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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Perturbation of the European free troposphere aerosol by North American forest fire plumes during the ICARTT-ITOP Experiment in summer 2004" *by* A. Petzold et al.

Anonymous Referee #1

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The paper is a valuable compilation of data and model results. The topic is interesting and new. The text is generally very well written. Campaigns like these greatly benefit from joint evaluation as done here. It should thus be published in ACP.

I propose some changes to make the paper more coherent. The documentation of the data seems to me inadequate and it would be necessary to add information, so that others can repeat the model experiments.

Detailed comments: P4932: " BCe values may be lowered by factor of 1.25" - seems

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Interactive Discussion

Discussion Paper

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to me odd English.

P4936: “FLEXPART suggests that almost pure forest fire aerosol was sampled”: I think this is exaggerating the capability of a trajectory model without wet deposition, aerosol dynamics and mass budgets closed. The footprint PES in figure 2b shows considerable passing right over the east coast of North America. High pollution levels in these areas, even closer to the receptor site may have contributed to the total aerosol load. To be well understood: I agree: The FLEXPART results support nicely the hypothesis that long range transported smoke from North America has been sampled.

Figure 1, 2 and table 1: Why is there just one minute picked in figure 2? In general I have difficulties to locate the measurements used in the analysis in time and space. Figure one is almost useless to identify the flight portions analysed in this paper. Table 1 is very general and could rather state explicitly when and where smoke encounters where measured. Where are the vertical profiles flown?

P4936 , Figure 3: This figure is mysterious to me. To obtain a map of BC column load one would need to integrate the emission and dispersion over a certain time frame. When are the CO tracers emitted, when is the emission ending? Which emission areas are activated for which period to get to this column load field? Is it the FLEXPART forward model?

P4936: spelling: “ĚHohenpeissenberg. In?? can be concluded”

P4937: suggested change “Such an absence of the NUC mode is HERE identified as oneĚ “

Figure 4: Isn’t there a contradiction in the size distribution vertical profile between the 22 and 30 July? Why is the aerosol in the supposed smoke layer on the 22nd of July peaking at smaller diameters than that of the 30th of July, although the plume of the 22nd is supposedly “older”? Wouldn’t we expect an aged aerosol (that of the 22nd) has a larger mode diameter? I don’t think there is a discussion on this in the paper. I think

this ageing hypothesis should be more clear and related to what was really observed. Instead the authors argue e.g. in the abstract “no steady state of the size distribution was observed”. I think this is not really thought through with respect to the observations presented in this paper.

Figure 4: excess CO is introduced in text and figure without reference to the appendix, nor is it explained shortly. Some short notation what it is, would be helpful.

P4939: “For the determination of the ratio BCe/Nnonvol the 5-percentile value of the BC concentration at Jungfraujoch is a more robust valueĚ” If we believe that the BCe fraction at background troposphere values is essentially constant within limits, then I do not think that the authors calculation of this fraction is totally correct. They combine the 10% and 90% percentile of number concentrations with the 5% percentile BCe concentration. I request to add the 10%, median and 90% percentile BCe concentration values measured at Jungfraujoch into the discussion and calculation of the error bar of the BCe/Nnonvol ratio.

P4940: “Significant differences between smoke plume profiles and non-smoke plume profiles occurĚ”. Significance implies some statistical testing has been done. I don’t see that.

P4941: “We set sigma-ap,min= 1sMm-1 (see appendix)”: I found 0.1 sMm-1 as detection limit. Probably a typing error, otherwise not consistent.

Figure 7a : Wouldn’t it be more fair for the comparison to plot the range of sigma-ap into which possibly the real values may have fallen (a bar going up to the detection limit instead of a point at the detection limit value). The model concentrations might be still too high!

P4941: “of similar magnitude as in the urban outflow from Paris”: Here a reference or citation is missing!

Figure 8: this figure is illisible, too small.

