Anonymous Referee #2

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We would like to thank the referee for the constructive comments and suggestions made to improve the manuscript. Our responses are given below.

The paper of Poulain et al. attempts a chemical mass balance of ambient particles by utilising a thermodenuder and various measurements to make a mass closure with the aim of characterising volatile (or refractory) fraction of ambient organic matter. The paper presents an ambitious attempt, but I am not sure if authors have realised all the ambiguities and uncertainties to make their work useful and inspiring for other researchers.

The paper needs to be re-evaluated after major revision before considering it acceptable for publication.

The major problem starts with the title. Not only it brings uncertainty to the term “refractory”, but more importantly, how relevant is the use of thermodenuder in gaining insights about refractory nature of ambient organic matter. Refractory term should normally be used in circumstances when there is no ambiguity in the thermal decomposition. For example, AMS refractory mass is defined as flash evaporation of particles at 600C: temperature is arbitrary, but at least flash evaporation is instantaneous. The denuder based study is arbitrary both in temperature and the process of thermal breakup. To me the title should rather be about thermal stability or volatility of ambient particles and not the refractory nature at arbitrary chosen temperature.

Response: We agree with the comment that generally speaking aerosol volatility depends on the vaporization temperature and residence time inside the thermodenuder (TD). However, this is a general comment attributed to all the TD and aerosol volatility works not specific to this study. This is one of the most important reasons why such measurements are usually difficult to be inter-compared. Therefore, this issue was precisely taken care of by mentioning explicitly the working temperature of the thermodenuder on the title of the manuscript.

In order to avoid any confusion, the title of the manuscript will be changed to “Chemical mass balance of 300°C non-volatile particles at the tropospheric research site Melpitz, Germany”.

Authors seem to suggest that T=300C is some sort of magic number everyone should follow. If not then how the results would compare if somebody runs the denuder at 250C, or a twice longer denuder. There is no information about the stability of the result if residence time was e.g. 10 times longer. For example, HTDMA community are in constant discussion and intercomparison of particle equilibration time in a humidifier (3, 6, 9 or even 30s) while the denuder with 1s residence time is assumed to be sufficient for thermally decomposing internally mixed aerosol. There is clearly a need running three denuders in sequence to see if volatile fraction changes as a result – otherwise what the current study tells us?

Response: It was absolutely not our purpose to consider the 300°C of the thermodenuder as a “magic temperature”, which was selected with some arbitrariness. The temperature of 300°C was chosen earlier in the development of this thermodenuder setup (Wehner et al., 2002; Wehner et al., 2004; Philippin et al., 2004) and was considered on the one hand to be high enough for evaporating the most important parts of the inorganic ions (especially ammonium nitrate and ammonium sulfate) as well as most organic carbon and on the other hand, as a *temperature low enough to prevent charring of organic compounds. The previous sentence will be added in the manuscript on section 2.3.*

We completely agree in the fact that comparing volatility measurements between different setups is quite difficult due to different working conditions including temperature, residence time and setups as mentioned by the reviewers. For these reasons and as mentioned before, working temperature was explicitly mentioned in the title as a reference value to provide an explicit definition of the term "non-volatility" used in the manuscript.

In contrary to the HTDMA community, the TD community is not so well structured and no large and systematic intercomparison workshops between different volatility setups were performed up to now to define a so-called "standardized" operation procedure or recommendations for both temperature and residence time. Only An et al. (2007), provided some comments on the influence of the retention time. Consequently, we clearly agree with the comment that there is a real need to perform such intercomparison in the future. However, drawing such conclusions is too far from the purpose of our work.

Nevertheless to verify the efficiency of our thermodenuder as a function of residence time, we conducted a dedicated sensitivity experiment that will, we hope, at least answer some of the questions raised by the referee. In this experiment, two technically identical mobility particle size spectrometers were set up at the research station Leipzig-TROPOS (Germany, urban background), each equipped with a technically identical thermodenuders upstream of the size spectrometer. The temperature of both thermodenuders was set at 300°C, as during long-term measurements in Melpitz. The flow rate passing through one thermodenuder was kept constant, at 0.3 l/min while it ranged between 1 and 10 l/min in the second thermodenuder. Results of this sensitivity test reveal that no remarkable effect of thermodenuder flow rate could be seen on the remaining volume concentration at least for our range of flow rate settings (1-10 l/min). We therefore see no indication that the residence time in the thermodenuder (3.1s at Q = 0.3 l/min) would be insufficient to evaporate all particulate volatile material at 300°C. The results of this sensitivity test are relevant enough to be included as an Appendix in the manuscript as following:

