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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling the formation and properties of traditional and non-traditional secondary organic aerosol: problem formulation and application to aircraft exhaust" *by* S. H. Jathar et al.

Anonymous Referee #3

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Jathar et al. present a re-formulated approach to modeling SOA formation from IVOC and SVOC emissions (so called Non-Traditional SOA or NT-SOA). They use data obtained during two aircraft engine emissions experiments in which emissions were sampled into a portable smog chamber, oxidized via reaction with OH and analyzed – accounting for the SOA formed through traditional VOC precursors and unspeciated I/SVOC precursors (by subtraction). The collected data provides the empirical basis for parameterizing NT-SOA formation using a hybrid model approach. The aim of the manuscript is to describe and justify the NT-SOA parameterization. Ultimately, the au-

thors hope that their approach will provide the basis for improved model treatment of NT-SOA formation across many different combustion processes. The scientific content of the manuscript represents an important contribution to the atmospheric community. Prior to publication in ACP, the authors should address the following points.

Points

The authors should seriously consider NOT using the terminology POC (Primary Organic Carbon) to describe I/SVOC exclusively. There are non-I/SVOC primary organic carbon emissions and as such the current terminology does not make sense and will likely lead to confusion/frustration in the wider atmospheric chemistry community.

The authors should provide a reference at: P 9947, L25 "Instead it is classified as an unresolved complex mixture (UCM) that is thought to be a complex mixture of branched and cyclic alkanes."

And also here: P 9948, L23 "but IVOC UCM is thought to be mainly composed of branched alkanes."

The authors need to elaborate on how the gas phase precursors and condensed phase oxidation products can have the same residence times in the atmosphere. This seems incongruous. p.9949 L5: first few generations of oxidation or about 5–10 % of the time spent by precursors and their products in the atmosphere.

Given these limitations, the authors state that they will focus only on the first generation of oxidation quantifying this further in section 3.2 stating: "The secondary PM data were measured after three to four hours of oxidation inside the smog chamber at typical atmospheric OH concentrations." But in section 3.4 the authors state that the inferred OH concentration suggest: "The OH exposure ranges from 4 to almost 50 h of atmospheric oxidation at a typical OH concentration" Perhaps I am misunderstanding the scope of the current experiment – Nevertheless, the authors should clarify this confusion.

Section 3.1 It would be useful for those not familiar with aviation fuels if the authors provided the compositional differences between the JP8 and FT fuels in this section. Rather than providing this information in the conclusions.

On P 9957, L11 the authors state: “The emissions were diluted with clean (HEPA- and activated-carbon filter) air to achieve concentration levels in the chamber that were representative of downstream of the engine exit plane.” Perhaps the authors could provide a reference or additional information that substantiates this claim. It would be useful to know what distance downwind of the engine exit plane the chamber dilution is set to mimic. Also, given that the air surrounding airports is often influenced by the other jet and non-jet combustion sources – the ‘real’ dilution environment likely has non-zero concentrations of organic aerosol. Given this consideration, it would be useful if the author could comment on the role that this airport C_OA would play in influencing the mass yield of NT-SOA. Also, if the chamber experiments are run over 50 h equivalent OH exposure in a 4 hour time period – the authors should comment on how their results would be influenced by this non-real-world oxidation condition. But perhaps the 50 h equivalent oxidation is not relevant for the current experiment.

In Table 1 the authors define the load conditions for the CFM-56 engine with % thrust values. The same rubric should be applied to the T63 engine. This quantification is actually very important given the differences in I/SVOC emissions at low, intermediate and high engine powers. Related to this – the authors define the 7% condition in section 3.1 as “idle/taxing” Besides the spelling error (should be taxiing) this loose definition leads to further confusion later on in the manuscript. Technically speaking the ICAO specified ground idle engine operating condition is 7% load, so considering ‘idle’ vs ‘non-idle’ conditions as much of the subsequent data is presented should classify the 7% data in the idle category. In its present form, the manuscript appears to treat the 7% condition as a non-idle condition.

Figure 2: Given the range of emission ratios for the different species, the Y-axis should be displayed as a log. It is impossible to ascertain the emission ratios for many of the

species listed. Also, a legend should be provided.

