Reply to the reviewer comments on manuscript acp-2013-687: "The Pagami Creek smoke plume after long-range transport to the upper troposphere over Europe – aerosol properties and black carbon mixing state"

by Dahlkötter, F., M. Gysel, D. Sauer, A. Minikin, R. Baumann, P. Seifert, A. Ansmann, M. Fromm, C. Voigt and B. Weinzierl

We would like to thank the two reviewers for their useful comments, which help to improve the scientific quality of this paper.

Following the general request to shorten the manuscript and to remove redundancies, we restructured the manuscript:

- All results describing the Pagami Creek plume including the microphysical and optical properties of the smoke particles, the coating thickness of individual rBC particles, the horizontal and vertical plume dimensions, the ground-based lidar measurements and aerosol classification, as well as source apportionment with HYSPLIT are now presented in section 3 (“Results”).
- Section 4 (“Discussion”) has been shortened as suggested. Instead of having 6 subsections, we condensed the discussion into 4 subsections starting with a comparison of the Pagami Creek plume results with other measurements in elevated aerosol layers during the CONCERT and with the literature.
- The subsection on the morphology and disintegration of the black carbon particles has been extended to give more information on this observation as requested.

We think that the new structure removed redundancies and improves the clarity of the presentation and the readability of the manuscript.

Below we reply to the individual reviewer comments. We highlight our replies to the reviewer’s questions in blue.

Reply to review #1 by Gavin McMeeking

Q: (…) The manuscript is a little long and has a large number of tables and figures and I believe could be condensed to focus more on the key findings related to the BC observations. The number distribution results could be moved to supplementary material, as they are not really central to the main thrust of the paper, as could some of the details on the SP2 analysis. The comparisons of rBC mass concentrations presented in Section 5.2, which compares the results to previously reported biomass burning measurements, are not particularly useful given the extreme differences in sampling locations and the plume age. If possible, a comparison of rBC/CO ratios would be more helpful.

A: Structure and length of the manuscript: Please see above.

BC/CO ratios: Unfortunately, no CO measurements are available for the CONCERT 2011 campaign. We looked at the IAGOS data to see whether there are CO measurements available and found one measurement of 198 ppbv at 9.5 km altitude recorded at 13:31:08 on 16 September 2011. However, this measurement was taken at 51.25N, 17.2 E which is about 150 km further to the east and slightly further to the south than our measurements in the plume. Given the observed inhomogeneity in the plume, we feel that this is not convincingly close enough to the DLR data to include in our paper.

Q: It would also be useful to know if any of the other previously reported emission measurements featured PyroCb, as this could affect the mixing state comparisons made in later sections.

A: Unfortunately, there is only very little information available for emissions of PyroCbs.

Q: The discussion of the disintegrating BC-containing particles could be expanded somewhat. A comparison is made between a UTLS background sample and the forest fire plume, but results for the polluted boundary layer segments could be added. Was there any correlation between coating thickness and disintegrating particle number fraction in the background or boundary layer regions? This might help determine if the disintegrating particle phenomenon is unique to biomass burning emissions or a feature of thickly-coated particles that are just more common in smoke plumes, as discussed. A figure or two would also be helpful, perhaps one showing the fraction of disintegrating particles versus total particle and BC core size in the smoke plumes.

A: We extended the section on the disintegration of BC-containing particles as requested. We added two additional figures. In Figure 13a, we show the relation of the scattering cross-section at the leading-edge and at the trailing-edge of the laser beam for BC-containing particles for the Pagami Creek fire plume together with the detection limit of the LSD and the range where data points are attributable to coincidence of BC-free and BC-containing particles in the laser beam. In Figure 13b, we plot the fraction of disintegrating particles to all rBC-containing particles as function of their coating thickness for a sequence measured in the Pagami Creek fire plume. Figure 13b shows that there is a correlation between coating thickness and disintegration particle number fraction.

Q: 28754, 27: Cappa et al. (2012) also compared ambient measurements to laboratory observations of coating enhancements.

A: We changed the text and included Cappa et al.

Q: 28762, 7: “spare to mention this mode” please re-phrase.

A: Done.

Q: 28762, 9: capitalize STP or write out

A: We capitalized it (“Stp”) at the beginning of this sentence.

Q: 28766, 19-23: Please clarify that the added uncertainty due to fresh/small rBC particles in the boundary layer arises from a mode of particles not captured by the log-normal fitting. I assume if the mode shifted you would capture the change in un-measured fraction fairly well.