Figure 8 top right: Why is there a gap in the middle of the graph?

P4943: Why do the authors use unit density as reference for the calculation of PM25? I think 1400 kg m-3 would be more appropriate for POM particles. I believe the calculations become more realistic. Assuming an error with a most realistic density of 1400 would then give a realistic error bar, which only can be compared to content estimates in emissions;

P4944: "However, plume dilution and uncertainties in PM add a high level of uncertainty to these numbers (on BC mass content after transport)."See comment above. Given the error bar on the content estimate: Can the authors really conclude here and elsewhere in the paper, that the transported smoke is "similar to fresh plume conditions".. Besides - why should plume dilution change the BC mass content of smoke particles?

P4945: (1) the coarse mode identified in aged size distributions is missing in ITOP observations": Which coarse mode? Not discussed before! Which coarse mode is used in 'climate models' for smoke, in which climate model ?

P4945: "plumes of various age were investigated during one field study. This permits a good comparability of observations." I can only see that two particle populations are discussed with respect to age, that of the 22nd and that of the 30th of July. 'Various' is exaggerated, or do the authors have more information on plume age in other cases. Then size parameters should be plotted as a function of plume age.

P4945: "from the nucleaction mode to the coarse mode"; Was the inlet system really capable to measure the coarse mode? What do the authors mean by coarse mode?

P4947: Is Turquety specifically providing an estimate for boreal forest fires?

P4947: "plume dilution is an issue": What kind of issue? What does this plume dilution discussion add to the principal findings of the paper? Should be refocused.

P4948: "values used so far in global climate models. Particles are of larger sizes but

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7, S2263–S2268, 2007

Interactive Comment

with lower number concentrations”: : Citation and reference needed, otherwise omit.

P4948: “Our data extent” typo - I guess etend is meant.

P4948: “As a hypothesis, the narrowing of the size distributionĚ” This discussion is not very convincing. Why should the size distribution narrow down with age by gas phase deposition? Why should it just happen on smaller sized particles? Any references ?

P4948: The CCN discussion can be omitted. Very speculative and general.

P4949: “ĚBCe/delta CO are of the same magnitude as for fresh emissions” Do the authors mean that the BC mass content and BC/CO ratio are within a factor of 10 (= one magnitude) of the emission values? Would that be an interesting finding? It would be helpful to quantify this estimate to put it more valuable relation to the export efficiciencies from Park et al.

P4949: “ and by an entirely internal mixture even for Aitken mode depleted particles”: Where is the proof of “entirely” mixture in this paper?

Table 6: An explanation of the symbol tau is missing in the header.

Table 6: I find the citation of Dentener 2006 in the table as reference to the smoke distribution assumptions very misleading. I can not find such data in that paper, nor are coarse particle modes suggested to be applied for smoke particles in that paper.

P4949: “ The median diameter of the accumulation mode growths with plume age while the size distribution becomes narrower”: This is interesting, but I don’t think that this is really shown conclusively in this paper. I guess it refers to the two cases of 22nd and 30th of July. First, the finding is based on very few cases (=2) along the ageing time axis. Second, we don’t know whether the plume of the 30th is from a different type of fire, or has a different impact from other sources. Third, there is quite some variability of the size distribution shown eg in Figure 4. This figure also indicates rather larger particles for the 30th than on the 22nd , in the smoke layer , which is in contradiction to the authors reasoning of the evolution of smoke size distributions. Also the error and the variability of the size distribution data in table 6 is not adequately discussed. A more careful statement in abstract and discussion on these size evolution issues would do the paper good.

P4949: “ after more than one weekĚ, no steady state of the size distribution is observed”: Omit. An observation of the steady state (or non-steady state) of the distribution would require a langrangian observation of changes (or not) in that distribution. That’s not done. Also the statement implies, that ‘after more than’ one week there are changes happening. The observations don’t really go beyond a week.

Please change abstract according to any changes in the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 4925, 2007.