to classify the atmospheric aging of OA components in the atmosphere.

The abstract is modified slightly to read as:

"In this study we compile and present together results from the factor analysis of 43 aerosol mass spectrometer (AMS) datasets (27 of the datasets are reanalyzed in this work). The components from all sites, when taken together, provide a holistic overview of Northern Hemisphere organic aerosol (OA) and its evolution in the atmosphere. At most sites, the OA can be separated into oxygenated OA (OOA), hydrocarbon-like OA (HOA), and sometimes other components such as biomass burning OA (BBOA). We focus on the OOA components in this work. In many analyses, the OOA can be further deconvolved into low-volatility OOA (LV-OOA) and semi-volatile OOA (SV-OOA). Differences in the mass spectra of these components are characterized in terms of the two main ions m/z 44 (CO2+) and m/z 43 (mostly C2H3O+), which are used to develop a new mass spectral diagnostic for following the atmospheric aging of OA components in the atmosphere. The LV-OOA component spectra have higher f44 (ratio of m/z 44 to total signal in the component mass spectrum) and lower f43 (ratio of m/z 43 to total signal in the component mass spectrum) than SV-OOA. A wide range of f44 and O:C ratios are observed for both LV-OOA (0.17 ± 0.04 , 0.73 ± 0.14) and SV-OOA (0.07 ± 0.04 , 0.35 ± 0.14) components, reflecting the fact that there is a continuum of OOA properties

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



C12108

in ambient aerosol. The OOA components (OOA, LV-OOA, and SV-OOA) from all sites cluster within a well-defined triangular region in the f44 vs. f43 space..."

The following paragraph in the introduction

"In this work, we apply factor analysis on 27 AMS datasets collected at many locations in the Northern Hemisphere over the last ten years, encompassing the majority of the sites included in Zhang et al. (2007) and Jimenez et al. (2009)."

is changed to:

"We present results from the factor analysis of 43 AMS datasets. As part of this work 27 of the datasets, which encompass a majority of the sites in Zhang et al. (2007), are reanalyzed. We focus mainly on the OOA component and the reanalysis allows for further deconvolution of the total OOA component reported by Zhang et al. (2007) into LV-OOA and SV-OOA components. The OA components resulting from this work were used by Jimenez et al. (2009) to form the basis of a modeling framework that links oxidation and volatility to capture the evolution of OA in the atmosphere. In this manuscript we combine the factor analysis results from the ambient datasets together to obtain a holistic view of how the AMS ambient component mass spectra change across environments with different sources and aerosol processes. The common features of the component spectra are used to develop a new mass spectral diagnostic for following the atmospheric aging of OA components in the atmosphere. Finally, since the AMS has been employed in many laboratory experiments over the years, a series of chamber data (both published and unpublished) are also integrated and compared to the ambient data. Chamber data provide the basis for simulating SOA formation in the atmosphere. Hence, it is important to evaluate whether the results from chamber experiments are representative of the atmosphere; similarities and differences between ambient OOA and laboratory SOA are examined and discussed."

2. The naming convention of the two OOA components (LV-OOA and SV-OOA):



9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- The naming convention is relative.

- These terms describe organic mass spectra that differ in quality as determined through use of a mathematical separation method, cannot be used as if they are site-independent quantities.

- The direct measurement is O:C, not volatility

It is fair to simply call the highly oxidized OA "HO-OA" and medium oxidized OA "MO-OA" (as suggested by Reviewer 1). Indeed, prior to introduction of the terms LV-OOA and SV-OOA, the two OOA sub-components were referred to as OOA-1 and OOA-2. We agree that the terminology HO-OA and MO-OA is more straightforward (and is more descriptive than OOA-1 and OOA-2), however, we prefer to keep the LV-OOA and SV-OOA for the following reasons.

As discussed in the Section 3.1, the assignment of LV-OOA and SV-OOA is not absolute for each site, i.e., the component with a higher f44 is referred to as LV-OOA and the component with a lower f44 is referred to as SV-OOA, regardless of the absolute value of f44. The only way to avoid any overlapping in the O:C distribution is to have an absolute cutoff threshold on the O:C or volatility scale, which is currently not available and is still being investigated.

