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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size-resolved aerosol chemistry on Whistler Mountain, Canada with a High-Resolution Aerosol Mass Spectrometer during INTEX-B" *by* Y. Sun et al.

Y. Sun et al.

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Response to the comments of referees

We thank both referees for their thoughtful comments. We have revised the manuscript in response to the comments. Listed below are our itemized responses to each reviewer's comments, which are repeated in italic.

Response to referee 1

In " Size-resolved aerosol chemistry on Whistler Mountain, Canada with a High-Resolution Aerosol Mass Spectrometer during INTEX-B " by Sun et al., the authors report measurements of the non-refractory sub-micron aerosol chemical composition,

acquired using an Aerodyne HR-ToF-AMS. The paper itself is well written and the technical descriptions are quite thorough. The manuscript in general should be published after the following issues have been addressed by the authors.
The result section is quite short. A more detailed discussion of the results, especially of the comparison between the different events identified by the authors, would definitely increase the value of the manuscript.
In response to the reviewer's comments, we have expanded the discussions on the results in the revised manuscript, particularly "Section 3.4. Characteristics of aerosols associated with different events ". The bulk aerosol composition, elemental composition of organic aerosol (OA), and the potential sources and processes for different events are compared and discussed in more detail.

A general problem of this manuscript is the way citations are used. Care should be taken to limit references to those strictly necessary for further understanding of the work in the context in which it is mentioned. For example, on page 20754, line 24, the citation of (Zhang et al., 2004a) is not a helpful reference to read to obtain further information describing the characterization of the particle beam collimator and the inlet transmission of the AMS. It mainly makes further reference to the publications of Jayne et al. (2000) and Jimenez et al. (2003), where the original description can be found.
In addition, there are also some inaccurate citations as for Matthew et al. (2008, see also comment below). This paper doesn't highlight the issue of particle acidity, as mentioned by the author, but focuses instead mainly on the relation between particle humidity/ particle water content and the collection efficiency. References should be limited to original publications describing the information of interest in the correct context.
We agree with the referee and have removed the citations to Zhang et al. (2004a) and Matthew et al. (2008). In the revised manuscript, we also eliminated references in which the same information can be found in earlier publications.

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Even more problematic are the frequent references to two papers in preparation, (Sun and Zhang, 2008) and (MacDonald et al., 2008). It is unclear exactly what these papers will describe, and how they will support the discussion of this work. In the opinion of this referee, it is essential that all data and work upon which this publication relies to draw its conclusions be either directly presented or previously peer reviewed. Dependence of conclusions on non-reviewed/un-presented work is unacceptable and should be resolved prior to publication. This can be addressed in one of three ways:
- If the citation is essential for this manuscript, and not just a reference advertising an upcoming work, the authors should describe and include the work in the manuscript if possible.
- If it is essential, but is too extensive to be included in this manuscript, the authors should publish the other manuscript(s) prior to the publication of this manuscript.
- If it is not essential for the publication, the citation should be removed from this manuscript.
We thank the referee for suggestions. We have removed the reference to Macdonald et al. (2008), which is still under preparation. To replace this reference, we now cite Leaitch et al. (2008) and McKendry et al. (2008) as both provide additional information referenced in this manuscript.
The reference to Sun and Zhang (2008) has been submitted for peer-review. It is cited as Sun and Zhang (2009) in the revised manuscript. The citation of this reference is essential and will allow interested readers to locate this reference after it is published.

Detailed comments:
p. 20751, line 15: " The average size distributions of sulfate and ammonium both showed a large accumulation mode peaking around 500-600 nm ... "
In the abstract, the authors report a maximum between 500-600 nm D_{va} for the average size distribution for sulfate and ammonium, in the conclusion a maximum between 400-500 nm is reported. This is inconsistent and should be corrected or

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clarified.
We thank the referee for pointing out this discrepancy. In the revised manuscript, the maximum D_{va} for sulfate and ammonium has been clarified to be $\sim$ 500 nm.

p. 20751, line 15: "... peaking around 500-600 nm in D_{va} ... "
Please, introduce new symbols and abbreviations properly, or avoid them completely within the abstract, as suggested by ACP/ACPD. The audience of ACP might be diverse enough to include people not that familiar with AMS specific terms like Dva.
We now define all the symbols, abbreviations, and acronyms the first time they appear in the texts.

p. 20752, line 14: (Akimoto, 2003) is not listed in the reference section.
"Akimoto, (2003) " is no longer cited the revised manuscript.

