

## ***Interactive comment on* “The role of plume-scale processes in long-term impacts of aircraft emissions” by Thibaud M. Fritz et al.**

**Anonymous Referee #1**

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This is an interesting and important analysis that emphasizes the potential impacts of plume processing of aircraft emissions prior to their incorporation into climate models at their grid scales. This is an important point that has been made previously on a number of occasions but is often ignored and not included in analyses.

I find some parts of the modeling results to be useful, and worthy of publication. However, there are a number of issues that should be addressed before this manuscript be accepted for publication, in my opinion. The gaseous chemistry regarding ozone formation is compelling, useful, and is quoted as agreeing with prior analyses. This is useful confirmation of the importance of plume processing for ozone impacts. The contrail impacts are also of interest in how particle properties are affected by plume processing. However, the details of the way the contrail modeling has been done need

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to be qualified to a greater degree, due to assumptions that are made (mono-modal soot distribution) and implied (dependence, or lack thereof, of water uptake on particle surface composition).

1) The approach shown for volatile PM (nucleation and growth of new particles, and uptake on soot particle coatings), seems incomplete and thus potentially flawed. No specific results are shown in plots nor discussed, and it is not clear that such results impact the chemistry nor contrails results that ARE shown. This perspective will be discussed further below, where I suggest removing or discussing in a much different way. 2) The contrail modeling has made some simplifications that may impact the results that the authors claim to be important. They need to discuss in more detail how the assumptions might qualify their results and the claimed quantification of the effects that they observe. 3) There are a number of more minor wording or presentation issues that I will identify below, along with some suggestions for how they might be addressed.

1. In section 2.2.3, page 6, line 15, “Soot and ice particles can also grow by condensation of water vapor, sulfuric acid, and nitric acid . . .”. Experimental results show that the growth of particle mass in aircraft exhaust plumes is dominated by organic species (and nitric acid is not usually observed in the initial plume regions). Leaving out organic species is leaving out a primary contributor to the mass of these newly formed particles (prior to water deposition in contrail formation) as well as the coatings on soot particles. Thus, the presented microphysical approach is missing the major contribution to mass. (Yet the authors do note that volatile organics are in the exhaust, section 3.1, page 10, line 18.)

However, there are no results presented in the paper that show the importance of this microphysical processing. Neither results showing newly nucleated sulfate aerosol nor the coatings on the soot particles (and their composition) are presented in the paper. It is not clear from the material presented how the eventual uptake of water is dependent on the condensed matter due to these species. Is the later water uptake affected by the surface composition? If not, there seems to be no impact of this analysis on the

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contrail results presented later in the paper. No size distribution results are shown, so it is not clear how the soot distribution and newly nucleated particles make up the input to the downstream mature plume modeling, and how they affect the subsequent analysis.

It is worth noting that many other modeling studies suggest that the “nucleation mode” is not important for contrail processes when the soot mode is present, due to the larger size of the soot mode. Thus, there is a basis for questioning the importance of this smaller mode. The question of the compositional changes of the soot surface due to condensation seems open, but unaddressed by the present study.

Unless more information is provided, it seems that this is an incomplete analysis that has limited bearing on the problem at hand, and it does not appear that the model has a means to include the effects of this analysis on the key results presented. I suggest this part of the analysis be removed or completely re-described.

2. In a related issue, the modeling assumes (section 2.2.3, page 6, line 5) that the soot distribution is a mono-modal distribution (“a single representative particle”). While that may make sense to define a more computationally tractable problem, the microphysical modeling discussed in 1. above seems to require a binned size distribution approach (page 7 line 3), so why is it necessary to force the soot to be mono-modal? But if the response to issue 1. above is to remove the volatile particle modeling, then perhaps the mono-modal soot distribution may be justifiable to simplify the computations.

However, if the approach is to accept a more limited modeling approach, based on a mono-modal soot distribution as has been done before (as referenced by the authors), then another separate question arises. The results show important differences due to differences in the fate of large particles versus small particles in the later plume processing (section 3.5.1, line 12 et seq.). If the initial soot distribution is mono-modal, the contrail particles will also be mono-modal for those particles that have had the same history (i.e., in the same ring). There needs to be more discussion of how the history

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of particles might generate a size distribution that differs from the initial mono-modal soot size distribution, if this is, indeed, what generates a polydisperse contrail particle size distribution.

3. Presentation issues and typos: a. In the introduction (page 2, line 29 - 30), “aviation is . . . the only direct, significant source . . .”, what about rockets? May not be as large, but rockets may still be significant.

b. Also, in the introduction: this is not meant to be a review article, but it might be worth mentioning that the importance of plume processing has a history that goes back to CIAP (CIAP monograph 3, 1975, DOT-TST-75-53, chapter 2 and references therein) and, {especially since the manuscript is a NASA sponsored study}, to NASA (Atmospheric Effect of Aviation: First Report of the Subsonic Assessment Project, 1996, NASA Ref. Pub. 1385, chapter 4 and references therein)

c. Figure 1. As a schematic, this figure seems to address only the 2.3 mature plume modeling part. There is an inset box that discusses the plume box model processing, but there is no schematic representation of the box model in the artwork. And there is no equivalent inset box that describes the discretized rings in the figure.

If the box model inset box were removed from the existing figure, I would suggest this figure would sit better in section 2.3, where the mature plume modeling is discussed. It has little schematic value for the box model as drawn so doesn't provide much benefit as placed in section 1. If, on the other hand, the figure was adjusted so that there were schematic aspects and inset boxes for both parts of the model, then perhaps a redrawn version might have reason to remain in section 1.

d. Section 2.1, page 4, line 13. “The output of this box model . . .” This sentence is confusing. The antecedent to “this box model” doesn't exist, since the model hasn't been mentioned yet, and is described in the subsequent section.

The preceding paragraph describes the physical phenomena to be addressed, but

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there is no mention of the box model that will be used. One solution would be to briefly mention how a box model (to be described in detail later) will be formulated to capture the elements described.

Another solution would be to drop that sentence and pick it up later after the two models are discussed. If this approach is taken, then the material in these two paragraphs (last two paragraphs of section 2.1) would just be discussing the physical phenomena in the two regimes and leave the box and ring models' discussion for the later sections. (If this approach is taken, the title of 2.1 might need to be adjusted.)

As written, the sentence is confusing, referencing models that haven't been introduced yet.

e. Section 2.2.2, page 5, line 22, Tremmel, (and by Lukachko et al., 1998 JGR 103, and in 2008, J. Eng. Gas Turbines and Power, 130, 2008) found that the conversion of S(IV) to S(VI) occurred primarily in the engine's turbine, and not in the plume to a significant degree. Later processing in the atmosphere happens also, but at time-scales much longer than the initial plume being addressed by this study.

f. Section 2.4.1, page 9, line 21. "equipped with GENx engines". In what sense is this engine represented in the model? In section 3.5.3, page 19, line 8, a soot emission index (EI) is given as 0.06 g/gfuel. (I assume this is a typo, and it is meant to be 0.06 g/kgfuel, or 60 mg/kgfuel). This seems very high for the GENx engine, especially at cruise at altitude. This (even after correcting for the typo!) is 1.5 times the {high} value used in the 1999 IPCC report of 0.04 g/ kgfuel. And where was the EI soot data obtained?

In addition, is the NOx EI chosen to be representative of the GENx from the ICAO Databank?

g. Section 3.5.1, page 16, line 1. Supersaturations of 102% to 108% are quoted, but is this respect to water or ice? (And is 108% observed in the natural atmosphere?)

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