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Interactive comment on "A new aerosol collector for on-line analysis of particulate organic matter: the Aerosol Collection Module (ACM)" by T. Hohaus et al.

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Reply to the comments of Anonymous Referee #2

The authors would like to thank the referee for her/his careful reading and positive feedback/helpful remarks that helped to improve the quality of the manuscript. In the following we respond to the individual comments and describe their realization. We have repeated the comments here in italics and added comment numbers for easy reference between points in the responses. Our replies follow each excerpt. Changes

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to the manuscript text are presented in bold.

General:

1. The title of the manuscript contains the phrase: "on-line" which could be an issue for intensive discussion considering that the sampling of aerosol and the analysis happen in separate steps. The authors re-qualify the measurement to be "quasi-online" in the abstract (P 1362, L 9). For the method described here it would be also an overstatement to call it "off-line" method (as filter or impactor samples usually are labeled with). However, for my understanding, the expression "on-line" is related to a continuity of sampling and measurement and does not fit to the working principle of the ACM-GC-MS.

[Response]: In the authors opinion the term "quasi on-line" describes the working principle of the ACM GC-MS most accurately. Therefore the description of the working principle was changed in the manuscript accordingly and the title of the manuscript now includes quasi on-line.

2. The introduction section could work out in more detail: what are the current "state-ofthe-art" techniques for OA specification, their advantages/disadvantages – making the development of new techniques such as the ACM-GC-MS so essential.

[Response]: The most common techniques for OA measurements (both off-line and on-line) are discussed in the introduction of the manuscript already and the authors refer to a number of recent review articles dealing with aspects of OA measurements (Canagaratna et al., 2007; Hallquist et al., 2009; McMurry, 2000; Turpin et al., 2000; Rudich et al., 2007). A more detailed discussion of OA measurement techniques and emerging issues is given in these reviews and beyond the scope of this manuscript.

2b. The collection surface is regularly cooled to -30° C and heated up to 270° C which C1033

might cause some material stress. Additionally there might be aerosol components that simply remain on the surface because they are not vaporizable even at cleaning mode temperatures (which are not further specified in the manuscript). Is this surface regularly cleaned or replaced from time to time? Furthermore this non-vaporizable inorganic substances may adhere onto the collection surface, and although this surface is chemically passivated this accumulating inorganic substances could cause interaction with the fresh aerosol sample – or does this Silcosteel work like a Teflon coating in a pan and the backflush cleaning mode is able to remove also the inorganic from the surface? If yes this should be clearly stated.

[Response]: We agree that only species that are volatile at the applied vaporizer temperature will evaporate in the ACM. We refer to the response to referee 1 item 2 for a detailed discussion of this aspect. With respect to the possible accumulation of e.g. inorganic substances on the collection surface no indication of interferences by such effects was found in the field measurements. In order to test whether the amount of inorganic material sampled in previous measurement intervals would affect the recovery rate of GC-MS detectable organic species, the ratio of ACM-GC-MS observed signal to AMS organic mass was investigated as function of accumulated AMS inorganic aerosol mass. No dependence on the total inorganic mass was found.

3. Concerning the procedure described in section 4.2: Comparing measurements by simultaneously varying two parameters (here the vaporizer temperature of the ACM-GC-MS and the aerosol loading into SMPS and ACM-GC-MS) has the risk that two effects could interfere with each other. It might have been tested previously that this is no issue for this particular case but there should be a comment on that in the text.

[Response]: As can be seen in figure 4 the data obtained at a vaporizer temperature of 225°C cover the full mass range explored here. The fit results are the same independent of whether the data points at other temperatures are included or not. Therefore we think it is appropriate in this case to conclude from the data shown both, that the

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octadecane mass measured with the ACM-GC-FID is equal to the mass provided and independent of the vaporizer temperature in the range 150 to 270 °C.

4. What is really missing in section 5.4 is a correlation between the mass spectra (MS) of both instruments while the AMS from Aerodyne (not mentioned which type) definitely is capable to provide mass spectra of OA - dependent on the AMS type in lower (C-ToF) or higher (HR-ToF) resolution. Furthermore, particularly this instrumental comparison appears to a reader to be halffinished.