Appendix A: Verification of thermodenuder efficiency

To verify the efficiency of our thermodenuder (Wehner type TD) as a function of residence time, we conducted a sensitivity experiment. For this purpose, two technically identical mobility particle size spectrometers were set up at the research station Leipzig-TROPOS (Germany), each equipped with identical thermodenuders upstream of the device (Fig. A1). The temperature in both thermodenuders was set to 300°C, as during the long-term measurements in Melpitz. Thermodenuder 1 was flushed by a constant flow of Q_1 = *3 l/min, which also corresponds to the nominal sampling flow during long-term measurements. Thermodenuder 2 was operated in the same fashion except that its sampling flow* Q_2 *could be varied between 1 and 10 l/min by means of an additional make-up flow. During the experiment, approximately 16 days of valid data were collected. Four different settings for* Q_2 *were employed: 1, 3, 5 and 10 l/min.*

The results of the experiment with ambient aerosols are depicted in Fig. A2. The upper graph shows the time series of the remaining particulate volume (< 800 nm) of both instruments (V_1 *and* V_2*). It can be seen that* V_1 *and* V_2 *follow each other very closely over most parts of the* *experiment, including large overall variations of total particle volume. The bottom graph of Fig. A2 confirms that the ratio* V_2/V_1 *between the volumes remaining downstream of the thermodenuders straddles around the value of 1.*

Fig. A3 and Table A1 sort the data into different values of Q_2*. For the nominal flow* $Q_1 = Q_2 = 3$ *l/min the thermodenuders agree very well on average (*$V_2/V_1 = 1.00$*). The same holds for* $Q_2 = 5$ *l/min. Increasing the residence time by choosing* $Q_2 = 1$ *l/min yields* $V_2/V_1 = 1.03$*, i.e. a slight decrease in the efficiency of thermodenuder 2, despite an increased residence time. For* $Q_2 = 10$ *l/min, i.e. the shortest residence time,* $V_2/V_1 = 1.05$*. The latter value might be interpreted as a decrease in thermodenuder efficiency as a result of decreasing residence time. It needs to be noted, however, that these deviations are on order of the measurement accuracy of the instruments.*

Our conclusion is: we cannot observe any remarkable dependency of the remaining volume after the thermodenuder on the sampling flow. We can safely assume this across the flow range 1 l/min < Q_2 *< 5 l/min.* $Q_2 = 10$ *l/min There is therefore no indication that the residence in the thermodenuder (3.1 s at Q = 3 l/min) would be insufficient to evaporate all particulate material volatile at 300°C.*

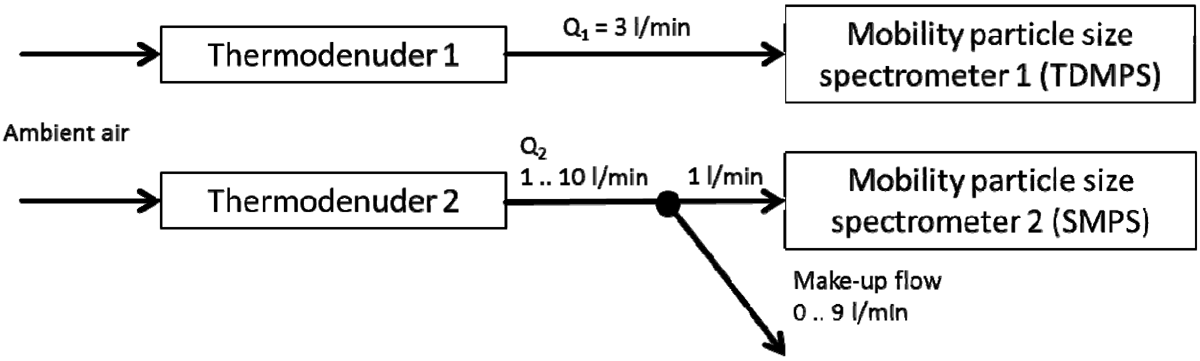

Figure A1: Experimental set-up to test the sensitivity of the thermodenuder efficiency as a function of residence time.