Regardless of the terminology, there will inevitably be some overlap in the O:C distribution as long as the component analysis methods are applied separately to each dataset, making the naming system relative to the O:C spanned by the OOA in a given study. For instance, even if we simply refer to the less oxidized component as MO-OA and the more oxidized component as HO-OA, we will still have a region in which the O:C of these two components overlap. If the relative way of naming these components at each site was not useful, we would not expect to see any clear distributions in O:C and volatility when considering the results from all sites together. However, as shown in Fig. 2, when taken together, the LV-OOA and SV-OOA from all sites fall into two distinctive distributions of f44 (and O:C) with little overlap, suggesting that these two OOA

ACPD

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



subtypes do have distinctive properties and volatility distributions. One exception is the Crete dataset, in which the aerosol sampled appears to be highly oxidized throughout the whole campaign (Hildebrandt et al., 2010). Two OOA components are identified in that study, but their separation is at the limit of what PMF can do (as described in that paper) and perhaps for that reason they appear to have similar volatility based on thermodenuder measurements. However, in this case, both OOA components are highly oxidized so essentially both factors fall into the LV-OOA range. Hence, it is not surprising that they have similar volatilities.

As mentioned in the manuscript, previous studies have shown a consistent trend in which the more oxidized component is less volatile and the less oxidized component is more volatile. With coupled thermodenuder-AMS measurements, the volatility of the two different OOA components in Mexico City and Riverside are characterized directly and it has been found that the more oxidized OA component consistently exhibits a lower volatility than the less oxidized component. Moreover, in Lanz et al. (2007) and Ulbrich et al. (2009), it has been shown that there is a good correlation of the time series of OOA-1 with sulfate and OOA-2 with ammonium nitrate; and thermodenuder measurements have also shown that nitrate has the highest volatility and sulfate has the lowest volatility among all AMS standard aerosol species (Huffman et al., 2009). While direct volatility measurements for many of the sites included in this study are not available, we found that for these sites it is also true that OOA-1 (LV-OOA) correlates better with sulfate and OOA-2 (SV-OOA) correlates better with nitrate. Recently, the link between volatility and oxidation has been used to develop a modeling framework to capture the evolution of OA in the atmosphere (Jimenez et al., 2009).

As described above, an absolute scale (O:C or volatility) does not yet exist for classifying ambient OOA subtypes. Overall, the terminology LV-OOA and SV-OOA is consistent with all previous studies that involved direct volatility measurements. Thus, we prefer to keep these terms as they convey the known physiochemical properties of the OOA subcomponents better and they are consistent with the framework already 9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



proposed in Jimenez et al. (2009).

3. Figure 1 should be removed.

We agree that the mass spectra of HOA, LV-OOA, and SV-OOA have been shown in other papers; however, we think that showing these mass spectra together with the total OOA spectrum is useful in the context of this paper. Fig. 1 makes the transition of the discussion from an AMS mass spectrum to f44 and f43 clearer and easier to follow, especially for non-AMS users.

Responses to specific comments are given below. Reviewers' comments are included in italics and author responses are in plain text.

Review 1:

The paper compiles a large amount of data sets the majority of which has already been published before (e.g., Zhang et al., GRL, 2007; Jimenez et al., Science, 2009; Lanz et al., ACPD 2009). While the authors give proper credit, it is not so easy to understand what the new and original contribution of this manuscript is. Here the authors should be more clear in the abstract and introduction.

To my understanding the new concept of this paper is the introduction of f43 (the ratio of m/z 43 to total OA), and to combine f44 and f43 as a diagnostic for atmospheric aging, as in Figures 4, 5, 6, 7, and 8. This approach is new and it is shown here that the atmospheric organic aerosol compounds can be characterized very well (with respect to atmospheric aging, oxidation and volatility) by their position in the f44 - f43 space, or, as in Fig 5, in f44 - 44/43 space.

Please refer to major comment 1 response. The reviewer summarizes the new concepts presented by this paper very clearly and we have incorporated the reviewers points into the manuscript as stated in the major comment 1 response.

An additional very important point is the comparison of the field data with laboratory data from smog chamber studies (again, in the majority previously published data).

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



This aspect is not mentioned at all in the title and in the abstract only with one (last) sentence. I would suggest giving more emphasis to the laboratory data in the abstract and introduction.