[Response]: Since the mass spectra obtained by the two instruments are from total organics (Q-AMS) and individual organic compounds (ACM-GC-MS), respectively, a direct comparison of the mass spectra obtained will not yield additional information. We therefore think that in terms of characterizing the ACM-GC-MS instrumental performance the given comparisons are the most instructive.

5. Not clear to me is why here arbitrary dimensions are given for the ACM-GC-MS while for the filter comparison the measured mass was possible to be calculated. Thus the correlation unfortunately doesn't allow for further conclusions than the linearity of two measurements with a coefficient of 0.97 but a comparison between resulting MS could probably provide more indication of the ACM-GC-MS performance. Thus the conclusion section 6 should be rephrased significantly or other data should be shown to confirm those conclusions. At current state the capability of ACM-GCMS to perform "quantitative measurements of ambient aerosol mass loading" is not shown.

[Response]: We agree that the ACM GC-MS data only represent a part of the organic aerosol compositon. In order to clarify that the ACM-GC-MS measures single organic species in the aerosol, the conclusion in section 6 was changed to: The correlation between the two sets of data is linear with a high correlation coefficient, indicating that the overall ACM-GC-MS measurements are representative of the ambient organic aerosol mass loading.

Specific:

7. -P1362, L 5: "However, current analytical methods are far from full speciation of organic aerosols and often require long sampling times." This is a very general statement and could be related to the sampling times the ACM needs.

[Response]: The statement was changed to now read: However, current analytical methods are far from full speciation of organic aerosols and often require sampling times of up to one week.

8. -P1362, L 11–12: "...and transfer gasified atmospheric aerosol particles." The word "particle" should be replaced by "components". After sampling and vaporization it is not an aerosol particle anymore which is analyzed but its chemical composition.

[Response]: The description was change to clarify that with the ACM the volatile and semi-volatile constituents of the particles are desorbed: After collection is completed volatile and semi-volatile compounds are evaporated from the collection surface through heating and transferred to a detector.

9. -P1362, L 15: Here, after the sentence is finished, it could be stated very concretely what are the shortest (or most reasonable) sampling times of the ACM. What is the temporal resolution that can be reached with the ACM-GC-MS?

[Response]: The sampling time for the ACM-GC-MS depends always (i) on the aerosol mass concentration present in the respective experiment (chamber aerosol concentration, ambient aerosol concentration), (ii) on the abundances of the volatile and semi-volatile compounds in the respective aerosols which are in the focus of the experiments conducted and (iii) on the sensitivity of the gas phase detector coupled to the ACM. Therefore in the authors opinion giving a number for the shortest sampling time could be misleading and misinterpreted as the overall shortest sampling time the ACM is capable of.The sampling times applied in the chamber and field experiment presented

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here are stated in the respective sections. For the chamber experiment this sampling time was chosen to match the GC measurement interval and thus optimize the duty cycle.

10. -P1363, L 19: "such as loss of compounds due to volatilization" as this statement is kept very general this is an issue for the ACM-GC-MS as well (particularly in the aerodynamic lens), isn't it?

[Response]: To clarify the statement the sentence was changed to read: Off-line time integrating bulk measurements are often subjected to positive and negative storage artifacts, such as loss of compounds due to volatilization, gas phase adsorption and reactions during collection that can alter the sample (Turpin et al., 2000).

11. -P1363, L 20-22: "uncertainties exist as to how representative these samples are for the atmospheric aerosol composition" this is valid for any instrument.

[Response]: the sentence was removed

12. -P1363, L 28 –P1364, L 1: "Therefore today an important focus lies on the on-line measurements of aerosol chemical composition in real time with high time resolution." Very general statement containing further "eye catcher" superlatives of which no one necessarily fits to the ACM-GC-MS as well.

[Response]: In the authors opinion these statements are qualified for the online methods mentioned (AMS and PILS). Since the ACM-GC-MS is referred to as quasi on-line through out the manuscript no overstatement of its potential is seen.

13. -P1364, L 11: "...cooled collection surface." The readers would like to know concrete values – or at least a statement in which temperature range this cooling happens.

[Response]: The temperatures applied to the ACM collector (for cooling and heating) can be set within a range from zero to a maximum/minimum temperature. Since this temperature should be chosen for each individual experiment the concrete numbers are stated with each experiment described in this manuscript and not in the general description of the ACM. In the instrumental description part of the manuscript the minimum and maximum temperatures for cooling and heating of the collector are added to point out which temperature range is possible.

