Responses to reviewers' comments for manuscript

Variation in chemical composition and volatility of oxygenated organic aerosol in different rural, urban, and mountain environments

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Reviewer 1 *(responses in italics)*

Organic aerosols are a major contributor to total aerosol mass concentrations and have implications for both human health and climate change. However, the formation of these aerosols is a complex supersaturation-driven process, involving highly dynamic vapor-particle interactions. Therefore, constraining the volatility of condensable vapors and the associated particles is critical for understanding the underlying oxidative chemistry and for better representation of organic aerosols in air quality models.

This paper presents data from ambient measurements of the chemical composition and thermogram of organic aerosols in various environments using the online and offline FIGAERO-CIMS methods. In addition, the authors estimated the particle volatility using a volatility parametrization and compared it with the thermal desorption profile in the lumped thermogram. The research topic of this paper is novel, the dataset is comprehensive, and the measurement techniques are state-of-the-art. Overall, this is a relevant study that fits within the scope of the ACP. However, the way the results are interpreted and discussed needs major revision to improve scientific rigor and to make it clearer to non-specialist readers. Here are my major comments:

We thank the reviewer for the constructive assessments of our manuscript. We appreciate your recommendation for publication in ACP and your time and consideration. We have carefully considered your suggestions and questions and addressed them in our revised manuscript. The point-to-point responses to the comments are given below.

1. While the dataset is comprehensive and covers various environments, I'm not convinced that it does not have a global representation. I would suggest that the authors remove text such as "across the globe" and "global dataset".

We understand the reviewer's concern and have removed the corresponding text as suggested throughout the manuscript.

2. Volatility calculation: It is not clear whether the authors used one parametrization for all data, or different parametrizations for data in different environments (or rather, for different types of compounds). If it's the former, I would suggest that the authors redo some of the calculations, because molecular formulas with the same number of carbon and oxygen atoms can have very different volatilities due to different functionalities (e.g., -OH and -OOH reduce volatility by the same amount). This could be a source of discrepancy between the calculated volatility and the thermal desorption profile. The authors can refer to this paper for further information (https://doi.org/10.1038/s41561-022-00922-5).

We have used the same parametrization for all data and have clarified that in the manuscript in Line 176. As parametrizations are developed based on different datasets comprising different compounds with different functionalities, different parametrizations can yield different results for the same dataset (e.g., Graham et al., 2023). However, for ambient datasets, to conclude the structure and functional groups from the molecular composition given by the mass spectrometer measurements will always remain speculative, at best, and requires ancillary data and measurements. We therefore believe that using the same parametrization for all datasets is the approach with the least bias. This is to some extent in agreement with the study by Nie et al. (2022) suggested by the reviewer. Nie et al. (2022) used different parametrization methods to calculate the volatility of gaseous oxygenated organic molecules (OOMs) detected by nitrate-CIMS, after identifying and classifying the OOMs based on their potential precursors using a novel workflow proposed in their paper. However, they also state that this workflow cannot be used to classify particle/aqueous-phase products, since the assumptions behind the classification are mostly based on gas-phase oxidation knowledge, and the data we present in our manuscript are from the particle phase.

We agree however with the reviewer that a comparison of different parametrizations could be instructive. Graham et al. (2023) did exactly this, investigating in depth the application of four different parametrizations to their chamber datasets on biogenic VOCs oxidized with either ozone or the nitrate radical and exposing the complexity of such an approach. They found that none of the four parametrizations investigated performed perfectly for all different biogenic terpene+NO_3 systems. For example, the Mohr et al. (2019) parametrization, tuned for -OOH functional groups, was found to work better for α-pinene+O_3 system, and thus it was speculated that the reaction products of α-pinene+O_3 have relatively more -OOH functional groups. The Daumit et al. (2013) parametrization assumes all oxygen atoms except those from -NO_3 are from -OH and =O functional groups and was found to work better for the α-pinene+NO_3 reaction products. These results underline again the complexity (and with that also the limitations) of assessments of the volatility (especially at the quantitative level) through parametrizations using the molecular composition as input, and even more so for complex ambient data from different locations and periods.