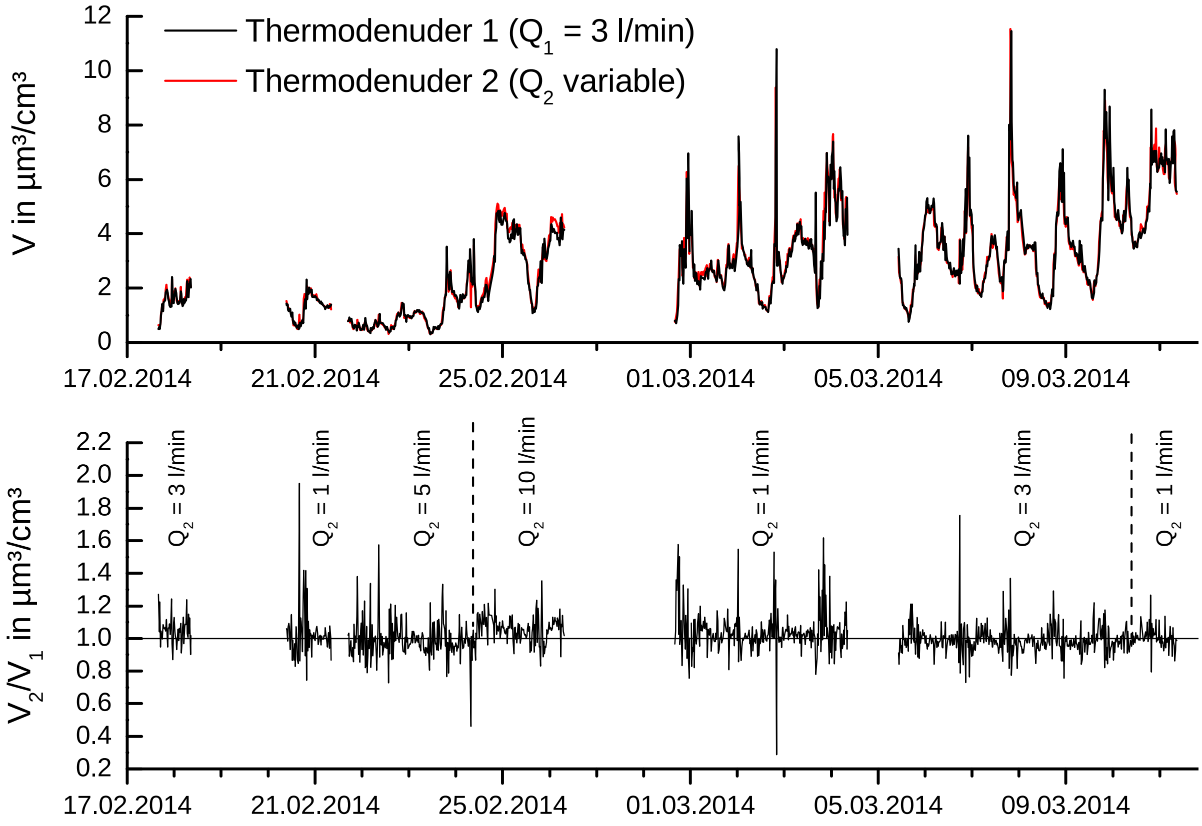

Figure A2: Upper graph: time series of particulate volume (< 800 nm) of ambient aerosol remaining after passage through the thermodenuders at 300°C. Bottom graph: ratio V_2/V_1 between the volumes remaining downstream of the thermodenuders.