p. 20753, line 21: "Sun and Zhang, 2008 ". This publication is referenced eight or nine times, however, no detailed description about its content is given. This is especially crucial, since this paper is in preparation, at least according to the reference section, and needs to be dealt with prior to publication of this work.
The reference to Sun and Zhang (2008) has been submitted for peer-review. It is cited as Sun and Zhang (2009) in the revised manuscript. The citation of this reference is essential and will allow interested readers to locate this reference after it is published.

p. 20754, line 9: "Details on the sampling site ... are given by MacDonald et al. (2008). " Even if the sampling site will be described in detail by MacDonald et al. (2008), it would be nice to have a short description within this manuscript, especially since MacDonald et al. (2008) is only in preparation.
A short description of the sampling site is given in the Introduction (line 74-77). It provides sufficient information relevant to this manuscript. The reference to MacDonald et al. (2008) has been replaced by Leaitch et al. (2008) and McKendry et al. (2008) in

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the revised manuscript.

p. 20754, line 12: "... at ambient pressure, which varied between 770-790 hPa during this study (see Sect. 3.1). "
Why is there a reference to paragraph 3.1? Also, how did the ambient pressure affect the inlet transmission of the instrument?
The reason of a reference to section 3.1 was that the time series of ambient pressure is shown in Fig. 3, which is discussed in section 3.1. As both reviewers raised the question, we realize that this cross-reference can be a bit confusing. We therefore have deleted such tags in the revised manuscript.
We did not characterize how ambient pressure at Whistler peak would affect the inlet transmission of the AMS. While changes in ambient pressure may lead to changes in particle velocity in the AMS and lens transmission efficiencies (Liu et al., 2007;Bahreini et al., 2008), the effects are expected to be small at Whistler peak, where the air pressure is typically 770- 790 hPa. This conclusion is also supported by the comparisons of the AMS data with the other measurements at Whistler Peak as well as from the Cessna aircraft (Fig. 2). This discussion is now included in the revised manuscript.

p. 20754, line 23: (DeCarlo et. al., 2004) is not listed in the reference section.
We thank the referee for pointing out this oversight. We have added the reference "DeCarlo et. al. (2004) " in the reference section.

p. 20756, line 20: "The HR-ToF-AMS was calibrated for inlet flow at the beginning of this study and for ionization efficiency (IE) and particle sizing every 3-5 days ..." If the instrument was calibrated that extensively, it would be useful if the authors gave a short summary of the results of these calibrations. For instance, how did the IE change during these calibrations. This would give the reader a better sense of the

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accuracy of the measurements.
In response to this comment, we have added Fig. S1 to show the IE calibration results in the supplementary information.
A short description has also been added in the revised manuscript:
"The measured IE values increased by $\sim$ 9 % between the first and the second calibrations while the ratio of IE to air beam (AB) signal increased by $\sim$ 20 %. This increase of instrument sensitivity occurred after a pump failure that led to a couple days of instrument down time. The IE/AB ratio remained stable for the rest of study (r.s.d. < 4 %, Fig. S1). "

p. 20757, line 13 and following: The authors describe why they used collection efficiency factor of CE = 0.5. This value seems also to be supported by the measurements of particle acidity. However, Fig. 3 shows clearly that for some periods during the campaign the relative humidity was above 90 %. As shown by Matthew et al. (2008), who investigated the dependence of the collection efficiency on particle humidity (and not particle acidity, as mentioned by the authors), a high liquid water content can also increase the collection efficiency of the AMS. Have the authors taken that into account, for example by drying the aerosol prior to sampling?
The relative humidity shown in Fig. 3 was for ambient air, not the relative humidity at the AMS inlet. As discussed in section 2.3.2, in this study, the RH at AMS inlet was lower because the AMS sampling line was not thermally insulated and the indoor temperature was typically 20-30°C higher than outside. We estimated that the RH at the AMS inlet was < 10 % most of the time. At such low RH, aerosol water will like to evaporate and particles entering the AMS will likely to be dry with low liquid water content. Thus, the CE = 0.5 used in this study is not inconsistent with the conclusion by Matthew et al. (2008).

p. 20757, line 18: "...by nephelometer and PSAP. "Please explain abbreviations!
"PSAP " stands for Particle Soot Absorption Photometer. The acronym is now defined

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in the revised manuscript.

p. 20761, line 15: "... meteorological input from FNL. "Please explain abbreviations!
FNL is one of the four different databases from Air Resources Laboratory (ARL). It refers to the GFS final analysis (FNL) for the northern and southern hemispheres.
We have clarified it in the revised manuscript "The back trajectory analysis was performed with meteorological input from Air Resources Laboratory FNL data archive. "

p. 20770, paragraph 3.4 - 3.5: This is actually the most interesting part of this paper, but it could be improved. A more detailed comparison between the different events would be very interesting. For example, does the elemental composition of the organic aerosol show changes for the different periods, and if so, how does it change. The trajectories shown in Fig. 9 suggest that for the DE1 and all OE events the air masses could be influenced by local, continental sources, while the DE2 air masses seem to be mostly influenced by the ocean.
We thank the referee for this suggestion. Detailed comparisons and discussions on the bulk aerosol composition, organic composition, and the potential sources and processes for different events have been added to this section.