In order to clarify that, we have now added a comparison of results from the parametrization initially used in the manuscript (modified Li et al. (2016) method (Isaacman-VanWertz and Aumont, 2021;Daumit et al., 2013)) to results from using the parametrization by Donahue et al. (2011) (Figure R1a) if making a simple assumption about predominant aliphatic/aromatic OOMs at urban sites in winter (i.e., UST-w, UKA-w, and UDL) and predominant isoprene OOMs in MCC-t and RAB (Nie et al., 2022). The $log_{10}C_{sat}$ (T) shows a similar trend for both parametrizations; however, the C_{sat} (T) using the Donahue et al. (2011) parametrization is on average 4 to 8 orders of magnitude lower, with the largest difference for UKA-w and smallest for UDL (Figure R1b). The discrepancies seem to be correlated with the average number of oxygen atoms (nO) in the bulk aerosol of the different environments (Pearson's R: 0.85; Figure R1b), confirming the influence of different functional groups on parametrization results. Our discussion of volatility results in this manuscript is therefore mostly qualitative. We also note that the correlation coefficients between thermal desorption-derived volatility and molecular composition-derived volatility for the two different parametrizations are comparable (Figure R1c).
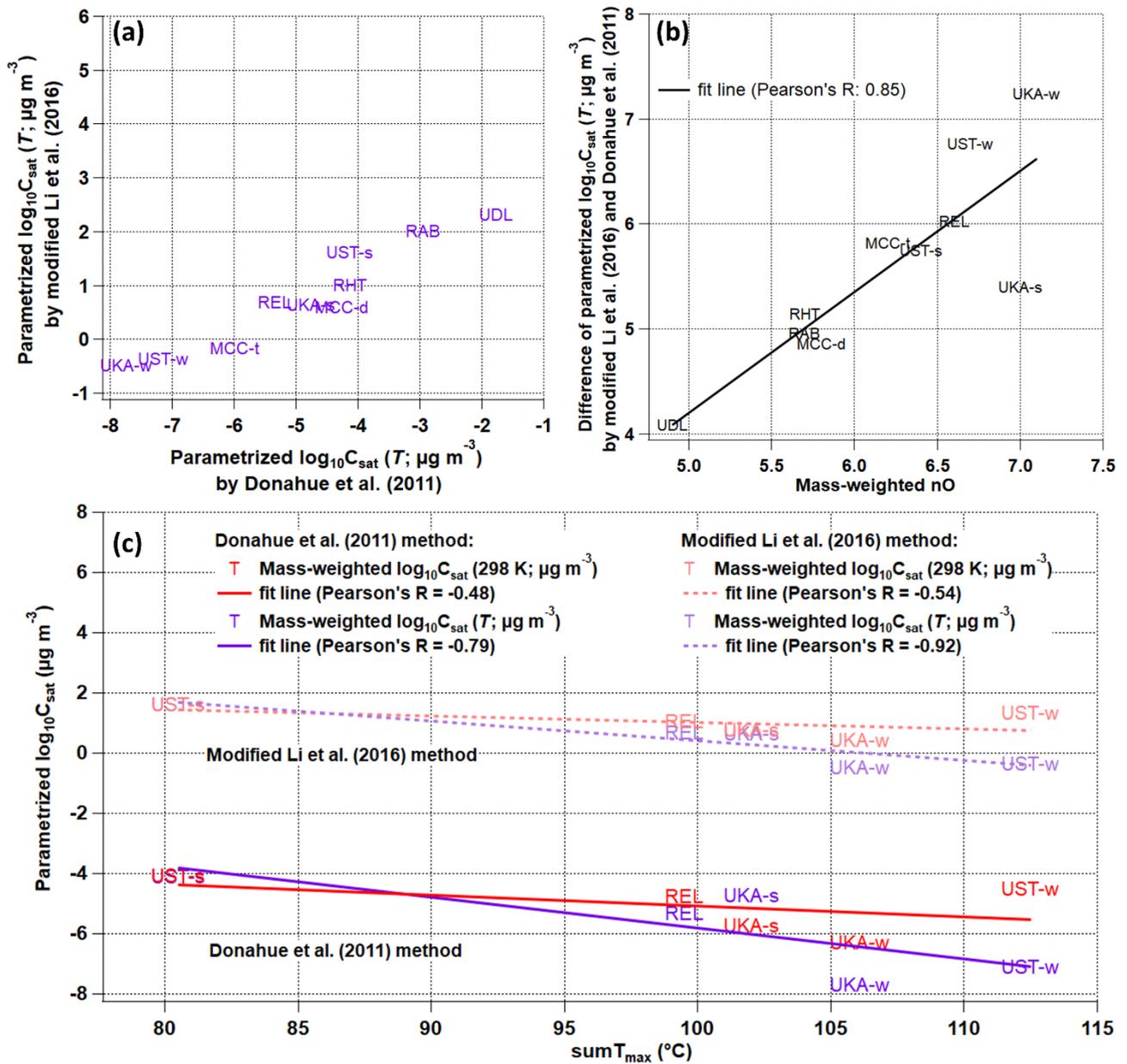
Figure R1. (a) *Comparison of the campaign-average mass-weighted $log_{10}C_{sat}$ (T) values using the modified Li et al. (2016) parametrization method (Daumit et al., 2013;Isaacman-VanWertz and Aumont, 2021) and the Donahue et al. (2011) method. **(b)** Comparison of the difference of campaign-average mass-weighted $log_{10}C_{sat}$ (T) values using the modified Li et al. (2016) parametrization method (Daumit et al., 2013;Isaacman-VanWertz and Aumont, 2021) and the Donahue et al. (2011) method with the mass-weighted number of oxygen atoms (nO). **(c)** Comparison of the campaign-average mass-weighted $log_{10}C_{sat}$ values and $sumT_{max}$ values for different locations and seasons (only datasets where the exact same FIGAERO setup was used), with mass-weighted $log_{10}C_{sat}$ (298 K) in red and mass-weighted $log_{10}C_{sat}$ (T) in purple; the colored lines are the fit lines for the corresponding markers.*

We have added Figure R1 as Figure S1 (all other SI figures are now Figure S2-S10) as well as more discussion in Lines 176, 184, 202, and 531 of the revised manuscript:

Line 176: "C_{sat} (298 K) was calculated using the approach by Li et al. (2016) as in equation (1) for all sites in order to not introduce more bias".