A: Correct. We extended the text to clarify this as requested.

Q: 28767, 25: suggest combining this material with Section 3.2

A: Done.

Q: 28771, 17: The paragraph describing the lidar depolarization ratios is a bit off topic compared to the rest of the paper and could be omitted to shorten the paper a bit.

A: We shortened the lidar analyses which were originally part of the discussion section and moved them into the new section 3.2 (“Estimates on the dimension and classification of the observed aerosol layer”). Here, we find it important to have this information since the lidar depolarization

ratios help to attribute the aerosol layer to the specific source (biomass burning aerosol with some large particles).

Q: 28777, 15: Suggest noting that our analysis in Akagi et al. (2012) relied on the time delay approach to classify coatings. Also remove “Anyway” in line 22.

A: Done.

Q: 28780, 9-13: I don’t quite follow this. Wouldn’t you also see a stabilization of the scattering cross-section from coincidence of a BC and non-BC containing particle? The BC particle vaporizes, so the scattering signal degrades, but then flattens as the second, non-BC particle continues transiting through the laser?

A: Thanks for raising this very good question which shows the complexity of these mechanisms. The position dependent correction for the laser beam intensity would fail for coincident particles that are, while simultaneously being in the laser beam, not exactly at the same position. This would cause an apparent time dependence of the inferred leading and/or trailing edge scattering cross section for coincident particles. We implemented this explanation to the text.

Q: 28781, 5: It becomes obvious in subsequent sections, but it should be explicitly stated here that the reported value of 50% is for in-plume measurements.

A: Done.

Reply to the review #2 by an anonymous reviewer

Q: (…) The presented content and methods appear sound and proof; however, the manuscript is largely excessive with many repetitions, unneeded information and literature discussions that have to be bundled. The authors should consequently shorten their manuscript and really focus on their key findings. This also includes the number of tables and figures and the unusual high number of used footnotes, which should in general only be used sparsely in scientific articles.

The structure (and length) of the article can e.g. be improved by combining the section on the results (Sect. 3) with the discussion part (Sect. 5), which would also avoid many repetitions and ease the reading.

A: Structure and length of the manuscript: Please see above.

Footnootes: We have integrated most of the footnotes into the main text.

Q: The finding of the disintegrating rBC particles is appealing, however, I miss a real quantification. An additional figure could be added here.

A: We extended the section on the disintegration of BC-containing particles as requested. For more detail please see our reply to review #1.

Q: A detailed error analysis and description of the main error sources of the different used instrumentation and inlet infrastructure (of the research aircraft) is currently missing and should be added.

A: The inlet system on the DLR Falcon research aircraft has been extensively tested and is successfully used for years. For example, Fiebig (2001) investigated the inlet system on the Falcon. In

addition, Schumann et al. (2011) show in Figure 7 the cut-off of the Falcon aircraft inlet, and the effect on the derived particle size distribution.

For the SP2, we have slightly extended the error discussion.

References:

Fiebig, M. 2001. Das troposphärische Aerosol in mittleren Breiten – Mikrophysik, Optik und Klimaantrieb am Beispiel der Feldstudie LACE 98. Ph.D. Thesis. Ludwig-Maximilians-Universität, München.

Schumann, U., Weinzierl, B., Reitebuch, O., Schlager, H., Minikin, A., Forster, C., Baumann, R., Sailer, T., Graf, K., Mannstein, H. et al., 2011. Airborne observations of the Eyjafjalla volcano ash cloud over Europe during air space closure in April and May 2010. Atmos. Chem. Phys. 11, 2245–2279. http://dx.doi.org/10.5194/acp-11-2245-2011.

Q: Page 28753, 28754, 28764: All footnotes should be part of the main text (in a shortened form).
A: Most of the footnotes were implemented in the main text. In two cases, we kept the footnotes, because adding it to the main text would disturb the readability of the text.

Q: Page 28754, Line 11: Is this estimate valid for all seasons or an annual average? Please clarify.

A: This estimate is based on monthly values. The maxima of the aerosol optical thickness of the European fine particle fraction (D_{opt} < 1 µm) are in April and summer. We clarified this in the text.

Q: Page 28754, Line 12: Please add “particle diameter” in the parenthesis.

A: Done.

Q: Page 28754, Line 16: Please add “surface albedo” or something similar in this sentence.

A: Done.

Q: Page 28756, Sect 2.1.: Please focus on the main instrumentation and flights used in this study (e.g. HONO, HCl, etc. are not being discussed here).

A: Done. We shortened this paragraph significantly.