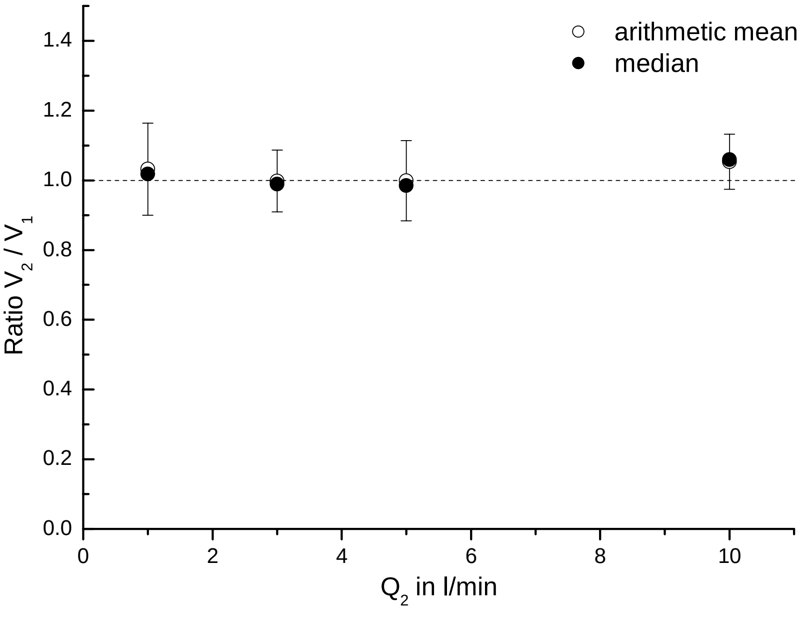

Figure A3: Ratio V_2/V_1 between the remaining volumes as a function of the sampling flow through Thermodenuder 2. Error bars correspond to standard deviation.

Table A1: Ratio V_2/V_1 between the remaining volumes as a function of the sampling flow through Thermodenuder 2. The minimum residence time t_{res} of the air sample in the heating unit is indicated in s.

Q_2 (l/min)	T_{res} (in s)	Volume ratio (V_2/V_1)		
		mean	*st. dev.*	*median*
1	9.4	1.03	0.13	1.02
3	3.1	1.00	0.09	0.99
5	1.9	1.00	0.12	0.99
10	0.9	1.05	0.08	1.06

Finally we have to apologize for a mistake regarding residence times reporting in the manuscript. Based on the heated part of the thermodenuder (500 mm length and 20 mm internal diameter), the real residence times were 3.8 to 3.1 seconds for a flow rate of 2.5 and 3 l/min respectively. It will be corrected.

The other major problem is the implication that thermal decomposition is comparable to a fraction of LV-OOA derived by PMF. Authors rightly suggest that ammonium nitrate and ammonium sulphate decompose at temperatures below 200C, but is this true for highly internally mixed particles? If sulphate is (partially) coated by BC or humic like OM, will it decompose equally well within 1s residence time? If not then ROA is overestimated. If, similarly, there is significant amount of dust or processed dust ($CaSO_4$) then ROA is also overestimated. More importantly, how the limited (ambiguous) particle volatility in the denuder is comparable to the fraction of non-volatile LV-OOA in terms of the chemical make-up? Taken the above altogether, I am not sure the term refractory is correctly used in the study and is useful to the community.

Response: The referee point out couple of important points that will be answered separately below.

- In previous work, Wu et al. (2009) compared the mass spectrum of humic and fulvic acids considered as surrogate for LV-OOA at ambient and 300°C. Although some modification of the mass spectra was stated and attributed to the loss of water molecules and a small decarboxylation, the structure of the mass spectra was preserved and could still be compared

with the ambient spectra. This was confirmed by Huffman et al. (2009) who successfully compared PMF results from ambient and coupled ambient-TD measurements and concluded that "any degradation of the mass spectra as a result of passage through the TD is compensated by the enhanced contrast between the different factors". Therefore, it appears to be accurate to use the ambient LV-OOA mass concentration derived by PMF to estimate its non-volatile fraction when its remaining mass fraction at a defined temperature is known.

The text of the section 3.2.3 will be changed as follow: „Previous field measurements, using a coupled TD-AMS set-up (Huffman et al., 2009) demonstrated that similar factor analysis components and time series results were observed when comparing ambient only PMF results with joint ambient-TD results. Therefore, it seems to be possible to extrapolate the non-volatile fraction at a specific temperature from the ambient PMF factors when their MFR at this temperature are known. […]. Additionally, changes of the mass spectra of the fulvic and humic acids at 300°C compared to the ambient mass spectra were attributed to the loss of water molecule and decarboxylation."