The title of manuscript is changed according to the reviewer's suggestion, as discussed in response to the next comment. The last sentence in the introduction is extended to read as:

"Finally, since the AMS has been employed in many laboratory experiments in multiple laboratories over the years, a series of chamber data (both published and unpublished) are compiled and compared to the ambient data. Chamber data provide the basis for simulating SOA formation in the atmosphere. Hence, it is important to evaluate whether the results from chamber experiments are representative of the atmosphere; similarities and differences between ambient OOA and laboratory SOA are examined and discussed."

Major comments:

Change the title, for example: "Organic aerosol components inferred from aerosol mass spectrometry: Worldwide datasets and laboratory data"

This is a good point. The title is changed accordingly.

Introduction, page 27749: Lines 24 to end: What is new in this study as opposed to Zhang 2007, Jimenez 2009 and Lanz 2009? This point has to be made very clear.

Please refer to major comment 1 response.

Skip Figure 1. There are been many papers showing mass spectra of HOA, LV-OOA (prev. OOA-1), and SV-OOA (prev. OOA-2), as Ulbrich et al., ACP, 2009, or Lanz et al., ACP, 2007.

Please refer to response to major comment 3.

Combine Figures 6, 7, and 8. It is not necessary to show all individual points and all

ACPD

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



individual compounds and experiments. It would be sufficient to indicate the areas of the graph where the data points are.

These figures are combined to be Figure 6a, 6b, and 6c. However, we think that it is useful to show the data in this detail (i.e., showing individual points for individual experiments) as it would be easier for these data to serve as references for results from future experiments (which may involve various SOA precursors).

Specific comments:

Introduction

Page 27749, lines 11-23: The direct measurement of the AMS is degree of oxidation, not volatility. Wouldn't it be more straightforward to call the subcomponents HO-OA (highly oxidized OA) and MO-OA (medium oxidized OA) instead of using volatility that is a kind of "secondary" information?

Please refer to major comment 1 response.

Page 27750, line 25: ME-2? If you mention it, you should briefly explain the difference to PMF. If it is not important for this data set, don't mention it.

The following text has been added to the revised manuscript: "In contrast to PMF, ME-2 allows for a priori constraints (partial or total) on the mass spectra and/or time series of the factors (Paatero, 1999; Lanz et al., 2008)."

Method

Are the PMF results taken from the cited papers or have the datasets been newly analyzed by PMF, and if so (as I understand), why? Are there marked differences to the published results?

As mentioned in the manuscript, the detailed PMF analyses from several sites have been presented elsewhere and the results are directly taken from those previous publications. For the rest of the sites, the PMF analysis is done in this work. To clarify this, 9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the following sentence:

"Details of the factor analyses of OA spectra from several sites have been presented elsewhere (Zhang et al., 2005a, 2005b; Lanz et al., 2007, 2008, 2009; Nemitz et al., 2008; Aiken et al., 2008, 2009b; Ulbrich et al., 2009; Docherty et al., 2008; Huffman et al., 2009; Slowik et al., 2009; Sun et al., 2009). In this work, the factor analysis of the organic aerosol spectra is based on PMF (Paatero and Tapper, 1994; Paatero, 1997)."

is changed to:

"In this work we present factor analysis results from 43 AMS datasets. As part of this work, we performed PMF (Paatero and Tapper, 1994; Paatero, 1997) based factor analysis of the organic aerosol mass spectra observed at 27 of the sites. For some of the urban downwind/rural/remote sites, a hybrid of PMF/MCA approach is employed (Cottrell et al., 2008; Nemitz et al., 2008). The factor analyses of the remaining 16 sites were performed previously and the results are discussed in more detail in the corresponding publications: Beijing (Sun et al., 2009), Riverside (Docherty et al., 2008; Huffman et al., 2009), Mexico City (Aiken et al., 2008, 2009b; DeCarlo et al., 2009), Pittsburgh (Zhang et al., 2005a, 2005b; Ulbrich et al., 2009), Zurich (Lanz et al., 2007), Egbert (Slowik et al., 2009), and the great Alpine region (Lanz et al., 2009). For the sites in Lanz et al. (2009), both PMF and ME-2 are used. In contrast to PMF, ME-2 allows for a priori constraints (partial or total) on the mass spectra and/or time series of the factors (Paatero, 1999; Lanz et al., 2008)."

Results Page 27752: "the Pittsburgh data set". References are given, but please add the basic information (project name, year, season, location: : :) here. However, my suggestion above was to skip Figure 1 anyway.