P 9958 Section 3.2: I understand that the purpose of this manuscript is not to give a detailed overview of the aircraft data itself, but the authors should at least describe how the Y-axis in Figure 2 is calculated and the underlying assumptions that are inherent in that calculation. In the caption to figure 2 the authors note that the CFM-56 data is the average of three trials and the T63 data is the average of two trials. At some point in the manuscript the authors should comment on the run-to-run variability for the same operation conditions and how such variability poses challenges to the NT-SOA parameterizations.

P 9958 Section 3.3 - Please provide a reference that supports the statement that T63 cruise results in mostly oxygenated species.

Throughout the manuscript the authors offer a few different definitions of NT-SOA: “SOA formed from POC vapors is defined as non-traditional SOA (NT-SOA).“

“NT-SOA is defined as the SOA mass formed through the oxidation of unspeciated POCs.”

“SOA formed from speciated VOCs is defined as traditional SOA (T-SOA)”

-The speciated VOC precursors (Table 2) that get incorporated into the T-SOA framework include species that are NOT VOCs such as: C11,C13,C14 n-alkanes. If POCs are defined as the sum of unspeciated emissions, it is unclear how the speciated IVOCs are treated in this framework. It appears that all speciated compounds (regardless of volatility) are treated as VOCs and added to the T-SOA totals. Given the importance (as the authors clearly state in the manuscript) of unburned fuel emissions in the C11-C15 IVOC range at engine idle operating conditions, the authors MUST do a better job quantifying and classifying the SOA formed from speciated IVOCs. Getting at the heart of this confusion highlights the issues that may arise from using ‘traditional’ vs ‘non-traditional’ terminology where traditional is tied to VOC precursors only. Given the

pace with which SOA models are changing to incorporate I/SVOC treatments, perhaps the authors should consider revising this terminology.

Figure 3. An explanation of how one calculates an average SOA emission factor is warranted. The caption points out the T63 data as the average of two trails but does not mention the multiple trials for the CFM-56. Does this mean that the data shown are just one of the three CFM-56 trails? If so, why was this one run selected over the others?

Figure 4a: Plotting the data as idle and non-idle engine operating conditions is confusing. For example, in the text the authors indicate that the CFM-JP8 takeoff condition needs to be excluded from the comparison in order for $\sim$ 50% of the SOA to be predicted. The upper panel of Figure 4a shows three distinct series of data points for non-idle conditions – it is not clear to the reader if the authors are treating 7% power as idle or non idle given their prior definitions in section 3.1 as mentioned above. Since there are three distinct series of data, I suspect the authors are plotting 7%, 30%, and 85% as non idle. If the authors expect the readers to interpret which engine condition is which in Figure 4a (implied by asking us to not include the takeoff condition when approximating the fractional error) they should use different symbols for each of the different engine power conditions. If the CFM-JP8 take off condition are the data points that lie along the 1:1 line then on average, the two other non-idle conditions do not account for 50% as stated in the text. Overall, it is confusing and somewhat misleading when you ask the reader to ignore a significant amount of data displayed in a graph in order to make a point about the other data included in the graph. At the very least, the authors should provide some justification for the exclusion of the data that fall along the 1:1 line in the T-SOA model approach.

Regarding the lower panel of Figure 4a – The authors ask us to disregard the FT-Idle data which falls on the 1:1 line. It is not clear why the two Blend-Cruise experiments resulted in 2 orders of magnitude difference in OA emissions. If this is the kind of run-to-run variability that the authors are encapsulating in their averaging routine, how

representative of real-world emissions are their results and how does such variability influence the NT-SOA parameterization scheme?

P9962 L22: “since a lot of the SOA formed in those experiments is explained by T-SOA” The authors need to quantify this statement.

Why is it likely that the T63-JP8 cruise emissions are mostly oxygenated species whereas the same is not true for the CFM-56 cruise emissions?

The authors should consider the role of ambient temperature (air being drawn into the combustor inlet) on the variability of calculated yields of NT-SOA for the same engine operating conditions.

In the text, Drozd et al. (2012) should be differentiated from the other publications since it is in preparation.

Considering differences in fuel composition and combustion conditions perhaps the authors could comment on how the presented NT-SOA parameterization would differ for biomass, diesel, or gasoline combustion.

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