3. Carbon number analysis: The author did not justify that nC alone can tell us much about the emission source, especially when comparing data from different environments. For example, nC for the urban sites is greater than that for the rural sites, but then nC for the MCC-d (anthropogenic influence) is less than that for the MCC-t (biogenic influence). Actually, I don't really see a pattern in the nC analysis. I would suggest that the authors condense the discussion and focus more on the oxidation state.

(Bianchi et al., 2022;Zha et al., 2023b); and 3) the impact of monoterpene oxidation products for MCC-t (also seen in the slightly higher contributions of C_{8-10} compounds other than C_5 compounds for MCC-t in Figure S2).

For clarification, we have modified the discussion in Lines 214 and 229 of the revised manuscript as following:

Line 214: "We focus our analysis of the bulk molecular composition of OOA from the different locations here on the average number of carbon (nC) and oxygen atoms (nO) as they affect the parametrized C_{sat}, and give an indication of potential precursor sources (Huang et al., 2019a) and oxidation processes, respectively."

Line 229: "The air masses arriving at Chacaltaya present a diverse mixture from various sources, which vary significantly between the transition and the dry seasons (Aliaga et al. 2021). While air masses arriving at Chacaltaya during the dry season are more affected by volcanic SO_2 emissions as well as anthropogenic compounds such as toluene (also seen in the slightly higher contributions of C_7 compounds in Figure S2) (Bianchi et al., 2022;Zha et al., 2023b), air masses reaching Chacaltaya during the transition season have more influence from e.g. the Amazon Basin (Aliaga et al., 2021;Zha et al., 2023b); hence, a higher contribution of monoterpene ($C_{10}H_{16}$) and especially isoprene (C_5H_8) oxidation products is expected (also reflected in the clear peak of C_5 compounds and the second hump of C_{8-10} compounds in Figure S2) (Zha et al., 2023a)."

4. Mass contribution of organonitrates: Lacking explanations. Why do remote mountain sites have such a high CHON fraction? I don't think "in accordance with their proximity to forested areas" explains it. And why "possibly due to the less efficient production rates of CHON"?

The higher CHO contributions at the rural sites (79.9±5.2%) compared to the mountain (66.2±5.5%) and urban sites (72.6±4.3%) are in accordance with their proximity to forested areas, as stated in Line 304. The less efficient production rates of CHON or their faster loss rates caused by hydrolysis due to high RH apply only for the rural station in the southeastern U.S. (RAB; 5.6±2.6%), as stated in Line 305 and several earlier publications (Lee et al., 2020;Hu et al., 2011;Lee et al., 2016;Pye et al., 2015).The reason for the mountain site MCC to have such a high CHON fraction could be related to the inorganic nitrogen pollution being transported up there from the polluted boundary layer of La Paz–El Alto metropolitan area (Bianchi et al., 2022), as well as the long range transport of CHON species from the Amazon Basin (Zha et al., 2023a). Besides, CHON contributes more (23.9±4.1%) in the dry season (MCC-d) than in the transition season (MCC-t, 18.7±5.9%), as a result of enhanced transport of anthropogenic pollutants from the La Paz–El Alto metropolitan area during the daytime in the dry season (Bianchi et al., 2022;Aliaga et al., 2021;Zha et al., 2023b).

Given this, we have removed text like "remote" or changed it to "mountain" for the mountain MCC site description throughout the manuscript. For clarification, we have added more discussion in Line 304 of the revised manuscript:

"The nitrogen-containing compounds (CHON) contribute 18.7–31.8% to the total OOA mass, except at the rural station in southeastern U.S. (RAB; 5.6±2.6%). This low contribution could be due to the less efficient production rates of CHON and/or their faster loss rates caused by hydrolysis under the high RH conditions there (83.1±15.2%; Table S1) (Lee et al., 2020;Hu et al., 2011;Lee et al., 2016;Pye et al., 2015). At the mountain site Chacaltaya, Bolivia, CHON contributes more (23.9±4.1%) in the dry season (MCC-d) than in the transition season (MCC-t, 18.7±5.9%), as a result

of enhanced transport of anthropogenic pollutants from the nearby La Paz–El Alto metropolitan area during the daytime in the dry season (Bianchi et al., 2022;Aliaga et al., 2021;Zha et al., 2023b)."

5. Tmax: "given the large spread in Tmax for …or over a whole campaign." I understand that the authors couldn't quantify the discrepancies between different instruments and operating conditions, but I think the authors should at least discuss how much uncertainty is associated with these discrepancies since the authors are doing the intercomparison after all.

