

***Interactive comment on* “Fine particles from Independence Day fireworks events: chemical characterization and source apportionment” by Jie Zhang et al.**

Anonymous Referee #1

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In their manuscript “Fine particles from Independence Day fireworks events: chemical characterization and source apportionment” Zhang et al. present results from aerosol measurements during and around the Independence Day fireworks in Albany, NY. Aerosol composition and species-resolved size distributions were measured using a HR-ToF-AMS. In addition, several instruments were measuring particle size distribution, particle mass concentration and black carbon concentration. Finally, a LIDAR was applied to determine backscatter and wind information. The authors present an analysis of the chemical composition of the fireworks aerosol as well as that of the aerosol during other times, not affected by the fireworks as determined by AMS. This includes a PMF analysis of the AMS organics. This analysis suggests that there have been

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other, less intense, events which were affected by fireworks activities during the days preceding Independence Day. In addition, PM concentrations during and outside the fireworks event were compared and the relative and absolute contribution of the fireworks to the ambient aerosol are discussed. Finally, LIDAR measurements were used to investigate transport of aerosols from fireworks activities both from Albany and from the nearby Town of Bennington. The manuscript is well and clearly written. The analysis of the data makes the impression that it was performed thoroughly. However, when reading the manuscript I found a couple of inconsistencies, very different depth of information for different issues, or unclear presentation of results (see detailed comments below). Furthermore, I think the presented analysis is well behind what could be done with this interesting data set and thus neglects a significant fraction of its potential. So far the authors present a pure description of some observations during an individual event with limited general importance. In order to qualify for ACP and to provide results that can be transferred to other, similar situations, it would be desirable to extend the data analysis beyond the current state. Some examples are: • Comparison of the size distributions (total and species-resolved) measured during the Albany fireworks event, during the arrival of the Bennington fireworks aerosol and during the times not affected by fireworks could provide interesting information on the characteristics and temporal evolution of fireworks aerosols. • Comparison of the organics mass spectra of the fireworks aerosol during the Albany fireworks event and during arrival of the Bennington fireworks could provide information on aging of fireworks aerosol. For this purpose the fireworks contribution to the organics needs to be determined in a different way, e.g. by separate PMF analysis of both time intervals or by determining the difference between event and non-event times, instead of using a constant PMF factor. • A more general determination of the contribution of fireworks aerosol to the aerosol burden at a city like Albany, including an estimate on how large the contribution within the affected days or the full year is, in comparison to contributions from other sources would enable the reader to better judge the relevance of such events. This could also include an estimate of the indirect contribution of such an event due to enhanced traffic

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emissions around the event.

Detailed comments:

Several fireworks events were identified during the measurement period, based on the appearance of the potassium ion in the aerosol mass spectra. I think the authors should explain the criteria more specific here since the potassium ion is also observed with the AMS when measuring biomass burning aerosols. It would be interesting whether during the observation of fireworks events with the AMS also other ions, which could be from the fireworks, were observed, e.g. Sr, Na or other metal ions.

I have a general problem with the description of the measurement locations. According to Figure 1, the wind direction was approximately 180° - 270° during the main FW event. According to Figure S1 the ASRC site was located about NW of the fireworks site, i.e. it needs wind from approx. 100° - 130° to transport the fireworks aerosol to the measurement site. The ACHD site was located approx. to the SE of the fireworks site, i.e. it needs wind from approx. 280° - 320° to transport fireworks aerosol to the measurement site. While for the relatively close ACHD site general dispersion of the fireworks aerosol in the area would probably also transport aerosols to the measurement site, this seems unlikely for the ASRC site, which is almost in the opposite direction compared to the needed wind direction. How can you explain the transport of aerosol from Empire State Plaza to the ASCR site under these meteorological conditions?

I find the presentation of time series of aerosol concentrations (e.g. in Figs. 2, 4, and 6) stacked on top of each other not very helpful when one tries to see temporal variability of any of the mass concentrations. Generally, temporal variability is seen well only for the lowermost trace and in addition – for graphs which show fractional contributions like Fig. 2 – for the uppermost trace. For all other traces only the largest changes can be identified in this type of graph.

The presentation of all the aerosol data for a 10.5 days interval makes it hard to identify the detailed features during the main fireworks event which were discussed within the

text. It would be very helpful for the reader to have graphs which show only the time interval around the fireworks event. In addition, it would be very helpful if Figure 7 would show the same time interval as those figures and if the time axes would agree (i.e. not using UTC for one graph and local time for the others).

P2L4: Fireworks aerosol is mainly found in PM1, not in PM2.5, as shown in this manuscript as well as e.g. in the Drewnick et al., 2006 paper.

P2L11-15: Here consumption figures of fireworks in the US are given. It would be interesting to also have numbers for July 4 and may be for the Independence Day fireworks in Albany. The 42% increase of 24-hour PM2.5 – how much is this in absolute concentrations?