p. 20773, line 28: "Sun et al. (2008) " Is this the publication cited before as Sun and Zhang (2008)? If so, please change accordingly. Otherwise, this publication is missing from the reference section
The reviewer is right. We meant to cite Sun and Zhang (2008), which is Sun and Zhang (2009) in the revised manuscript.

p. 20775, References: Please, give the full author list for each reference. This is actually mandatory according to the ACP/ACPD submission guidelines!

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We now give the full author list for all references in the revised manuscript.

Fig. 1: This figure is not needed, especially since DeCarlo et al. (2006) is referenced several times
Although a schematic of HR-ToF-AMS is already shown in DeCarlo et al. (2006), it is included in this paper for easy reference, especially since this manuscript focuses almost exclusively on an ambient study results from this instrument.

Fig. 2: In (b) and (c) the label for the 1:1 line is out of the picture.
A 1:1 line has been added on Fig. 2 (b) and (c).

Fig 6: Why was m/z 28 excluded on the right axis? Please explain.
The organic *m/z* 28 (mainly CO^+) is set to 0 in the standard AMS Data Analysis Software (SQUIRREL) because quantification of organic *m/z* 28 is a challenge due to the interferences of very large $N_2{}^+$ signal from gas phase N_2. Therefore the organic mass spectra reported in previous studies are generally normalized to total organic signal without *m/z* 28. However, some previous studies have observed that the organic *m/z* 28 is $\sim$ 0.9-1.3 of *m/z* 44 for ambient organic aerosol (OA) (Zhang et al., 2005;Takegawa et al., 2007). We therefore set *m/z* 28 = *m/z* 44 for OA in this study (see section 2.3.2). As a result we have two ways to normalize the organic mass spectra, i.e., with *m/z* 28 and without *m/z* 28, as we done in Fig. 6. Using the axis without *m/z* 28 is convenient for comparing the mass spectra observed in this study to those reported in previous studies.
We have clarified the meaning of axes in the captions of Fig. 6 and Fig. 15 as follows: "The left axes show the percent contribution of each peak to the total signal using the revised fragmentation table, i.e., with *m/z 28* (CO^+ = $CO_2{}^+$). The right axes indicate the percentage of each peak using the standard AMS fragmentation table (Allan et al., 2004), i.e., without *m/z 28* (CO^+ = 0). "

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Fig. 7: How do the authors define "good S/N "?
Fig. 8: Again, how is a "good S/N " defined?
A W-mode mass spectrum is defined as having "good S/N " when the ambient organic aerosol loading was above 0.7 μg m^{-3} for this study. 0.7 μg m^{-3} was decided based on visual inspection of ambient W-mode mass spectra as well as those of particle free air. At organic aerosol concentration < 0.7 μg m^{-3}, the W-mode spectra are too noisy to allow a good estimation of the elemental composition. This is explained in section 2.3.2.

Fig. 11: Axes are not labeled.
Axis labels have been added in Fig. 11.

Fig. 12: Part of the RH trace is outside the diagram.
RH readings beyond 100 % were not shown in Fig. 3 and Fig. 12, because these readings are wrong due to the sensor being iced up.

Response to referee 2
In this manuscript, the authors have investigated aerosol characterization by HR-ToF-AMS at the Whistler Mountain during INTEX-B campaign. Impact of the air masses to the aerosols chemical composition and size distribution are detailed. Authors presented general results and detailed 5 specific cases (OA1, OA2, OA3, DE1 and DE2) correlated to specific air masses.
This manuscript is very interesting and globally well written. The different sampling techniques, data analysis methods and parameterizations used are particularly well documented. This manuscript is suitable to be published in Atmospheric Chemistry and Physics after authors considered the following comments and remarks that should be addressed in the revision before the paper being published.
Specific comments:
-Page 20751, Abstract: Authors wrote ẗhe average size distributions of sulfate and

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ammonium both showed a large accumulation mode peaking around 500-600 nm in D_{va} ; in the conclusion S̈ulfate and ammonium appeared to be internally mixed with a large accumulation mode peaking around 400-500 nm änd in the text (section 3.2.2, page 20764, line 6) referred to 500 nm for sulfate and ammonium size distribution. Only one value must be used.
The peak value of D_{va} for sulfate and ammonium has been clarified to be $\sim$ 500 nm in abstract, text and conclusion.