We would like to clarify that the "large spread in T_{max}" per campaign referred to in our manuscript is mostly due to the fact that we aggregate all compounds of all measured mass spectra at a certain location, and these compounds span orders of magnitude of volatilities and hence tens of degrees in T_{max} (Huang et al., 2019b; Lopez-Hilfiker et al., 2014, 2015). Lopez-Hilfiker et al. (2014) have shown that for individual compounds, thermograms and corresponding T_{max} are highly reproducible under stable conditions (within ~2 °C). Differences in instrumental settings between locations and campaigns affect average T_{max} to a lesser degree compared to the chemical composition of the bulk aerosol. Different FIGAERO geometry was speculated to not induce significant T_{max} changes (Ylisirniö et al., 2021). Doubling the ramp rates is expected to induce a T_{max} difference of maximum 5 ºC (Schobesberger et al., 2018;Ylisirniö et al., 2021;Thornton et al., 2020). Correction of the T_{max} values to a filter mass loading of 1 µg following our approach outlined earlier (Huang et al., 2018) only leads to a T_{max} change of <5 °C.

For clarification, we have added more discussion in Lines 420, 476, and 489 of the revised manuscript:

Line 420: "Sum thermogram shapes vary for different environments […] given the large spread in T_{max} […] over a whole campaign period using the same instrument and operating conditions (Thompson et al., 2017) (see an example of $C_6H_{10}O_5$, the molecular formula corresponding to levoglucosan, in Figure S7). However, T_{max} variation due to different instruments or operating conditions is generally smaller than that from the difference in monomers and dimers (Wu et al., 2021) and other factors like the presence of isomers (Thompson et al., 2017;Masoud and Ruiz, 2021) and particle-phase diffusivity, viscosity and matrix effects (Huang et al., 2018;Ren et al., 2022). Different FIGAERO geometry was speculated not to induce significant T_{max} changes (Ylisirniö et al., 2021). The ramp rates for all measurements shown in the present study varied between 6.7 and 13.3 ºC/min (see Table S2), inducing a T_{max} difference of maximum 5 ºC (Schobesberger et al., 2018;Ylisirniö et al., 2021;Thornton et al., 2020). Correction of the T_{max} values to a filter mass loading of 1 µg following our approach outlined earlier (Huang et al., 2018) only leads to a T_{max} change of <5 °C."

Line 476: "For the complex ambient particle matrix, other factors than the pure compounds' vapor pressures come into play, such as the presence of isomers with different vapor pressures (Thompson et al., 2017), thermal decomposition contributions of larger molecules (Lopez-Hilfiker et al., 2015), and matrix effects and viscosity (Huang et al., 2018;D'Ambro et al., 2017;Wang and Ruiz, 2018) even for same compounds in solutions mixed with different species (Ren et al., 2022)."

Line 489: "Such variation may be due to actual changes in effective volatilities of the observed OOA components, due to experimental factors, or both. However, as we discussed earlier, T_{max} variation due to different instruments or operating conditions is generally smaller compared to those due to changes in physico-chemical properties. Therefore, the relative variations in thermogram shapes and T_{max} we present and discuss here in our study are dominated by the presence of isomers (Thompson et al., 2017;Masoud and Ruiz, 2021) and particle-phase diffusivity, viscosity and matrix effects

(Huang et al., 2018;Ren et al., 2022). Due to unavailable calibrations of the relationship between T_{max} and C_{sat} (Wang et al., 2020; Ylisirniö et al., 2021) for most locations, the analysis and interpretation of OOA volatility based on FIGAERO-CIMS thermograms is challenging and, in the absence of additional constraints on OOA composition and thermal behavior, currently remains at the qualitative level (Graham et al., 2023;Voliotis et al., 2021), especially for complex field data."

6. Thermal decomposition: "the decomposition fraction is estimated to be 5.8–35.9%", and "The discrepancy could be due to thermal fragmentation of larger oligomeric molecules, which bias the Csat results towards higher volatilities and the sum thermogram shape to a lesser extent due to the dominance of monomer species". Then how does this affect the volatility calculation using the fragmented formulas? To what extent should we trust the calculated volatility? Need more discussion here.

As we stated in Line 554 of the manuscript: "There have been attempts to separate the thermal decomposition contribution for individual thermograms (Lutz et al., 2019;D'Ambro et al., 2018;Buchholz et al., 2020; Wu et al., 2021), but this poses the threat for introducing new uncertainties due to the difficulty in e.g. differentiating isomers from thermal decomposition products, and monomers from dimers in ambient samples with complex VOC precursors (Graham et al., 2023;Voliotis et al., 2021)." The decomposition estimation approach by Wu et al. (2021) we used in our study represents one option currently available to the best of our knowledge for controlled laboratory studies, but still it is a conservative approach for complex ambient particle matrix. As a sensitivity analysis, we have calculated the average mass-weighted $log_{10}C_{sat}$ (298 K) and $log_{10}C_{sat}$ (T), and $sumT_{max}$ values after removing the fraction of signal from potential thermal decomposition products (Table R1). As expected, the fragmentation effect on sum thermogram is small except for those multi-mode thermograms dominated by the second mode (e.g., MCC-d, RHT, and UDL; see Figure S6). The change in C_{sat} is roughly within +1 order of magnitude, which is within the uncertainties of the parametrization method (Isaacman-VanWertz and Aumont, 2021).