With the large number of datasets included in this study, it would be cumbersome to specify the basic information of each field campaign in the text. As noted in the manuscript, the details (locations, times, previous publications, etc.) of all datasets included in this study are given in the supplementary material.

ACPD

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



Regarding the Pittsburgh dataset, the sentence is changed to read as:

"For instance, in the Pittsburgh dataset (acquired in September 2002) studied by Ulbrich et al. (2009), the variation..."

As mentioned earlier, we think that it is worth keeping Fig. 1 as it is useful in the context of this paper.

Page 27753, lines 11-18: This discussion underlines the questionability of the naming convention LV-OOA and SV-OOA. The measured value is oxidation (O:C ratio or f44).

Please refer to response to major comment 1.

27754, line 22: Where is Chebogue Point (elevation, latitude, longitude, season and year of measurement)?

As noted in the manuscript, the details (locations, times, previous publications, etc.) of all datasets included in this study are given in the supplementary material, including Chebogue Point.

The sentence is changed to read as:

"The remote/rural Chebogue Point (Nova Scotia, summer 2004) measurements show a range of O:C that appear to reflect air masses of different ages and source regions."

27755 lines 19 – 21. If Figure 1 is deleted, these two sentences can also be deleted.

As discussed earlier, we would keep Fig. 1 as it is useful in the context of this paper. Hence, these two sentences would be retained too.

27756 line 21 – end: The discussion on instrument-to-instrument variability is confusing. First, it is said "Some of the differences ... are likely due to instrument-toinstrument variability", later, the statement is " Thus, it is unlikely that this variability is enough...". Please be more precise.

To clarify, the sentence

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



C12116

"Some of the differences in the f44 and f43 values shown in Fig. 4 are likely due to instrument-to-instrument variability"

is changed to

"Some of the differences in the f44 and f43 values shown in Fig. 4 can possibly arise from instrument-to-instrument variability."

27759, line 14, and Appendix: This is basic maths. I don't think the Appendix is necessary. Just giving the result in the text on page 27759 is sufficient.

Although the math turned out to be relatively straightforward, it is actually not instantly obvious how the line in the f44 vs. f43 transforms into the curve in the f44 (and O:C) vs. 44/43 space. We think that it is still useful to include the derivation in the Appendix.

But please give in the caption of Figure 4 the slopes and intercept instead of coordinates for the two lines.

The slopes and intercepts are now stated in the figure caption.

Figure 5: According to the definition of the lines in Fig 4 and the transformed curves in Fig 5, the curves in Fig 5 approach a limit for f44 of about 0.03 (max f44 in Fig 4), corresponding to O:C = 1.2. Is there a further meaning? Does this value represent a atmospheric maximum oxidation state?

Yes, in Fig. 5 it appears the O:C increases sharply at the beginning of oxidation then plateaus around a value of 1.2, suggesting that this may represent a maximum atmospheric oxidation state. We note that CO has an O:C of 1.0 and CO2 of 2.0, so for aerosol with O:C of 1.2, decomposition to the gas-phase may be an important process countering additional oxygenation (Kroll et al., 2010, submitted).

This is added to the revised manuscript: "It appears the O:C increases sharply at the beginning of oxidation then plateaus around a value of 1.2, suggesting that this may represent a maximum atmospheric oxidation state of the aerosol."

ACPD

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Fig 6: Some points are connected with lines, some aren't.

As mentioned in the caption "The lines connecting the data points indicate the range of f44 and f43 observed in each experiment. For ïĄą-pinene ozonolysis, only the values at the end of the experiment are shown to avoid crowding the plot".

Fig 9: What the rationale in plotting only the highest f44? It is said that the reason is not to overcrowd the plot, but can't this procedure introduce a strong bias in the plot?

The highest f44 indicates the highest level of oxidation reached in each experiment. With respect to the highest f44 value, the lowest f44 value would move down and left in the plot, which essentially would show the same overall trend as the highest f44 points. With the large number of data points, showing only the highest f44 values from the experiments clarifies the trend.

Page 27762, line 20: What is meant by "time-dependent" OOA data? I don't understand this explanation. If highest lab f44 values are chosen, wouldn't it be justified to use only the highest f44 from field data?