- The most important point in our approach is to properly identify the influence of the non-detected and non-volatile compounds like mineral dust and sea-salt to the non-volatile composition. As properly mentioned by the referee, the presence of such compounds on the PM_1 *size range will contribute to the overestimation of the non-volatile organic aerosol (NVOA). For this reason, mass closure of the* PM_1 *was made and discussed in terms of comparing AMS and MAAP to TDMPS as well as by considering off-line filter results. Any discrepancy between chemical composition (AMS+MAAP) and TDMPS might reflect the presence on a significant fraction of non-detected compounds. The mass balance of ambient* PM_1 *made for the two sampling periods emphasized a low contribution of non-detected compounds compared to organic and black carbon according to both instrumental and method uncertainties. Consequently, based on the different uncertainties (V-TDMPS, TDMPS, BC density…) our NVOA estimation should rather be considered as an upper limit than an absolute concentration.*

The following sentence will be added to the conclusion: „Since contribution of undetected and non-volatile compounds (e.g. dust, other inorganic ions) cannot be completely excluded, the present NVOA values have to be considered as the upper limit that may be expected for each period".

- It is true that internally mixed particles might evaporate differently from pure salt. However, Huffman et al. (2009) using direct AMS measurements downstream their thermodenuder, compared thermogramms of nitrate and sulfate obtained during field studies and pure ammonium nitrate and sulfate salts. The authors reported a residual mass fraction of inorganic salts at high temperatures which might either be related to incomplete evaporation of inorganic salts from internally mixed particles or reflects the presence of non-volatile organonitrate / organosulfate compounds. Nevertheless the inorganic mass fraction remaining was very low below 5-10% at 250°C and even lower mass fraction remaining has to be expected at 300°C. This is also confirm by the results we have obtained from our sensitivity test where no indication was found that the residence time in the thermodenuder (3.1 s at Q = 3 l/min) would be insufficient to evaporate all particulate volatile material at 300°C.

The uncertainties of the many parameters used in this study make the chemical balance very fragile. Authors may be right and lucky at the same when deriving rather arbitrary parameter like density. Will the same approach work in other geographical location? Given different density of OM, BC or the presence of crustal material, or significant amount of sea salt, may completely render the usefulness of the method. Or the PMF results at different location may suggest different partitioning between SVOOA and LV-OOA which would suddenly result in different quantitative conclusions about volatile LV-OOA fraction. The results of this study are already suggesting about 50% uncertainty in refractory LV-OOA which is huge.

Response: Influence of dust and sea-salt to ambient and non-volatile PM_1 particles was already discussed in the answer of the previous comment. We will only focus on the PMF results here. We agree that changes on the LV-OOA volatility between the sampling periods can be interpreted on the manuscript as resulting from the uncertainty of the LV-OOA volatility measurements and then some more explanations are needed. Although the change on the LV-OOA volatility is not reflecting some uncertainty on the volatility measurements, it can be related to the change of the physical properties of the factors.

Although similar PMF factor can be identified for different measurements periods or sites, it does not mean that mass spectra of these factors are ever 100% identical. Some variations on the mass spectra can results on changes on the initial sources (e.g. different biomass burning conditions, or SOA precursors) and/or different aging processes. Since the AMS and PMF cannot really describe the chemical composition of the organic aerosols, the better way to highlight these variations is to follow their oxidation level. Ng et al., 2010 summarized the changes in the oxidation level of different PMF factors obtained during several field campaigns over the northern hemisphere in a triangle space. The less oxygenated compounds are falling in the bottom of the triangle while during aging processes the OOA tend to reach the top of the triangle space. Comparing factor analysis results of 25 AMS measurements across Europe, Crippa et al. (2014) *provided an excellent demonstration of the diversity of the LV-OOA factor (Fig. SI-4 in Crippa et al., 2014). These differences between similarly identified factors might also lead to changes in the physical properties of the factors like hygroscopicity and volatility (Jimenez et al., 2009). Consequently, to better answer this question, the two LV-OOA mass spectra were compared in the Ng's triangle space. Results show that during winter, the LV-OOA was slightly more oxygenated than during summer which is in agreement with the idea that volatility of organic aerosol decrease with increase of the oxidation level (Jimenez et al., 2009). Consequently, changes of the volatility properties of the LV-OOA have to be considered as principally resulting from the differences in chemical composition (illustrated by the oxidation level of the factor) rather than related to uncertainties of the LV-OOA volatility determination.*
This will be included as described in the following:

"Differences in the volatility properties of the two LV-OAA factors may result from the differences in their oxidation states. To check this hypothesis, mass spectra differences were highlighted and plotted in Figure 8, f44 (the fraction of m/z44 to total organic mass spectra) vs. f43 (the fraction of m/z 43 to total organic mass spectra) in the triangular space presented by Ng et al. (2010). This triangle plot represents a simple and practical approach to compare different organic factors with different oxidation levels since they will fall in different area of the triangle. The less oxidized factors (e.g. HOA) are usually present in the bottom of the triangle whereas the most oxidized factors (e.g. LV-OOA) are usually present in the upper part of the triangle. The top part of the triangle tend to suggest that SOA oxidation level become more and more similar after long aging processes (Ng et al., 2010). Comparing the position of each LV-OOA factor highlights that winter LV-OOA was located slightly above the summer LV-OOA indicating that winter LV-OAA was more oxygenated than the one in summer. This small difference in term of oxidation state might explain the observed change in the volatility properties of the two LV-OOA factors and the fact that the less volatile factor (winter time, MFR 52%) is also the one with the highest oxidation level."

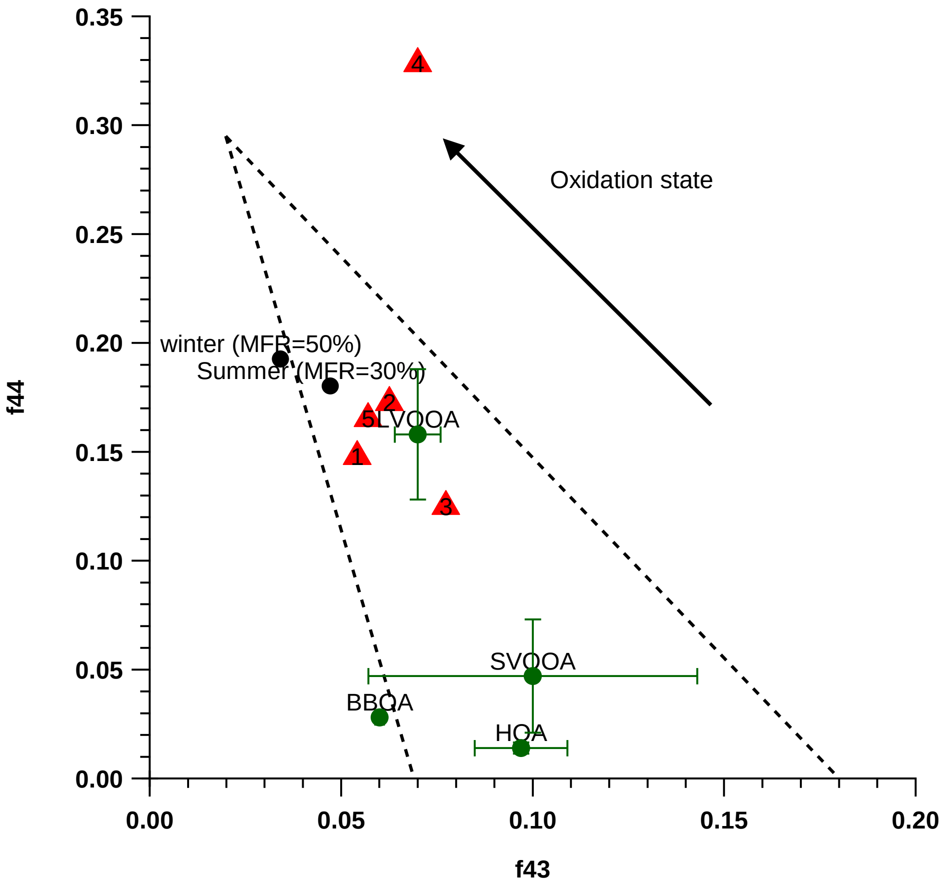

Figure 8: f44 vs. f43 of each LV-OOA factors in the triangle space determined by Ng et al. (2010). The green points referred to the average values (+/- standard deviation) of factors used by Ng et al. (2010). Red triangle and numbers referred to different examples of LV-OOA factors (1: Hersey et al. (2011), 2:Lanz et al. (2007), 3:Ulbrich et al. (2009), 4:Robinson et al. (2011), 5:Crippa et al. (2013)) illustrating the individual variability of LV-OOA.

Additionally, the sentence in the abstract was modified to: "Our results suggest that LV-OOA was more volatile in summer (May-June 2008) than in winter (February/March 2009) which can be linked to a difference in oxidation levels (lower in summer)."