Table R1. *Comparison of the campaign-average mass-weighted $log_{10}C_{sat}$ and $sumT_{max}$ values for different locations and seasons before and after removing the fraction of signal from potential thermal decomposition products.*

Name	$log_{10}C_{sat}$ (298 K) ($\mu g\ m^{-3}$)		$log_{10}C_{sat}$ (T) ($\mu g\ m^{-3}$)		$sumT_{max}$ (°C)	
	Original	*Removed*	*Original*	*Removed*	*Original*	*Removed*
MCC-t	1.6	0.9	-0.2	-1.0	81.7	81.7
MCC-d	2.2	1.5	0.6	-0.3	107.7	99.2
REL	1.0	0.8	0.7	0.4	99.5	99.5
RAB	2.0	2.0	2.0	1.6	94.5	94.5
RHT	2.2	1.9	1.0	0.8	146.3	124
UST-s	1.6	1.5	1.6	1.4	80.5	80.5
UST-w	1.3	1.3	-0.4	-0.4	112.5	112.5
UKA-s	0.8	0.8	0.6	0.8	102	102
UKA-w	0.4	0.2	-0.5	-0.7	106.1	106.1
UDL	2.8	2.7	2.4	2.2	113	88

For clarification, we have added more discussion in Line 346 of the revised manuscript:

"This has an insignificant effect on the bulk molecular composition of OOA particles, and e.g. within the corresponding standard deviation range of nC, nO, and O:C ratios. If we remove the fraction of

signal from potential decomposition products (which is still not ideal), the change in C_{sat} is within the uncertainties of the parametrization method (Isaacman-VanWertz and Aumont, 2021). However, please note that the decomposition estimation approach we used in our study was initially used for controlled laboratory studies by Wu et al. (2021), and thus remains a conservative approach for ambient particle matrix with complex VOCs."

7. In my opinion, some of the conclusions are too strong and lack supporting information. For example:

7.1 "we achieve a comprehensive picture of the relationship between volatility and chemical composition of OOA particles", what is the exact relationship?

It is the aim of our study to investigate the relationship/interconnection between molecular composition-derived volatility and thermal-desorption-derived volatility for complex ambient particle matrix, since this is not always straightforward given the rather large uncertainties of both methods (Isaacman-VanWertz and Aumont, 2021;Voliotis et al., 2021;Masoud and Ruiz, 2021;Wu et al., 2021). For clarification, we have rephrased the sentences in Lines 559 and 576 of the revised manuscript:

Line 559: "Overall, however, for a limited number of our datasets from the exact same instrument, the lower apparent volatility (i.e., higher sumT$_{max}$) agrees qualitatively with the lower $\log_{10}C_{sat}$ values, corroborating potential relationships and interconnections between volatility and chemical composition across different environments and systems despite the large uncertainties and artefacts of both methods."

Line 576: "[...] suggesting a potential relationship/interlink between the volatility and chemical composition for the different locations and systems.

Here, using FIGAERO-CIMS measurements of OOA particles from various locations, we achieve a comprehensive picture of the relationship/interconnection between volatility and chemical composition of OOA particles in different rural, urban, and mountain environments, which are characterized by different ambient meteorological parameters, trace gas levels, emission sources/chemistry, and resulting distinct OOA molecular composition and volatility."

7.2 "however, the effects on the bulk molecular composition and sum thermograms of all detected OOA compounds are small as these thermally-unstable oligomers do not dominate the OOA mass." I would suggest the authors reword it because 35.9% is not small.

Similar to previously reported thermal fragmentation contributions for nitrate SOA in a recent chamber study (1–27%; Graham et al., 2023), the thermal decomposition fractions for our study are 5.8–26.6%, except for one dataset for MCC-d (~35.9%), the sum thermogram of which is multi-modal and dominated by the second mode (see Figure S6). For more details about the thermal fragmentation effect on sum thermograms, please refer to the response to the Major Comment 6. We have also rephrased this sentence in Line 584 as following: "[...] however, the effects on the bulk molecular composition and sum thermograms of all detected OOA compounds for most sites studied here are not significant as these thermally-unstable oligomers do not dominate the OOA mass. For some locations with multi-mode thermograms dominated by the second mode (e.g., MCC-d, RHT, and UDL; see Figure S6), the effects would be larger."

7.3 "and that environmental conditions (e.g., ambient temperature) play a lesser, secondary role through their influence on sources and chemistry of a particular environment," I don't think any

strong conclusions can be drawn about source and chemistry, because there are no analysis of source apportionment and oxidative chemistry.

We agree with the reviewer that we cannot draw strong conclusions about sources and chemistry in our study. However, in previous studies source apportionment was performed for several locations we present here, such as the rural site in southeastern U.S. (RAB) by Massoli et al. (2018), the rural site in Hyytiälä Finland (RHT) by Lee et al.(2018), the urban site in Delhi India (UDL) by Kumar et al. (2022), and so on. These references are properly cited and added to support our discussion in this comparison study.

