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Interactive comment on "Wildfire smoke in the Siberian Arctic in summer: source characterization and plume evolution from airborne measurements" *by* J.-D. Paris et al.

Anonymous Referee #2

Received and published: 21 October 2009

This paper describes the results of YAK-AEROSIB campaign in Siberia. Airborne measurements of CO, CO2, O3, EBC and aerosol numbers were performed during multiple flights. Biomass burning plumes from areas around Yakutsk were encountered and also modeled using the transport model FLEXPART. Emission factors for those Siberian fires and aerosol lifetimes in the plumes are determined. Conducting an aircraft campaign in Siberia is certainly not an easy task and the data resulting from this campaign are certainly unique and the results presented here are clearly worth publishing in ACP. The paper is generally well written and organized. I have two major and a few smaller comments that need be addressed before this paper can be published.

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Main Comments:

1) My main concern with this paper is the calculation of the emission ratio (ER) and emission factor (EF) of CO, which has a large uncertainty that is not clearly enough acknowledged in the paper. Usually ERs are determined by calculating the slope of a scatter plot, in this case CO versus CO2. In Figure 4 CO2 data are missing at the end of the flight, which makes the analysis presented her more difficult and uncertain, but nevertheless, it looks like it is still possible to generate a statistically significant slope from a scatter plot in the biomass burning plumes V and VI. Is there a reason, why this was not done? If not, how does the slope compare to the ERs used here? The values for the ERs given in Table 1 are very uncertain, mainly due to the uncertainty in the background of CO2. Given the background uncertainty of about 2 ppm deltaCO2 in plume VI is 1.1-5.1 ppm, which results in a very large uncertainty in the ER of CO. The slope of the scatter plot is not dependent on the background mixing ratios and is therefore much more certain. Given this large uncertainty in the ER of CO and the fact that the EF of CO2 is taken from the literature, the error estimate of the CO EF seems very optimistic. The following calculation of the total CO emissions from the Siberian fires relies on this uncertain number, which was determined from only two plumes from the same fire. This large uncertainty should be more clearly acknowledged in this paragraph. Looking at the total CO emissions might still be useful to show the importance of the Siberian fires for the northern hemisphere.

2) The determination of the background mixing ratios is one of the important parts of the presented analysis and I think it should be presented in a separate chapter of the paper. Especially the CO2 background values are used to calculate delta CO2 in Table 1, but only later in the paper it is shown, how the background is estimated. The authors estimate background values with altitude profiles and I would suggest adding such profiles (average and clean air) to this chapter, which would also help the first part of the Results section.

Other Comments:

Page 18208: The above-mentioned altitude profiles will greatly improve the discussion on the stratospheric influence on ozone and the CO2 gradient to the surface.

Page 18208 line13: Higher ozone in the free troposphere can also be caused by stratospheric ozone and not only by surface deposition. This should be mentioned in the text.

Throughout the paper: Please use the term mixing ratio instead of concentration with the units ppm or ppb.

Page 18208 line 27: What is the rationale for using emissions within the last 10 days of transport for anthropogenic CO? It seems to me that BB plumes were followed further back in time as shown in Figure 10. Anthropogenic sources should be made consistent with BB sources and the actual transport times should be mentioned in the text.

Page 18209 line 8: What is the grid size for the BB emission inventory used in FLEX-PART? The model resolution is likely also a source of uncertainty close to the fire locations.

Page 18209 line 13-18: Looking at Figure 5 it seems the FLEXPART CO altitude profile has steps at about 1km resolution close to the fire up to about 5 km. What is the cause for that and what is the vertical model resolution? The model distributes the fire emissions homogeneously throughout the lowest 3 km. Is the fire CO rapidly lifted to 5km? Looking at Figure 5 the vertical distribution of the fire CO is likely the main uncertainty in the model calculation.

Page 18209 line29: The thick cloud layer over the hot spot area certainly will influence the MODIS fire detection. What is the influence of this on the FLEXPART BB emission inventory?

Page 18210 line 13: What causes the low ozone in this plume? In the conclusions, possible ozone destruction is mentioned and should be discussed here as well.

Page 18212 line 13: Please explain the terms C[CO2] to C[PC] in the text.

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Page 18213 line 20: Please explain in more detail how the EF was calculated. What plumes were used (average of the two) and what CO2 EF.

Page 18214 line 3-13: It should be easily possible to calculate the total BB CO emissions for the Siberian fires from the FLEXPART emission inventory. How does that result compare to the calculation presented here?

Page 18214 line 13: Also here give more detail on how the ER is calculated.

Figure 9 is not mentioned in the text and does not add anything to the paper and I would suggest removing the Figure.

Page 18215 line 15: Most of the BB particles and therefore most of the EBC mass will be in the size range of the ultrafine particle number concentration. A short discussion should be added explaining how the EBC mass can decrease faster the aerosol number concentration even though the loss processes are mainly wet and dry deposition for both.

Table 1: Please indicate the uncertainties for all the values given.

Figure 1: The red and blue dots are very small and hard to distinguish.

Figure 5, 7, and 8: Can the plume numbers also be marked in these Figures?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18201, 2009.