P2L21: Here two Sun et al. papers are cited. The Sun et al., 2010 paper is not in the reference list. Both papers do not seem to be adequate references for the general measurement capabilities of advanced aerosol mass spectrometry technology like the Jayne et al. and the DeCarlo et al. papers.

P3L4-7: Here it seems that some statements are exaggerating. Why was the meteorological situation such that it provided a “unique opportunity” for investigation of the contribution of FW displays to ambient aerosol concentrations? As stated above, the wind direction seemed to be even very unfavorable. Also I do not see how the “health influence” of FW-related aerosols was investigated in this study.

P3L16 and L22 and P4L15: It would be helpful if the authors would specify above what the elevation was measured. I guess in L16 it is a.s.l. and in L22 it is above ground, on P4L15 it could be both. Furthermore, if here and in the following paragraph some detailed information is provided, it would be nice if this would be done more consequently: Why is the sampling height provided for the ACHD site but not for the ASRC site? Where was the AMS instrument located, where the SMPS and the pDR-1500? Why is the sampling line length for the inlet duct at ASRC (?) provided, but not the length of the sampling line from the duct to the instruments and also not for the inlet

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at the ACHD site? For estimate of inlet line losses, also the diameters of the lines and their orientation would be needed.

P3L19: According to the statement here, the gap in the measurement data was due to maintenance of the instruments. To me it seems very strange that maintenance started at midnight, that maintenance lasted for 44 hours, and that all instruments have exactly the same downtime.

P5L5 and the whole paragraph: “. . . slow evaporation and ionization of potassium . . .”. “evaporation” should be “vaporization”. While potassium might vaporize slowly, ionization is probably not slow. In addition to this effect, the quantification of potassium in the AMS is also difficult due to the fact that potassium can be surface-ionized at the vaporizer surface during the vaporization process. This effect will directly affect RIE of potassium. Since this effect – and as a consequence the RIE of potassium – strongly depends on the vaporizer temperature, the history of the vaporizer and the tuning of the AMS, it makes little sense to use RIE values for potassium obtained in measurements with other AMS but it would be better to determine the own RIE value.

P6L2: The statement that “all instruments exhibited the same general behavior” seems a bit exaggerated. There are sometimes strong differences between the various PM2.5 measurements up to more than a factor of two. There are also sometimes strong differences in the trends.

P6L3-5: I also do not agree with the explanation of the increasing trend in aerosol concentrations with hygroscopic growth. An increasing trend is observed from 6/27 until 6/29 according to Figure 1. During this time RH values varied strongly and no correspondence of the increase in PM and RH can be observed for this whole time interval. Furthermore, hygroscopic growth of particles should not affect AMS measurements of mass concentrations and also not measurements performed with a dryer before the instrument.

P6L8-11: The aerosol concentration peaks during the nights of July 2-3 were also

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identified as FW-related due to their high K signal. Is there any other evidence that these peaks are actually from FW – e.g. similar K/S ratio as observed during the July 4 peak? Was there fireworks reported or observed in the area around the measurement sites? According to the magnitude of these additional FW peaks either a massive fireworks event in the order of 30% of the July 4 fireworks or fireworks which were burned very close to the AMS site would be needed to generate these peaks. Is this realistic?

P7L3-5: It is very hard to assign all these numbers to the respective species.

P7L8-10: If SO₄ and NO₃ are from K-rich salts and if slow vaporization of K results in under-estimation of K, this should also result in under-estimation of the related SO₄ and NO₃.

P7L10-11: I do not agree with the statement here. According to Figure 2, organics and K show broader peaks than SO₄. If all three are from the same source (fireworks), is this difference a sign of chemical processes occurring with the FW aerosol?

P7L13-15: If SO₄ fraction varies between 8.5 and 16.4% (a factor of two), I would not call this “fairly stable”. As a consequence I would not say that this indicates constant sources in and near Albany as stated in the following sentence.

P7L19-21: If there was a large difference in the mass fractions of the aerosol components before, during, and after the FW event, this would be interesting to see in pie charts, e.g. in the supplement.

P7L22: If the FW event lasted from 23:00 until 02:00 (line 1, same page), this is 3 hours, not 4 hours.

P7L24: What is “volume mobility diameter”?

P7L27: Not the surface ionization but the slow vaporization of K-containing species will make the K size distributions complicated.

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P8L11: Can the authors provide any information on the potential sources of the evening BBOA?

P8L19: It should be Fig. S3b, not S3d.

P8L24 and other locations: Percentages given with a precision of 0.1% and at the same time with likely uncertainties of 10% or more do not make sense.