By comparing the volatility either derived from molecular composition or from thermograms with various environmental and measurement parameters (Figure S5 and S9), we conclude that compared to the chemical composition, the environmental conditions play a smaller role in the apparent volatility of OOA particles for the measurement locations we investigated in our study. However, the environmental conditions such as ambient temperature could affect the sources and chemistry of a particular environment, such as seasonal patterns of biogenic emissions and human behavior (e.g., residential heating), and thus the chemical composition.

7.4 "Our study thus provides new insights that will help guide choices of e.g. descriptions of OOA volatility in different model frameworks" The authors would need to explain more about this.

More explanations are added in Lines 50 and 601 of the revised manuscript:

Line 50: "Our study thus provides new insights that will help guide choices of e.g. descriptions of OOA volatility in different model frameworks such as air quality models and cloud parcel models."

Line 601: "Considering the major contribution of OA to total aerosol mass concentrations and implications for both human health and climate change (Jimenez et al., 2009;Nel, 2005;IPCC, 2021), our study provides new insights that will help guide choices of e.g. descriptions of OOA volatility in different model frameworks. For example, with a better constraint on the volatility of condensable vapors and the associated particles using e.g. the up-to-date CIMS measurements, the study could contribute to understanding the underlying oxidative chemistry and a better OA representation in air quality models. The potential contribution of co-condensation of organic vapors to aerosol forcing or to future cloud radiative effects could also be better accounted for in cloud parcel models (Heikkinen et al., 2023). A better understanding of [...]".

Reviewer 2 *(responses in italics)*

General comment

The authors present evaluations of a combination of aerosol field data taken in 5 different regions of the world (India, Germany, Bolivia, USA, Finland). The central instrumentation is FIGAERO-CIMS, a method often applied in field and laboratory studies. Some seasonal aspects are addressed for the Bolivian and German data sets.

The focus is on comparison of campaign averages for vapor pressures / volatility in relation to particle composition and some other atmospheric parameters. The data set the paper is based on represents a lot of work and effort and is quite impressive.

The manuscript is well written and well organized. The presented material is well chosen and suited to support the discussions and results presented in the manuscript. The manuscript is interesting to read in that presents some critical aspects of vapor pressure and volatility determinations.

The difficulty of the manuscript lies in selection of observations (sites). I believe that they are too singular in time and space to conclude something from the comparison with respect to particle properties in the atmosphere. (I understand that such observations are limited.) This prevents conclusions but very general ones. That is probably the reason why the authors focus more on the methodological aspects. However, whenever they found something interesting, which may be related to atmospheric processes, they step back and question the relations by referring to the experimental difficulties and operational aspects of FIGAERO measurements. The best indication is the statement on page 12 beginning in line 387 and ending in line 398.

And I am not sure if the results support the conclusion that just more efforts ("alternative approaches") are needed "for more quantitative estimations of volatility from FIGAERO-CIMS measurements" (line 534f). Overall, I would say the conclusions are bit weak regarding the atmospheric aspects.

I would still suggest to publishing the paper in ACP as it addresses important aspects and limits of FIGAERO approaches, which should be realized by a broader community. I suggest that authors should address the minor aspects below.

We sincerely thank the reviewer for the thoughtful evaluation and positive feedback of our manuscript. We are grateful for your recognition of the work and effort we put in this study. We appreciate your comments for further improving our manuscript. Below, we address your points in detail:

We agree with the reviewer's comments that the measurement locations presented in our study are limited in time and space. More (long-term) observation would be beneficial for drawing stronger conclusions related to atmospheric aspects for OOA particles in different environments and systems. Therefore, we have removed texts like "across the globe" or "global dataset" throughout the manuscript. However, we would like to stress here that the aim of this work is to investigate the relationship/interconnection between molecular composition-derived volatility and thermal-desorption-derived volatility for complex ambient particle matrix. Overall, for a limited number of our datasets from the exact same instrument, we do find the lower apparent volatility (i.e., higher $sumT_{max}$) agrees qualitatively with the lower $log_{10}C_{sat}$ values, corroborating the potential relationships and interconnections between volatility and chemical composition across different environments and systems. Please refer the changes in the manuscript to the response to the Major Comment 7.1 by Reviewer 1.

And as the reviewer pointed out, one interesting finding of our study is about the large T_{max} variation in different environments and systems in Section 3.2.2. We do agree with the reviewer that this could be related to atmospheric processes. The relative variations in thermogram shapes and T_{max} we present and discuss here in our study are dominated by the presence of isomers (Thompson et al., 2017;Masoud and Ruiz, 2021) and particle-phase diffusivity, viscosity and matrix effects (Huang et al., 2018). Please refer more details to the response to the Major Comment 5 by Reviewer 1. The changes in the manuscript can also be found there.