As noted in the manuscript, the ambient OOA data include only the time periods when variable partitioning of semi-volatiles is expected (i.e. time periods when there is relatively low temperature, and/or changing temperature). The highest f44 periods do not necessarily meet this requirement. To avoid confusion, the term "time-dependent" will be omitted in the revised manuscript.

Fig 11: Plots a and b can be combined in one graph. You might consider reducing the number of points (increasing the averaging time).

Since the aerosol loadings span a wide range for these experiments, we would not be able to see the changes in f44 with respect to loadings as clearly if we were to combine all the plots into one graph.

We have increased the averaging time to reduce the number of points.

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Fig 4, 6, 7, 8: If you don't agree to combine the graphs 6, 7, and 8 into one graph, you should at least plot all 4 graphs on the same x-scale.

We have made the x axis of these 3 graphs to be the same.

Review 2:

One very important point is the use of the terms "low-volatility oxygenated organic aerosol" (LV-OOA) and "semi-volatile oxygenated organic aerosol" (SV-OOA) throughout the paper. These terms describe organic mass spectra that differ in quality as determined through use of a mathematical separation method. The authors state that what is called LV-OOA in one dataset could be SV-OOA in another due to the site dependence of this analysis. Nevertheless, the terms SV-OOA and LV-OOA are used throughout the article almost as if a site independent quantity had already been defined, which is not the case. As of yet, these designations have little or no relevance as analytical terms for classification of different organic aerosol fractions in general and as such should either be omitted or used only with great caution. If the authors insist on using these terms, then much more attention should be payed to the context and consequences of their usage. Similar misconceptions arise in making comparisons to organic compounds, for instance comparing LV-OOA to HULIS. This should not be done in the absence of a fixed definition for LV-OOA.

Please refer to response to major comment 1.

Figure 1 shows typical mass spectra for LV-OOA and SV-OOA for a single measurement site. However, they are presented in a way that might mislead the reader into assuming that these are typical mass spectra for these classes. This figure should be removed to avoid any misconception.

As mentioned in the response to major comment 3, we think that Fig. 1 is useful in the context of this paper. The mass spectra from the Pittsburgh dataset are shown as an example because the LV-OOA and SV-OOA from this site have been discussed in

ACPD

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



detail previously, and indeed the mass spectra of LV-OOA and SV-OOA from this site are pretty typical for these components.

Although section (3.3) is entitled "Comparison with laboratory data", a large part is dedicated to comparison between different chamber experiments rather than comparison between ambient and laboratory generated data. Furthermore, a large fraction of these chamber experiments were performed using very high organic mass loadings. These conditions might be irrelevant for atmospheric conditions (see, for example, Shilling et al., 2009). For instance, Figure 9 compares different chamber experiments in which biogenic precursors exceed even oxygenated organic aerosol mass concentrations in highly polluted urban areas.

We do not agree that a large of part of section 3.3 is comparing different chamber experiments rather than comparing chamber experiments with ambient data. All the chamber data are present in the f44 vs. f43 space, and compared to the triangle region in which all the ambient OOA factor fall into. The fact that most of the chamber data are in the lower half of the triangle suggests that chamber SOA is not as oxidized as ambient LV-OOA. As mentioned in the manuscript, this could arise from the limited oxidation exposure in the chamber and the higher loadings used in the experiments.

A small point of criticism is the title. Since all measurements originate from the northern hemisphere and mostly from highly populated urban areas, I don't think that "worldwide" is appropriate.

Agreed. We will change "worldwide" to "Northern Hemisphere".

Furthermore, there is no real separation between urban, rural and remote datasets. They are instead classified as either "urban" or "urban downwind/rural/remote". This suggests that all measurements have been anthropogenic influenced. It would have been much more interesting and helpful if there had been a real separation between "clean" remote, rural and urban influenced datasets, even if that would have meant a reanalysis and separation of individual datasets. 9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Most of the rural/remote sites included in this study have some anthropogenic influence. Without a global modeling study, which is outside of the scope of our manuscript, we cannot perform a direct classification of the sites into "clean" (i.e. with OA dominated by natural sources) and "urban-influenced" datasets.

Specific comments: Abstract: I think, the abstract should more clearly emphasize the main idea of the paper - the mathematical separation of aerosol mass spectra into two different types (f44, f43). This would strengthen the position of the paper and its uniqueness despite the fact that most data has already been published.