14. -*P1364*, *L* 12: "...heated the collection surface." The same as mentioned before. [Response]: It is referred to item 13 for the response of the authors.

15. -P1364, L 12: "...transferred by a carrier gas..." which one? This gap would be less obvious by leaving out "by a carrier gas" if the authors want to keep this for the detailed instrumental description part.

[Response]: That the type of carrier gas is not specified in this part of the manuscript was on purpose because the ACM is not bound to use a certain type of carrier gas. Every (preferable inert) gas can function as a carrier gas. The type of carrier gas is typically determined by the coupled gas phase detector. For example He, N₂ and H₂ (typical GC carrier gases) and also purified air are possible. The sentence was change to indicate that more than one carrier gas can be chosen for the ACM depending on gas phase detector coupled to the ACM: **The ACM can be operated with a variety of carrier gases (e.g. He, H**₂, N₂) often determined by the type of gas phase detector coupled to the ACM.

16. -P1365, L 6: the aperture dimensions, as well as all further, should be given in SI dimensions.

[Response]: Done.

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17. -P1365, L 9: please state, at least in brackets, which volume flow rate (value) is ensured.

[Response]: The values for the volume flow rate was added to the manuscript.

18. -P1365, L 12: There should be stated why it is necessary for this system that the vacuum system can be isolated.

[Response]: The sentence was extended as follows: The vacuum system can be separated from the collector with an automated controlled vacuum isolation valve to prevent carrier gas from compromising the vacuum and to prevent loss of volatile and semi-volatile compounds during desorption and transfer of the compounds to the detector.

19. -P1365, L 15: Is the kind of stainless steel ("316") necessary to know for understanding the working principle?

[Response]: The description of the stainless steel type was deleted.

20. -P1365, L 21: The new information is that the cooling of the collector surface happens with liquid nitrogen. Still open: to which temperature.

[Response]: It is referred to item 13 for the response of the authors.

21. -P1365, L 21: "The cold nitrogen is..." - from "liquid nitrogen" in the sentence before the word "cold" becomes redundant.

[Response]: The word "cold" was deleted.

22. -P1365, L 24: Here the accuracy is given to hold the cooling temperature – still the reader doesn't know the value of "set temperature".

[Response]: It is referred to item 13 for the response of the authors.

23. -P1366, L 2: please replace, here and elsewhere, the imperial by SI dimensions.

[Response]: The imperial dimensions where replaced with SI dimensions with except of the dimensions for the tubings where tubings with typical imperial dimensions were used for the set up of the ACM GC-MS or the experiments. In the authors opinion it is more instructive to use common and known descriptions of e.g. "1/16" stainless steel tubing" than "stainless steel tubing with an outer diameter of 1.5875 mm".

24. -P1367, L 5: "The cartridge heater is heated..." to which temperature and with which heating rate ($^{\circ}C s$ -1)?

[Response]: The cartridge heater can only be switched on or switched off. When voltage is applied to the cartridge heater it heats up without a controlled heating rate to the preset temperature. The maximum temperature was added in the instrumental part of the manuscript.

25. -P1367, L 10: "...a general rule is set in this stage to a higher temperature than the desorption..." how much higher?

[Response]: The backflush temperature has to be chosen in an iterative manner. If after cleaning at the higher backflush temperature residues of the volatile and semivolatile compounds can be detected in a subsequent blank measurement the backflush temperature has to be increased. For all experiments presented in that paper a blackflush temperature of 30 °C higher than the desorption temperature was sufficient to obtain a blank measurement showing no residues. During the backflush mode no additional measruements are done which can be used for checking the state of the possible residuals on the collection surface.

26. -P1367, L 15: "This cleaning of the collector is performed after each measure-C1040

ment. The duration of the backflush mode can be set to meet the actual requirements regarding possible remaining contaminations." Is the state of cleaning checked with measurements that are part of the cleaning mode or how else is the duration for the cleaning mode set?

[Response]: For the authors response it is referred to the previous item 25.

-P1368, L 3: "The coupling of the GC to the transfer line of the ACM was accomplished using a 1/16" Swagelok connector." Is this essential to know for understanding? If not it could be taken out.

[Response]: The sentence was deleted.