Minor comments

1. I would say that "volatility" of/in a mixture depends on the chemical composition, i.e. on the vapor pressure of the components, and the physical conditions, mainly the temperature. Translated to atmospheric situations this means that chemical composition depends on emissions and the atmospheric chemistry on the way to the observation point and the physical conditions depend on the let's say the (local) meteorology. Since you are looking at campaign averages ("bulk apparent volatility of OOA particles") you are looking at a kind of a systemic property of aerosol particles, but what you are searching for is still the physical aspects of vapor/pressure of an ensemble of compounds. However, in re-constructing the systemic volatility from the individual components, one is a priori limited by the mass spectrometric approach, which can give (here) chemical formulas at best, and the limits of vapor pressure information for the individual compounds or detected formulas. (On top the operational aspects of FIGAERO measurements.)

We agree with the reviewer that one important limitation of reconstructing systemic volatility from the individual compounds or detected formulae using FIGAERO measurements is the potential divergence from the sum of individual parametrized C_{sat} due to non-ideal intermolecular interactions (Compernolle et al., 2011;Isaacman-VanWertz and Aumont, 2021), as we also stated in Line 332 and 547 of the manuscript, in addition to the uncertainties related to the parametrization for e.g. nitrated organic compounds (Graham et al., 2023). We have added more discussion in Lines 84 and 547 of the revised manuscript:

Line 84: "This could induce uncertainties associated with volatility estimates particularly for the complex ambient particle matrix (O'Meara et al., 2014), in addition to the potential divergence from the sum of individual parametrized C_{sat} due to non-ideal intermolecular interactions (Compernolle et al., 2011;Isaacman-VanWertz and Aumont, 2021)."

Line 547: "[...] the bulk OOA volatility for the complex ambient particle matrix may also diverge from the sum of individual parametrized C_{sat} due to non-ideal intermolecular interactions (Compernolle et al., 2011;Isaacman-VanWertz and Aumont, 2021), in addition to the highly uncertain parametrization of C_{sat} for organonitrates with multiple nitrate groups (Graham et al., 2023)."

2. Could it be that what you tried in this manuscript is inherently an impossible task? A way out could be to drive everything empirically and relate the observations to classes of conditions. However, for that the presented data set is too particulate. Could you explain or justify explicitly your approach using campaign averages?

We agree with the reviewer that it would be too particulate to investigate the classes of conditions and atmospheric processes for each individual dataset. The reason why we use campaign average

values is that we would like to focus our analysis on the systemic bulk molecular composition and bulk volatility of OOA from different environments and systems (to avoid any extreme events or sudden plumes), in order to study whether there is any connection of chemical composition and volatility between molecular composition-derived volatility and thermal-desorption-derived volatility for complex ambient particle matrix.

3. Independently, I am asking myself when the use of campaign averages make sense. Naively, I would say if you had a bimodal distribution of conditions for example, then the campaign average cannot be observed by measurement. This would be different for a simple monomodal distribution of conditions where is a certain chance to indeed observe the campaign average. Can you comment on that?

We think that campaign averages including bimodal distributions still show characteristic features. Taking sumT$_{max}$ values as an example, they are derived from campaign average sum thermograms, which are the summed signal evolution of all CHOX compounds as a function of desorption temperature. We can thus see the distribution of the mode of the sum thermograms over the range of the desorption temperature (single-mode vs. multi-mode) and robustness of using either mean or median sum thermograms (see Figure R2 and also Figure S6). As for log$_{10}$C$_{sat}$ (T), it would be affected by both the composition and meteorological conditions such as ambient temperature (see also Figure S4). Therefore the standard deviations of log$_{10}$C$_{sat}$ (298K) may indicate variations of the distribution. The standard deviations of log$_{10}$C$_{sat}$ (298K) for different locations (Table R2) are within the uncertainties of the parametrization method (Isaacman-VanWertz and Aumont, 2021), and therefore suggests small variation of the log$_{10}$C$_{sat}$ (298K) for the measurement locations investigated in our study.
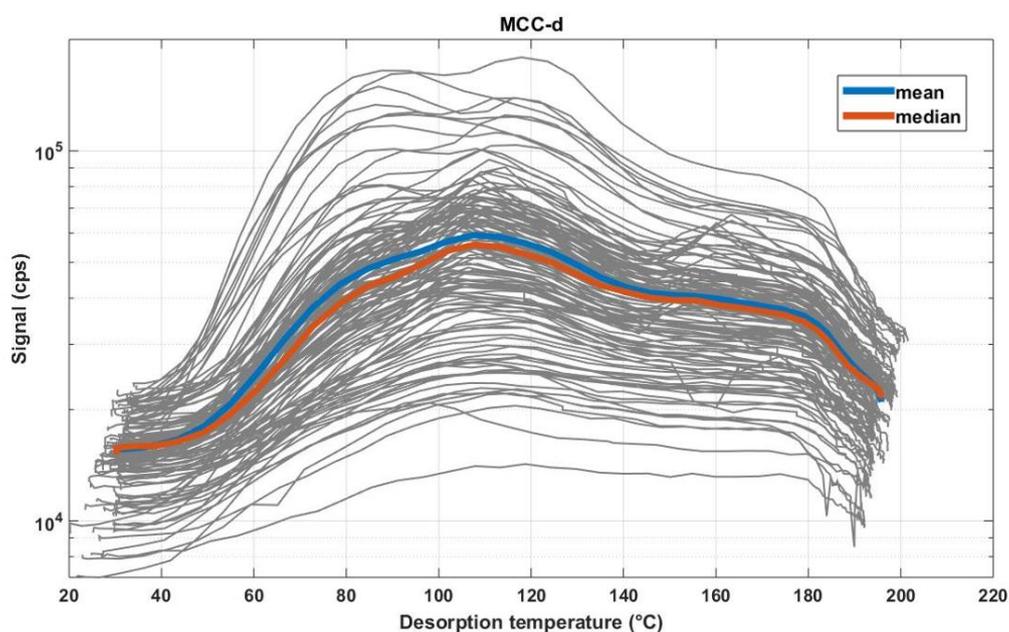
***Figure R2.** An example of multi-mode sum thermograms of CHOX compounds for MCC-d. All campaign sum thermograms are in gray lines, mean one in blue and median in red.*