Some minor (other) comments

Abstract. Authors claim that their results suggest that organics was more volatile in summer, but the percentage of ROA was higher in summer (53%) then winter (47%). Something does not make sense.

Response: This is a confusion of 2 different parameters: 1- the volatility of the organic is reflect by its mass fraction remaining (MFR) at the working temperature of 300°C. The lowest is the MFR, the more volatile is the compound. Since our results estimate a MFR of 30% for summer LV-OOA and 52% for winter LV-OOA, it is true to conclude that in summer LV-OOA is more volatile than in winter. 2- the mass concentration of ROA inside the non-volatile particulate matter is calculated by the equation ROA = [LV-OOA] × MFR. Ambient concentration of LV-OOA was higher in summer (mean concentration 2.96 µg m^{-3}*, leading to* *a mean non-volatile fraction of 0.88 µg* m^{-3}*) than in winter (mean concentration of 0.64 µg* m^{-3}*, leading to a mean non-volatile fraction of 0.33 µg* m^{-3}*). At the same time, average black carbon concentrations were relatively similar for both periods as mentioned in the manuscript. Therefore, the resulting non-volatile OA mass fraction (53% in summer and 41% in winter) as well as mass concentration appears to be larger in summer even if LV-OOA is in fact more volatile.*

P26987, line 16. Authors assert that AMS cannot detect sea salt, but that has been disproved by Ovadnevaite et al. 2012 (JGR). That study does not invalidate refractory nature of sea salt, but sea salt can certainly be quantitatively detected by AMS.

Response: It is true that Ovadnevaite et al. (2012) were able to detect and quantify sea salt using an HR-ToF-AMS at the marine research station of Mace Head (Ireland). However, at standard AMS working conditions direct measurements of sea-salt by AMS in a similar way as nitrate and sulfate in quite challenging. Sea-salt starts to evaporate at 600°C and fully evaporate at 650°C. Therefore, the HR-ToF-AMS used at Mace-Head was specially tuned for this purpose including a vaporizer temperature setting at 650°C instead of standard 600°C. Nevertheless, manuscript will be changed in order to clearly mention the point that in some conditions, sea-salt can be detected by AMS as follow:

"Because soot, crustal material and sea salt can usually not be properly detected by the AMS, while Ovadnevaite et al. (2012) demonstrated the possibility to measure sea-salt, the AMS is commonly considered to only provide non-refractory PM_1 *aerosol chemical composition".*

P26988. Authors rightly consider uncertainty in mass absorption coefficient, but that would introduce not only systematic uncertainty, but also would add significant variability as the mass absorption coefficient can be constantly changing. For example, absorbing humic like substances would result in higher mass absorption coefficient.

Response: We agree with this comment. It is absolutely true that the mass absorption coefficient can be constantly changing as it was mentioned in the manuscript (p26988, l11): "However, model calculations and experimental results from the literature suggest that the mass absorption coefficient of atmospheric soot particles is not constant, but may depend on the state of mixture with other compounds".

The mass absorption coefficient used in this study was estimated by Nordmann et al. (2013) by coupling MAAP measurements and laboratory analysis of the corresponding MAAP filters by Raman spectroscopy for 7 different sampling sites over Germany and covering 3 different periods of the years 2009-2010. The resulting value of 5.3 $m^2\ g^{-1}$ *was considered by the authors as representative for the soot mass absorption coefficient over the Central Europe. Although this value appears to be more accurate that the default value; it cannot exclude the fluctuations of this coefficient during measurements due to present of other absorbing compounds. This sentence will be included in the manuscript.*

P26991, line 5. Authors assert TDMPS uncertainty at 10% (probably based on sizing precision in Wiedensohler et al (2012, AMT)), but it should rather be 33% due to volume conversion when making its use in mass balance.

Response: It is absolutely true that the 10% uncertainty of the TDMPS is based on the sizing precision of Wiedensohler et al. (2012). However, in the same paper, authors also highlight that same uncertainty of 10% was found when considering either number or volume size distribution (See Fig. 8 and 9 from Wiedensohler et al. (2012)).

P26992, line 9. R2 value is called "measure of determination", in other places "correlation", but in fact it is VARIANCE, or the percentage of the total variance explained by the common factor. The true correlation is "r", not R2.