27. -P1369, L 9: Here the first time a concrete temperature is mentioned for the collection surface, which seems to be quite late. Anyways the question comes up: why that low temperature? Why not close to zero or at room temperature? Does a temperature of-30°C not cause further problems (condensation of gas/vapor that still may be present in the aerosol sample, though in low quantities)?

[Response]: For the discussion why a subzero temperature is chosen as a collection temperature and changes to the manuscript it is referred to the authors response to the comments of Anonymous Referee #5 for item 1. From the AMS instrument which has the same inlet system and therefore will transfer the same amount of gaseous species into the collection region of the instrument, it is known that only gases with mixing ratios in the ppm range are observed. This means that possible interferences from gaseous species are restricted to N2, O2, CO2, Ar and H2O of which only H2O may condense at the temperatures chosen for sample collection. But choosing a higher temperature e.g. room temperature evaporation of volatile and semi-volatile compounds from the particle phase under the high vacuum conditions during sampling would become more likely. This would introduced higher uncertainties to the results obtained with the ACM.

28. -P1369, L 13 – 20: In this section the reader gets the first time an impression about the time scales of a sample/analysis cycle. What is not really clear herein and in previous text: It should be stated explicitly if simultaneously to the analysis part with the GC/MS (or else) a new sampling cycle is started or if the analysis has to be completed before a new sample is initialized.

[Response]: Indeed during the GC-MS analysis of the evaporated sample a new collection can be started optimizing the ACM-GC-MS duty cycle. In order to clarify this the following sentence was added at line 20: Note that during the GC-MS analysis a new sample collection can be started.

29. -P1370, L 2 – 5: "The obtained mass spectra for each compound were compared with the EI spectra of the NIST library database. The comparisons achieve good results and the automatic search and compare algorithm of the MS data acquisition software (Masslab 1.4) identified all compounds with a high probability." For this comparison with results from a MS spectra database the quality of agreement should be demonstrated with values. Just with general comments like "good results" or "identified with high probability" the reader is not able to evaluate this.

[Response]: We added the minimum Reverse Fit Factor (REV) given by Masslab and stated also the value for a perfect agreement of the predicted and measured mass spectra. The sentence reads now: The comparisons achieve good results and the automatic search and compare algorithm of the MS data acquisition software (Masslab 1.4) identified all compounds with a reverse fit factor REV > 886. REV indicates how likely it is that the obtained spectrum contains the library entry with REV=1000 being a perfect match.

30. -P1370, L 20: the "conversion factor" could be specifies in more detail, please.

[Response]: The description of the conversion factor was extended to state the origin of this factor in the equation. The manuscript was changed to: The factor 10^{-21} accounts C1042

for the necessary conversion of the particle volume concentration and density to calculate the ACM collector mass loading in g.

31. -P1370, L 20: "Note that no impurities or artifacts could be observed..." The phrase could be clearer in terms of possible impurities or artifacts which were present in quantities smaller than the detection limit of the instrument – if possible proven by a figure.

[Response]: The sentence refers to possible impurities that might be present in the particles from contaminations of the solution or aerosol generation set-up. To clarify the statement the sentence was changed to read: Note that within the limits of detection no impurities could be observed in the GC/MS-FID chromatogram for all octadecane particle measurements.

32. -P1370, L 25 – 26: "The linear fit to the data shows that the slope is one." In fact it is not "one". Rephrase this, please, into e.g. «slope is with 1.06 +/- 0.04 very close to one» which would be the most honest.

[Response]: The sentence was changed to read: The slope of the linear fit to the data is 1.06 \pm 0.04 .The recovery rate for octadecane is 100% within the measurement uncertainties.

33. -P1370, L 26 – P1371, L 2: "Possible losses of octadecane at all temperatures over the complete pathway of the ACM system were found to be minimal." What, in numbers, means "minimal" and what is this statement based on? Is this measured somehow? If yes it should be presented in this manuscript.

[Response]: Since this another way of putting the result of 100% recovery we omit the sentence for clarity.

34. -P1372, L 11 – 12: "The experiment was conducted at ambient pressure and C1043

temperature." And a few lines later (P1372, L 16 – 17) it is mention "The initial conditions inside the chamber after flushing were 27% relative humidity, a temperature of 20 °C...." but a few lines prior (P1372, L 4 – 6) "This is provided by a floor heating system which temperature periodically changed in the range of 30° C±4°C over the duration of 2 h." So either this heating feature of the chamber was not used and this sentence (concerning the chamber) could be taken out or this feature was used and the measurements did not happen at ambient temperatures (considering also that the relative humidity is affected significantly with temperature).