Table R2. $log_{10}C_{sat}$ (298K) values (average ± 1 standard deviation) for the different locations.

Name	$log_{10}C_{sat}$ (298 K) ($\mu g\ m^{-3}$)
MCC-t	1.6±0.3
MCC-d	2.2±0.4
REL	1.0±0.6
RAB	2.0±0.2
RHT	2.2±0.2
UST-s	1.6±0.2
UST-w	1.3±0.4
UKA-s	0.8±0.6
UKA-w	0.4±0.5
UDL	2.8±0.2

4. line 385: Please can you shortly explain in the experimental sections what sum thermograms are, for non-FIGAERO users. What is exactly is summed in a sum thermogram?

More descriptions are added when first introducing sum thermograms in Line 418 as suggested: "Campaign-average sum thermograms of CHOX compounds (i.e., the sum of mass spectral signal evolution of all CHOX compounds as a function of desorption temperature) for the different locations [...]".

5. line 399- 431: Shouldn't sumTmax tell us something about the persistency of the particles, when they are moving out of the source region?

The desorption temperature at which a compound exhibits maximum signal (T_{max}) correlates with the compound's enthalpy of sublimation and can be used to infer its saturation vapor pressure (Lopez-Hilfiker et al., 2015;Mohr et al., 2017), which in turn influences particulate mass and lifetime in the atmosphere. The sources and transformations of particles affect their role/fate in the atmosphere. After the particles are moving out of the source region, the processes/transformations would affect $sumT_{max}$, i.e., the bulk volatility and thus the physico-chemical nature of the particles (Thornton et al., 2020). Relative variations in thermogram shapes and $sumT_{max}$ we present and discuss here in our study are dominated by the presence of isomers (Thompson et al., 2017;Masoud and Ruiz, 2021) and particle-phase diffusivity, viscosity and matrix effects (Huang et al., 2018). Please refer more details to the response to the Major Comment 5 by Reviewer 1. The changes in the manuscript can also be found there.

6. line 432-448: If I understand correctly, this questions the approach using Li et al. vapor pressure parametrization. If so, that should be mentioned.

In fact, the discussion here are mainly focused on the factors affecting the volatility derived from thermograms, instead of from the Li et al. parametrization method. The results in our study show some discrepancies between the two methods, and based on these results alone it is not possible to judge which method is more correct, which is also not the aim of our study.

7. line 456f: Here you show something interesting, but you discuss it away. If you don't trust the finding, why mentioning it?

We agree with the reviewer and have moved the corresponding text related to the correlation of sumT$_{max}$ with SO$_2$ to the figure captions of Figure S9 in the Supplement to avoid confusion.

8. Figure 2: Please, take out the legend from the figure. It is hiding information. Could you tabulate the values of log10Csat in Table S1?

Revised Figure 2 and S4, and added the values of $log_{10}C_{sat}$ (298 K) and $log_{10}C_{sat}$ (T) in Table S1 as suggested.

9. Figures S4 and S8: It could be helpful to correlate log(Csat) and sumTmax also with the campaign averages of the OA mass concentrations. For ideal mixtures, those determine the critical threshold which "vapor pressures" are sufficient for a compound to remain in the condensed phase. And that should be related to the bulk apparent volatility of OOA particles. The data look like a correlation and if so, that should be mentioned in the main manuscript.

We agree with the reviewer that OA mass concentrations should be related to the bulk apparent volatility of OOA particles. And we did this correlation as well in Figure S5 and S9. However, the correlations of campaign-average mass weighted $log_{10}C_{sat}$ (T) values and sumT$_{max}$ values vs. OA mass measured by AMS/ACSM are weak (0.3 and 0.1, respectively) and therefore we didn't discuss more in the main manuscript. Clarifications are added to the figure captions: "Org and PM$_{2.5}$ data were total non-refractory mass concentration from a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne Research Inc.) or an aerosol chemical speciation monitor (ACSM, Aerodyne Research Inc.)."

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