

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.

J.-D. Paris et al.

jean-daniel.paris@lsce.ipsl.fr

Received and published: 24 November 2009

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 here 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

C7459

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 Δ CO₂ 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.

- Response: The reviewer comments on the method for determination of ER and EF. It is right that ER_{CO} determination from a regression slope has less uncertainty due to background estimation. Initially we tried two methods (i.e. based on maximum enhancement, and on regression slope) and found them to give closely similar results for the plumes reported here (typically, within 1 std dev, with significant correlation coefficient as a basis for the regression slope; $r^2 = 0.66$, $p < 10^{-6}$), but not necessarily for all other plumes close to the source (during other flights, not shown in detail in the paper). For plumes in other flights correlation coefficients were not so significant. However we changed that and report now in the revised ms values of EF_{CO} based on the linear regression slope. We added a note on that in the revised ms between Eq. 3 and Eq. 4. Regarding acknowledging the uncertainty, we added a sentence in section 3.3 discussing the EF: “The uncertainty reported here should be considered with caution since we did not measure EF_{CO2} directly”. Also, we have softened the wording of our conclusions through the text according to the reviewer’s suggestion

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 Δ CO₂ in

C7460

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.

- Response: Following the reviewer's suggestion we gathered the scattered elements on background determination in a new section, making it clearer for the reader to locate this information. We did not however add a figure with the vertical background of CO₂ because there would be only two values to show: the well-mixed boundary layer and the free troposphere. We report on the BL value that is relevant to this study.

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.

- 1. This point was addressed above in the response to the General comments

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.

- 2. A role of stratospheric O₃ is possible and now we have acknowledged this in the revised ms. We also additionally discussed earlier results (Paris et al., 2009b) where we have shown that in other summer campaigns above Siberia we found little correlation between lower tropospheric O₃ and stratospheric input.

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

- 3. We change the term of concentration for mixing ratio where necessary through the paper

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

C7461

with BB sources and the actual transport times should be mentioned in the text.

- 4. The backward modelling was actually carried out over 20 days and not over 10 days as erroneously mentioned in the text. We thank the reviewer for spotting this mistake, which is now corrected in the revised ms. We consistently used 20 days for both forward and backward simulations and for all tracers, throughout the paper.

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

- 5. For the forward simulations, the tracer was released at the exact location of each fire detection, so the releases are independent of a particular grid. However, in the case of the backward simulations, the emission sensitivity was calculated with a resolution of 0.5° and all fire detections were aggregated at that resolution. The reviewer is right that in the vicinity of the fires, this limitation is a possible cause for model errors. However, we also found that the forward and backward calculations were quite consistent, so this does not seem to be the major reason for model errors.

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.

- 6. For the forward simulations, of which results are displayed in Fig. 5, a 1-km resolution was used for the FLEXPART output, so the step changes are caused by the relatively coarse vertical resolution of the model output grid. However, notice that the transport calculations are independent of this grid. Thus, while the output does not resolve vertical structures below 1 km, the overall effect of smaller-scale processes would be reflected in the output. A note on that has been added to the revised ms in

C7462

the caption of Fig. 5. For the transport in the lower atmosphere, the vertical resolution of the ECMWF data is probably most important, as it determines the accuracy at which for instance boundary layer heights can be determined. The vertical resolution of the ECMWF data is variable, from a few 10 meters at low altitudes to a few hundred meters in the lower free troposphere.

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?

- 7. Biomass burning emissions are calculated directly at the fire locations observed by MODIS and, therefore, depend on the satellite's viewing conditions. Clouds definitely affect viewing conditions and, therefore, fires underneath clouds may largely be missing. There is no obvious way to deal with missing detections and our only measure was to assume that fires are burning for 48 hours, which introduces some temporal smoothing to the fire emissions. If a fire is not detected on one day but either on the following or preceding one, it would be reflected in our inventory but would have a reduced source strength on both days.

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.

- 8. p18210L13 low O₃ and the anticorrelation with CO in plume 1 is now discussed in the revised ms. Page 18212 line 13: Please explain the terms C[CO₂] to C[PC] in the text.

- 9. p18212L13 The terms C(. . .) are explained in the revised ms

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. - 10. ERco calculation details are now discussed in the revised ms. we chose the method of maximum enhancement for EBC since the long integration time of the instrument did not allow computing coherent

C7463

ent regression slope. EFCO was estimated using regression slope in the two plumes together.

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?

- 11. We calculated the total CO emission derived with the algorithm used by FLEXPART for the region 30-180 deg E and north of 40 deg N. According to this algorithm, a total of 27 million tons of CO were emitted for fire detections over forested areas, and another 30 million tons of CO were emitted by fires detected over other land use types, for the period March to October 2008. This is now mentioned in section 3.1 in the revised ms.

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

- 12. ERbc calculation is done in the same way as for CO. This is acknowledged in the revised ms.

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

- 13. Fig 9 has been removed from the revised ms

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 than the aerosol number concentration even though the loss processes are mainly wet and dry deposition for both.

- 14. BC decrease slightly faster than total aerosol mass concentration (5.5 vs. 5.1 d). The mismatch between size-dependent sensitivity of the two measurement techniques and the fact that we compare decrease in number vs decrease in mass concentration can explain a large part of this difference. In fact, the ratio of DN3-200 to DBC remains

C7464

approximately constant ($\sim 3000 \text{ cm}^{-3} / (\mu\text{g m}^{-3})$) for all plumes excepted Plumes 1 and 2, where the low excess above background makes this ratio uncertain.

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

- 15. Table 1 uncertainties added

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

- 16. Figure 1 These dots are small area polygons; the size of dots and of the whole panel has been increased in the revised ms. Figure 5, 7, and 8: Can the plume numbers also be marked in these Figures?

- 17. Fig. 5,7,8: plume numbers have been added in the plots

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

C7465