[Response]: The floor heating was used during the experiment to have the chamber content continuously mixed by convection. However it is also stated in the manuscript that the effect of the floor heating results in a periodic temperature fluctuation in the chamber by about \pm 0.25 °C. The changes in relative humidity in the chamber were during the experiment \pm 2%. Therefore the temperature in the chamber was throughout the experiment at ambient conditions. The sentence in the manuscript was changed to clearly state the effect of the floor heating system: The use of the floor heating results in a periodic temperature and relative humidity fluctuation in the chamber during the experiment by about \pm 0.25 °C and \pm 2%, respectively.

35. -P1372, L 13 – 14: "The air flow through the chamber during flushing was $25m^3h-1$." Is this essential to know for better understanding?

[Response]: The sentence was deleted from the manuscript.

36. -P1373, L 1: "Note that the initial monoterpene and ozone concentrations are orders of magnitude above ambient levels..." Here it would be nice to have a concrete number of how many orders of magnitude.

[Response]: We added the scale to the sentence and it reads now as follows: **Note that the initial monoterpene and ozone concentrations were two to three orders of**

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magnitude above ambient levels (Guenther et al., 1995; Kesselmeier and Staudt, 1999).

37. -P1373, L 5 –6: "Hence the observed yields for products formed in this experiment not necessarily represent ambient yields." Either this is a contraposition to the cited statement of Atkinson (1997) then it should be much clearer. Otherwise it could be taken out as any reader can be assumed to be aware of this.

[Response]: The sentence was deleted from the manuscript.

38. -P1373, L 14 –15: The CPC "cut-off" is a commonly used but not very proper laboratory-slang expression. Better is, and one can read that increasingly, the "50 % detection particle diameter" of a CPC.

[Response]: Changed.

39. -P1373, 18 – 19: "The SMPS was measuring the particle diameters between 14 nm and 750 nm." The SMPS usually measures the number concentration of particles of a selected particle (electromobility) diameter (with an uncertainty, the DMA-specific band width).

[Response]: For clarity we changed the sentence to read: **The SMPS was measuring the number concentration of particles with diameters between 14 nm and 750 nm.**

40. -P1374, L 6: "The filters were preheated for 10 h at 600° C before sampling" If there is a reason for that it should be stated.

[Response]: The preheating of filters prior to sampling is a standard procedure used to remove possible contaminants from the filters. The used times and temperatures

are instructive in the sense that they give indications of which volatility fractions of contaminants were removed by this procedure.

41. -P1374, L 7 –8: "The sampling line of the filters was situated 3m away from the ACM-GC-MS sampling line." Does this mean that the aerosol taps were placed in the chamber with 3 meters distance to each other?

[Response]: Yes, the ACM GC-MS and the SMPS had a separated sampling line than the filters and these were 3 meters apart, also in the chamber.

42. -P1374, L 15 –21: "ACM-GC-MS blank measurements were conducted regularly between the chamber samples. For the blank measurement a High Efficiency Particulate Airfilter (HEPA) was installed between the chamber sampling tubing and the ACM-GC-MS inlet. The same measurement cycle of the ACM-GC-MS was repeated with the installed filter. The blank measurement were used to check for contaminations or residues inside the ACM-GC-MS and for adsorptions from the gas phase onto the collector." Unfortunately a figure showing those blank measurements comes later in the manuscript. You could place "c.f. section 5.6" somewhere in this paragraph. Do those blank measurements show a dependency on the duration of the cleaning mode? Is the cleaning state of the ACM measurable in this way?

[Response]: In the experiment presented in this manuscript the blank measurements showed no residues after the ACM was for 10 minutes in backflush mode. We didn't change this duration during the SOA and ambient experiments to prevent that possible residue interfere with our aerosol measurements. The influence of the duration of the backflush mode with a subsequent blank measurement was investigated during the octadecane calibration measurement. However as the results from the characterization with octadecane suggest even for small durations no residues were detected. But in general if volatile or semi-volatile compounds are not completely desorbed from the collection surface this type of measurement can be used to measure the cleaning

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state of the ACM. We added a reference as suggest in the manuscript for the blank measurement.