P8L6-P9L7: This section with the discussion of the PMF results is not very satisfying. The discussion on the background aerosol sources seems very incomplete and short and does only provide a rudimental overview over the various aerosol sources and their variations. Also the discussion on FW effects on the organic aerosol seems rather incomplete. It would be interesting to have a discussion on how fireworks activities like the Independence Day fireworks quantitatively affect the concentrations of the various types of organic aerosol: HOA – by more people being on the streets to observe the display; BBOA – as a by-product of fireworks burning; FW-OA – as a direct emission of the fireworks; OOA – as a potential product of FW-related gas phase emissions. Especially in order to make this study relevant to a broader audience and for FW events outside of Albany, NY this could be very helpful.

P9L13-15: I have the impression that the calculation of organopotassium is highly uncertain. Due to uncertain RIE, slow vaporization effects and surface ionization of potassium the absolute K concentration already is uncertain. The calculation of inorganic potassium is probably even more uncertain due to additional uncertainties of the ammonium fraction of the various salts. Finally, the difference of the two is even more uncertain. If this approach is used, an estimate of these uncertainties would be needed. The fact that resulting organopotassium correlates with FW-OOA is not surprising since potassium correlates well with FW-OOA and organopotassium is probably only a relatively small fraction of total potassium.

P9L15-18: It is unclear to me what the purpose of the calculation of the density of the aerosol is. In this calculation the contribution of refractory aerosol components with

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likely high density (metal salts or oxides) and of black carbon is neglected. With these additional uncertainty the resulting uncertainty of the density calculation becomes so large that also a typical density for urban aerosol could be assumed.

P10L4: I do not agree with the assumption that the ratio of FW-related aerosol and background aerosol does not change during transport: While the FW-related aerosol gets diluted during transport, this is not the case for the background aerosol.

P10L6: In addition to the 2-hour average also the 24-hour average of the aerosol should be mentioned here, if these values are compared to the 24-hour threshold value.

P10L7-10: Here the temporal trends of various aerosol components measured during the time after the FW display are used as evidence for conversion processes within the atmosphere. How can the authors be sure that during this time always air masses were probed, which encountered fireworks activities during the night of July 4 and not simply different air masses with different influence of fireworks were probed?

P10L12: Define the “carrier-to-noise ratio” and explain what kind of information this ratio provides.

P10L14: The local time interval is 3 hours long, the UTC interval only 2 hours.

P10L12 – P11L6: This whole section is very confusing to me. There are many inconsistencies between the observations and their interpretation. E.g.: According to the LIDAR weak easterly winds were measured. This is contrary to the wind directions provided in Figure 1. What is correct? Many of the features that are explained in the text are not really visible in Figure 7. What means “high altitude FW transport” – at which altitude was the fireworks burned? There was “high altitude” and “low altitude” aerosol observed in the LIDAR. It is claimed that the low altitude aerosol was the result of subsidence or diffusion of the high altitude aerosol. However, there is no connection visible in the two clusters in the LIDAR signal. Since all the aerosol data are provided for 10 days and the LIDAR signal are only for a couple of hours it is almost impossible

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to see all these short-time features in the aerosol data. The 500 m back trajectory passes over the town of Bennington. However, according to Fig. S8 the affected air mass was at an altitude of 1000 m over Bennington. It is unrealistic that this air contained aerosol from the local fireworks in Bennington. It is also unrealistic that the 500 m altitude air was sampled at ground. The aerosol arriving from Bennington (after 40 km of transport) produces a more intense signal in the LIDAR measurement than the aerosol from the much closer Albany fireworks. Is this realistic? This whole section seems to be highly speculative. If it is not it should be much clearer connected to the LIDAR measurements.

P11L8-9: What does “Chemical characterization and source apportionment ... are quantified ..” mean?

P11L21-25: This information is not part of the presented study.

Figure 3: Why where the AMS size distributions measured over such a long time interval. This does not allow distinguishing between size distributions dominated by FW events and those of the background aerosol.

Figure 7: This Figure is rather useless for the reader. The features presented in this figure are not explained. The scale has no units and it is not explained what it shows. The wind markers are not visible. It would be nice to present time on this figure consistently with the other graphs.

Figure S3: Diurnal pattern of an individual event (FW-OOA) does not make a lot of sense.

Supplement page 4: The strong dips in SV-OOA and LV-OOA during the main fireworks event and the peak in the residual signal during this time show that PMF does not clearly separate the various types of organic aerosols. This is often observed when individual events occur only during short fractions of the whole time of measurements. A possible approach to improve the PMF separation could be to apply PMF separately

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only to a short time interval around the main FW event.

Figure S4a: This figure shows that BC provides a substantial contribution to the overall aerosol. Therefore it cannot be neglected neither in the comparison of total AMS concentrations with those of other instruments nor in the calculation of aerosol density.

Figure S5: After the main FW event some events with very small particles occur. Can these events be explained?

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