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Interactive comment on "On the effects of hydrocarbon and sulphur-containing compounds on the CCN activation of combustion particles" *by* A. Petzold et al.

Anonymous Referee #2

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General Comment:

The paper by Petzold et al. presents results from comprehensive investigations of gaseous and particulate emissions from a gas turbine engine which were carried out within the European project PartEmis. Samples were taken at the nozzle exit of an aircraft test engine which was operated in a simulation test run under so-called old and modern engine conditions and with fuels of different sulfur content. Therefore, the results discussed in the present paper may, within experimental uncertainty and limitations, be representative for engine emissions under cruise conditions. The paper

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offers, based on the application of an impressive set of well advanced and established methodologies, a broad and very interesting spectrum of results especially on the very important question, how the CCN activation of combustion particles is affected by surface coating or internal mixing with sulphuric acid and organic carbon. On the other hand, the role of CCN activation may critically depend on the temperature at which water saturation is exceeded. At temperatures below about $-35°$C ice nucleation activity will become more important. This should be considered e.g. in the introduction and the discussion of contrail effects on page 2613.

It appears to me that part of the results presented in this manuscript have already been subject of previous publications e.g. by Petzold et al., GRL, 2003, Gysel et al., GRL 2003, Hitzenberger et al., GRL, 2003, and Nyeki et al, GRL, 2004. One of the major new aspects of the present manuscript seems to be the analysis of the organic, elemental, and total carbon content of the exhaust aerosol. If this is the case it should be more clearly pointed out somewhere in the introduction and the discussion of the results should be more specific to the new aspects of carbon content analysis. In this context, I recommend major revision and extension of the chapters which describe the off-line carbon analysis methods (see major and minor comments and questions), especially because there is given only one reference to EGA analysis (Puxbaum, 1979) which may not be directly accessible to all readers.

The scientific questions addressed in this manuscript are well in the scope of ACP. However, I recommend major revision for publication in ACP.

Major review points:

(1) In line 28 of the introduction you state that CCN (cloud condensation nuclei) activation is the "key process with respect to any indirect effects of combustion-related particles on the global climate". This may be true for warm and mixed-phase cloud conditions but totally disregards that aircraft-emitted particles mainly may act as ice nuclei rather than CCN at heights (and temperatures) where most aircraft engine par-

ticles are emitted to the atmosphere.

(2) In the first paragraph of page 2602 you mention the advantage of the present experiments in comparison to laboratory flame burners. This suggests, if I understand right, that an aircraft engine is an ideal model for all other combustion engine sources to the atmospheres. I doubt this is the case, for the same reason as a laboratory burner is an ideal source for all laboratory investigations. I strongly recommend to be more careful with arguments towards a "real combustion engine" and against "laboratory burners". Both approaches have advantage and disadvantage.

(3) P.2603, l.11: In Figure 1 there is no a or b. The caption refers to upper panel. The sketch in the upper panel is completely unclear to me. Please explain in more detail: Do you show here a cross section of the nozzle? What is the area and direction of gas flow? Where are the sample points actually located? What mean the angles given in the drawing? Were the sampling tubes directed against the gas flow or in any other orientation?

(4) Concerning the cooling profile along the sampling line discussed on page 2604, you mention that the sample was first cooled and then kept at a temperature of about $150°$C for the undiluted part. You then mention possible aerosol losses due to diffusion and gravitational settling. What about thermophoretic effects? It is not sufficient here to just mention possible loss processes. Can you quantify or estimate the sampling artifacts? If you concern about losses in the undiluted sample line at $150°$C what about sampling artifacts during cooling from $1200 \pm 200K$ to $150°$C? This should also be specified, if possible.

(5) As mentioned above, section 2.3 about the off-line chemical analysis needs major revision and extension. For instance it may not be obvious to the reader why the Puxbaum and Cachier methods use different temperature thresholds to determine elemental carbon (EC). Are the systematic deviations shown in Figure 2 reflecting this difference? Do you have an explanation for the systematic trend with FSC which is indi-

cated in the correlation data shown in Figure 2? The systematic difference between the two methods may be well-known to the authors but not to the reader. Please include a brief explanation, if any, especially if it is important for the conclusion drawn from the respective data. If there is a significant fraction of semi-volatile OC in the EGA analysis I would not be surprised about a systematic deviation of the EGA EC content to the results from the Cachier method. What about using the same threshold temperature for determining EC in both methods? Nothing is said about possible contribution of scattering to the transmittance measured in the combined thermo-optical method. I'm also missing any statement about the accuracy of the carbon analysis methods. To which of the two analysis methods do you refer in line 22 of page 2606? I would be confused if you refer to both because in Figure 2 you showed significant systematic deviations. There is a mismatch between good agreement mentioned in the first paragraph of section 3.1 and the systematic errors and inconsistencies addressed above. The representation of Figure 4 is of poor quality. Could you e.g. mark the identification of the main peaks? Why don't you convert the runtime on the horizontal axis to a temperature scale? Can you give at least one example of the transmittance data together with the thermal carbon signal?

(6) P.2607, l.20 ff: If nothing is known about particle shape which normally is very irregular for combustion particles the estimates made here can in fact be extremely uncertain. Therefore I suggest to skip this paragraph together with Figure 6.

(7) P.2608, l.13 ff: How did you convert mobility equivalent diameter measured by the DMA to aerodynamic diameter? Obviously you scaled the DMA volume and surface distribution to match the measured mass distributions. This must be mentioned here. Why don't you show a DMA mass distribution in the left panel or give an idea of the agreement between the two methods of mass determination? How did you determine the surface coverage here? If you can give only a max. (maximum?) value of 0.1 monolayers the coverage seem to be below detection limit already for the high FSC case.

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(8) The sulfuric acid surface coverage seem to depend significantly on the sampling temperature and the temperature of the DMA systems. Can you give an idea whether the data shown in Figure 8 are representative for particles emitted under cruise conditions? What kind of sheeth air did you use in the DMA systems? Can you exclude partial evaporation of the sulfuric acid coating inside the DMA, e.g. upon dilution with dry sheeth air?

(9) Part of the conclusions on page 2617, l.14 ff is only correct if complete internal mixing is assumed. Please indicate what you know or assume about the mixing state, if not yet done (I may have missed a hint on that).

Minor comments and questions:

Title: Please check if the title is appropriate. What hydrocarbon compounds do you have in mind here? I did not find this explained in the manuscript.

p.2600, l.5: . . . suite of aerosol, gas, and chemi-ion measurements

p.2602, l.3: . . . more general conclusions are drawn from this study?

p.2603, l.23: This feature made possible additional investigations . . .

p.2604, l.21: Do you mean on-line here? Or did you really measure locally inside the exhaust flow?

p.2605, l.5: How do you define the threshold diameter for nucleation particles? The size distributions shown in Figure 5 do not indicate any second mode when approaching 10 nm.

p.2605, l.10: How can the present investigation achieve this adaption if the CCN activation is measured only at one saturation ratio? I think through modeling, which may be mentioned here.

p.2607, l.7: The intensities of semi- and nonvolatile OC peaks vary . . . (Is there a reason for that?)

p.2607, l.10: Please specify if the size distributions shown in Figure 5 are corrected for sampling dilution (I guess they are).

p.2615, l.13: . . . is of the order