43. -P1374, L 19 –20: either "...the blank measurements were..." or "...the blank measurement was...."

[Response]: Corrected.

44. -P1375, L 1 –7: "The first approach..." is the beginning of one sentence. What is missing is the clear specification of the "second approach".

[Response]: The sentence was completed adding the information for the second approach and reads now: For the identification of the chromatographically separated compounds two approaches were used. The first approach was to identify the peaks by comparison of the mass spectra measured with the MS with the content of the NIST database and the second approach was calculating linear retention indices and comparing the result with literature values.

45. -P1376, L 4 –5: Either "index" is replaced by "indices" or "correspond" is replaced by "corresponds".

[Response]: Corrected.

46. -P1376, L 25: The molecular weight dimension "Da" (Dalton) is not a SI dimension [Response]: Canged.

47. -P1378, L 4: Either "3mm circular pieces were...." or "a 3mm circular piece was...." [Response]: Corrected. 48. -P1378, L 9: "A ND correction owing to the inhomogeneity of particle loading across the filter was done accounting for the overestimated nopinone concentration on the cut out filter piece." This suggests that the correction is particularly related to the nopinene. But the particle loading on a filter generally should be corrected for independent on the aerosol species. Thus the sentence should start with "A correction owing to the inhomogeneity...". Furthermore, for all of the mentioned corrections the correction factors could be stated in a way that the correction process is reproducible for a reader.

[Response]: The sentence was corrected as suggested. Also for all correction (BF, PVD and ND) described in section 5.5 the range of the correction were added to the manuscript.

49. -P1378, L 14: "The nopinone concentration drop continuously..." it should be "drops" instead of "drop".

[Response]: Corrected.

50. -P1378, L 25: That Billerica is located in Massachusetts is perhaps less important than the country "USA" which could be stated here.

[Response]: "USA" was added to the sentence.

51. -P1379, L 7: "…which in indicative of…" The "in" should be replaced by "is" [Response]: Done.

[nesponse]. Done

Figures Tables

52. - Table 2: the dimension of M-SOA is given in μ m⁻³ which is not a mass dimension as it should be according to the caption.

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[Response]: The dimension was corrected to μ g m⁻³.

53. - Table 3: The RIs have no dimensions.

[Response]: Table 3 doesn't list any retention indecies (RI) and RI are dimensionless.

54. - Neither in Fig. 4 nor in the caption or in the text it is mentioned if the error bars come from repeated measurements (if yes how many) or if they result from the instrumental errors of the SMPS and the ACM-GC-MS. In fact the bars seem to display a constant percentage of sampled mass. The question is also if instrumental uncertainties like SMPS band width are considered? Or the particle losses inside the SMPS. Furthermore the Fig4 caption talks about "…a line through the origin…". In the same figure the fit intercept proofs the regression not to be a line through the origin.

[Response]: The error bars in figure 4 originate from instrumental errors and take into account the instrumental uncertainties. The figure caption was changed to read: Octadecane mass measured with the GC-FID vs. the collector loading (calculated from SMPS measurements) together with a linear fit through data points. Numbers show the desorption temperatures for each measurement.

55. - Figure 5: please add dimensions to the ordinate axis label.

[Response]: Done.

56. - Figure 7: Instead of showing another time series of the ACM-GC-MS comparison with the filter samples (as shown already in Fig 6) the authors could think about showing the comparison of measured total mass with both methods fitted by a linear regression.

[Response]: As also suggested by anonymous referee 1 we changed Fig. 7 to a correlation plot and also updated the discussion in the manuscript. For the changed

Figure and the discussion we refer to the authors replies to Anonymous Referee #1 item 28 and 29

57. - Figure 8: It should be explained somewhere how to understand the increasing background for retention time < 33.8 min and constant values for retention time > 34min.

[Response]: The rise in the background is due to GC column bleeding. For details it is referred to to item 24 of the authors response to the comments of Anonymous Referee #1.

58. - Figure 9: The error of the total organic mass measured with the AMS could be easily implied as error bars into the graphic.

[Response]: We applied the error bars for the total organic mass measrued with the AMS. The errors are dominated by the uncertainty of the relativ ionazation efficiency of organic aerosol.

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