-Page 20753, line 21 "Sun and Zhang, 2008 ": authors referred 7 times to this work but it is corresponding to a paper in preparation. Authors must give either a more precise reference that reader can refer to or detail a little bit more results of this works.
-Page 20754, line 9 "MacDonald et al. (2008) ": same comment.
As stated in our responses to the first reviewer, we have removed the reference to Macdonald et al. (2008) and instead cited Leaitch et al. (2008) and McKendry et al. (2008) for relevant information. We kept the reference to Sun and Zhang (2008), which is Sun and Zhang (2009) in the revised manuscript, since this paper provides information that supports some of the discussions made in this paper and it has been submitted for peer-review.

-Page 20759, line 3: Authors referred to a "good S/N " but did not clearly define it. How did the authors define it?
A W-mode mass spectrum is defined as having "good S/N " when the ambient organic aerosol loading was above 0.7 μg m^{-3} for this study. 0.7 μg m^{-3} was decided based on visual inspection of ambient W-mode mass spectra as well as those of particle free air. At organic aerosol concentration < 0.7 μg m^{-3}, the W-mode spectra are too noisy to allow a good estimation of the elemental composition. This is explained in section 2.3.2.

-Page 20767, line 1: The nominal formula for organic aerosol composition is a

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very interesting result and it is one of the most important results of this paper. However, more detail interpretations are expected. Indeed, Authors presented in this manuscript, different aerosol events (OA1, OA2, OA3 and DE1) corresponding to different air masses, we can attempt to also have a comparison of the nominal formula for each of these different events. For this reason, I strongly recommend to add this kind of comparisons in the revised manuscript.
We have taken the referee's suggestion and reported the nominal formulas of organic aerosol for different events in the revised manuscript. The compositional differences and associated sources and processes for different events are also compared and discussed in more detail.

Technical comments:
-Page 20752, line14: citation "Akimoto, 2003 " is not referred in References.
The reference of "Akimoto, 2003 " has been removed from the revised manuscript.

-Page 20754, line 3: why referred to section 3.1?
The reason of referring to section 3.1 was that the time series of ambient pressure is shown in Fig. 3, which is discussed in section 3.1. This tag has been removed from the revised manuscript to avoid confusions.

-Page 20754, line 23: citation "Decarlo et al., 2004 " is not referred in References.
We thank the referee for pointing out this oversight. We have added the reference "DeCarlo et. al. (2004) " in the reference section.

-Page 20757, line 18: definition of PSAP.
"PSAP" stands for Particle Soot Absorption Photometer. The acronym is now defined.

-Page 20761, line 15: definition of FNL.

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FNL is one of the four different databases from Air Resources Laboratory (ARL). It refers to the GFS final analysis (FNL) for the northern and southern hemispheres.
It is now clarified in the revised manuscript:
"The back trajectory analysis was performed with meteorological input from Air Resources Laboratory FNL data archive. "

-Page 20767, line14: "H_2O^+ = 0.25 CO_2^+ ", it should be 0.225 according to Aiken et al,(2008).
We thank the referee for pointing out this typo. It is now corrected.

-Page 20774, line 9: "m/z 48 (SO^+) and C_4^+) " the bracket after SO^+ must be deleted.
The extra ") " after SO^+ has been deleted in the revised manuscript.

-In Fig 2: reference to line 1:1 appears only in plot a. and d. and seems to be missing in the other. It should be better to add it in the legend box.
A 1:1 line has been added to Fig.2 (b) and (c).

-In Fig 4: should be "SO_4^{2-} ", not "SO_4^{2}- ".
Corrected in the revised manuscript

-In Fig 6 and 15: it is not so easy to understand the raison of different left and right axis. The authors should explain a little bit more how to read these graphs.
The meanings of the left and right axes are clarified in the figure captions in revised manuscript as follows:
"The left axes show the percent contribution of each peak to the total signal using the revised fragmentation table, i.e., with *m/z* 28 (CO^+ = CO_2^+). The right axes indicate the percentage of each peak using the standard AMS fragmentation (2004), i.e., without *m/z* 28 (CO^+ = 0). "

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-In Fig 7: it should be a good improvement to add the different periods of interest (i.e.OA1, OA2, OA3, DE1 and DE2) on the graph as done in Fig. 3.
We have marked the periods of interest on Fig. 7 in the revised manuscript.

-In Fig 11: names of the axes may be helpful for the reader.
Labels for the axes in Fig. 11 have been added in the revised manuscript.

-In Fig 13: definition of STP.
STP is spelled out "standard temperature and pressure " in the revised manuscript

-In Fig A2 middle panel: should by "NO_2^+ ", not "NO_2+ " in the left axis legend.
Corrected in the revised manuscript

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