Response: We disagree with the comment. The coefficient of determination (R^2) is commonly used for fitting to express how well the data points fits. However, and as suggested, we now describe the linear correlation coefficient by the r-Pearson value (r).

P26992, line 19. Authors need a proof that ammonium sulphate evaporates at 150C in internally mixed particles, not only of pure species in laboratory setting. Please refer to consequences above.

Response: This is an interesting point and we fully agree that internally mixed particles might react differently than pure species. However, as mentioned before our sensitivity test demonstrated that using our setup and our working conditions, there is no indication that the residence in the thermodenuder would be insufficient to evaporate all particulate volatile material at 300°C. Therefore, a complete evaporation of ammonium sulfate can be expected. Moreover, Huffman et al. (2009) using direct AMS measurements downstream their thermodenuder, compared thermogramms of nitrate and sulfate obtained during field studies to pure ammonium nitrate and sulfate salts. The authors reported a residual mass fraction of inorganic salts at high temperatures (200-250°C) which might either be related to incomplete evaporation of inorganic salts from internally mixed particles or reflects the presence of organonitrate / organosulfate compounds. Nevertheless the inorganic mass fraction remaining was very low below 10-5% at 250°C and even lower mass fraction remaining has to be expected at 300°C.

Consequently, the text will be modified to consider this aspect as follow: "In real environment, particle composition is much more complex than pure standards; therefore evaporation of inorganic salts in internally mixed particles might slightly differ from that of pure salts. However, Huffman et al. (2009) reported only a residual mass fraction of nitrate and sulfate at 250°C slightly below 10% which cannot be clearly attributed to either inorganic salts from internally mixed particles that did not fully evaporated or non-volatile organosulfate / organonitrate compounds. Consequently, at 300°C, contribution of ammonium nitrate and sulfate are expected to be quite negligible (<10%)".

Line 26. This is rather a major comment. Authors invoke the presence of marine particles when explaining strange mode at 400nm, but they quantitatively show that marine species contribution was small in PM2.5 and negligible in PM1. Something does not add up here. Can it be that larger particle could not reach thermal equilibrium in the denuder due to short residence time? Clearly, larger particles would require longer time to decompose inside out than smaller ones. This re-iterates the impact of residence time on the results. Was the size effect studied in at least laboratory setting?

Response: There is confusion between PM_1 *from the winter campaign and* $PM_{2.5}$ *from the summer one. Contribution of marine particles was only observed during winter time based on air mass trajectory and Berner impactor measurements. As mentioned in the manuscript during the winter campaign chloride, sodium, calcium, potassium and magnesium all together represented around 4% of the total* PM_1 *mass. Consequently for the winter period, no significant marine particle can be expected in the submicron size range (*PM_1*). The marine aerosols mentioned in the manuscript and by the referee are located in the range 1-2.5µm which is out of the scanning range of the V-TDMPS. The summer* $PM_{2.5}$ *measurements which only referred to the summer campaign due to the lack of* PM_1 *measurements at this time indicated that just a very small contribution was expected due to the low concentration of sodium and chloride ions for this time period in contrast to the winter measurement period.*

The TDMPS has an upper size cutting of 800 nm and then cannot measured super-µm particles. However after passing through the thermodenuder the diameter of the super-µm particles might shrink to sub-µm range due to the loss of their 300°C volatile fraction. Consequently, these particles would be counted by the V-TDMPS measuring downstream to the thermodenuder at 300°C but not during ambient measurements. A direct consequence is an overestimation of the VFR and MFR which might represent a large artefact for volatility measurements. Because no measurement of the super-µm particles downstream of the thermodenuder was performed, further investigations are needed to better understand how these particles evaporate inside the thermodenuder.

For these reasons, we made the suggestion that the strange mode above 400 nm was assumed to be related to a diameter shrink from coarse mode to sub-µm particles. This hypothesis was confirmed by the impactor measurements which clearly show the presence of a larger super-µm mass concentration when concentrations of non-volatile particles larger than 400 nm increased. Finally, in order to provide homogeneous conclusions and properly compare winter and summer periods, the same upper size cutting was also used for the summer campaign and no marine influence was detected. According to the comment of the second referee a time depend method was employed to better estimated the upper size limit of the V-TDMPS measurements. We invite the referee to read our answers to the first referee.

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