This comment has been addressed in the response to major comment 1 above.

While f44 is introduced as the ratio of m/z 44 to total signal in the component mass spectrum, this is not done for f43. I assume f43 is the ratio of m/z 43 to the total component mass spectrum?

The definition of f43 is added.

P27747, L1: "In this study we present results from the factor analysis of 43 aerosol mass spectrometer (AMS) datasets". This is in contrast to the statement in the introduction: "In this work, we apply factor analysis on 27 AMS datasets".

Among the 43 datasets, 27 of them are reanalyzed with PMF in this work. To avoid confusion, the sentence is changed to

"In this study we present results from the factor analysis of 43 aerosol mass spectrometer (AMS) datasets (27 of them are reanalyzed in this work)".

Also, "AMS" is introduced as "aerosol mass spectrometer (AMS)" while later (P27748, L6) it is redefined as Aerodyne Aerosol Mass Spectrometer (AMS).

The sentence "The Aerodyne Aerosol Mass Spectrometer (AMS) provides quantitative data on inorganic and organic aerosol species in submicron non-refractory aerosol particles with high-time resolution."

ACPD

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



is changed to

"The Aerodyne AMS provides quantitative data on inorganic and organic aerosol species in submicron non-refractory aerosol particles with high-time resolution."

P27748, L12: (Paatero and Tapper, 1994) and/or (Paatero 1997) should be cited when PMF is first introduced.

The citations are added.

P27748, L24: I assume, BB POA stands for "biomass burning primary organic aerosol"?

Yes (as mentioned in the manuscript).

P27760, L14ff: "In order to allow accurate parameterizations of SOA formation and evolution in the atmosphere, it is desirable that the laboratory conditions be as representative as possible of those in the atmosphere." How representative are the conditions in the laboratory studies? The organic mass of the biogenic precursors of these studies exceed those found in urban environments by one order of magnitude.

It is true that some of the older experiments in the combined dataset used here had relatively high loadings, however, that was typical of all earlier studies. Over recent years, the SOA community has come to realize the importance of running chamber experiments with more atmospherically realistic loadings. The more recent chamber experiments are performed with loadings which are similar to ambient concentrations in polluted areas and only about an order-of-magnitude larger than over forested regions (25 ug/m3).

Figure 1: I would suggest removing this Figure and all according statements from the manuscript. See the comments above.

As discussed in the response to Review 1, we think that Fig. 1 is useful in the context of this paper.

ACPD

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



Figure 6: The lines representing the ranges are very confusing.

The lines connecting the data points indicate the range of f43 and f44 observed in each experiment. In order to avoid overcrowding of the plot, for a-pinene only the values at the end of the experiments are shown.

Figure 10: Why is m-xylene shown in this plot? Also, since neither organic loadings nor OH exposure are responsible for the differences in f44, there is no point in color coding these data points. If the authors wanted to make the point that the f44 values from datasets are not comparable due to differences in laboratory conditions, i.e., chamber size, then why show this Figure at all? It raises the question as to how intercomparable laboratory experiments are in the first place.

This plot shows the f44 vs. OH exposure for all the low-NOx aromatics experiments. Contrary to the reviewer's comment, the purpose of this plot is to show that we can compare chamber experiments when they are performed under similar conditions. For instance, the fact that loadings for the Caltech and CMU toluene experiments are similar allows us to evaluate the effect of oxidant exposure on f44. It is clear from Fig. 10 that the larger OH exposure in the Caltech chamber results in higher f44 than in the CMU chamber.

Figure 11: By connecting the data points the plots become messy for regions where data points are dense. Also, it is unclear in which direction f44 changes. I would suggest coloring the data points with different colors representing each successive experiment, like it has been done in Figure 10 for the organic loadings.

Each of the figures in Fig. 11 is from one single experiment. Loading increases initially owing to SOA formation. For a-pinene and m-xylene photooxidation, after SOA mass peaks, loadings decrease because of wall loss.

In the revised version of the figure, the points are smoothed to make the data less dense.

ACPD

9, C12106–C12123, 2010

> Interactive Comment



Printer-friendly Version

Interactive Discussion



Interactive comment on Atmos. Chem. Phys. Discuss., 9, 27745, 2009.

ACPD

9, C12106–C12123, 2010

> Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