Q: Page 28757, Line 10: Were all five CPC’s being used for the analysis? If not, describe only the ones being used (incl. manufacturer, model, cut-off).

A: In the initial analysis all five CPCs were used. For the number size distribution shown in Figures 1 and 6, just one CPC (TSI CPC 3760A; cut-off at 14nm) was used.

During the measurements, the CPCs were set to different lower cut-off diameters providing information for example on the particle number concentration in the nucleation mode. In addition one CPC were operated on 16 September behind a thermal denuder providing measurements of the non-volatile particle number concentration (the setup of the CPCs, e.g. which CPC or how many CPCs were operated behind a thermal denuder varied during the campaign). For our analysis, particle number concentration in the nucleation mode was important, because the method applied to derive the particle number concentration in the Aitken mode is only justified, when no particles in the nucleation mode are present. In addition, the data from the non-volatile CPC were important to check the consistency of the SP2 BC measurements.

Q: Page 28759, Line 24: What is the assumed uncertainty on this assumption (refractive index) and how does it influences the corresponding particle diameter?

A: The uncertainty of the optical sizing associated with uncertainties of the assumed refractive index is about 5% (± ~ 10 nm) for a 200 nm particle and about 8% (± ~ 50 nm) for a 600 nm particle when varying the refractive index from 1.59+0.00i to 1.50+0.00i or 1.70+0.00i. We clarified this in the text.

Q: Page 28760, Line 5: I would suggest adding a variable sign after the delta, e.g. deltaD_coat.

A: Done, we now use $\Delta_{D\_coat.}$

Q: Page 28760, Line 7: Please use the same notation of the refractive index as before (plus sign between real and imaginary part, see also Page 28761, Line 7).

A: Done. We changed the notation so that it is consistent throughout the paper.

Q: Page 28760, Line 22: I doubt that the assumption (15% uncertainty on rBC mass) is really justified if all uncertainties (incl. the aircraft sampling system) are thoroughly included. Please clarify on the aerosol sampling and SP2 losses and its combination to a total uncertainty assumption.

A: Besides other factors, the accuracy of the SP2's mass concentration measurement depends on the calibration material chosen (e.g. Gysel et al., 2011). For our study, we used fullerene soot as calibration material, because this has been shown to be equally sensitive to fullerene soot and ambient BC from sources where fossil fuel was dominant .

In 2011, 6 SP2s from different research groups were brought to the AIDA chamber in Karlsruhe to assess the reproducibility of the SP2 measurements. During these tests, the mass size distributions of the rBC cores agreed within ±10 % for all 6 SP2s (for more details see Laborde et al., 2012). In summer 2012, the DLR Falcon and the NASA DC8 aircraft flew wing-by-wing within the framework of the DC3 field experiment. During the SP2 intercomparison flight, the rBC mass concentrations measured with the NOAA SP2 on the DC8 and with the DLR SP2 on the Falcon agreed within < 4% (Markovic et al., 2014, in prep.).

The aerosol inlet at the Falcon is well characterized. For typical Falcon speed, the cut-off is near 2.5 µm at ground level and decreases to about 1.5 µm at 10 km altitude (Fiebig, 2001). Since the upper detection limit of the SP2 for rBC was 0.44 µm equivalent diameter for CONCERT 2011, we do not expect that the rBC measurements are affected too much by the inlet cut-off of the Falcon inlet. In addition, the sampling lines were kept short in order to avoid particle losses as a result of long sampling lines. Given all this, we think that 15% uncertainty on rBC mass is justified.

References:

Gysel, M. , M. Laborde, J. S. Olfert, R. Subramanian, and A. J. Gröhn, 2011: Effective density of Aquadag and fullerene soot black carbon reference materials used for SP2 calibration, Atmos. Meas. Tech., 4, 2851–2858, 2011, doi:10.5194/amt-4-2851-2011.

Laborde, M., Schnaiter, M., Linke, C., Saathoff, H., Naumann, K. H., Moehler, O., Berlenz, S., Wagner, U., Taylor, J. W., Liu, D., et al., 2012: Single Particle Soot Photometer intercomparison at the AIDA chamber, Atmos. Meas. Tech., 5, 3077-3097, 10.5194/amt-5-3077-2012.

Q: Page 28763, Line 7: Table 2 is not really needed, since only two flights detected the smoke plume and also Fig 3. shows the same message. The table could therefore be removed (or moved to an additional supplement).

A: We think that Table 2 is important to show that elevated aerosol layers were present during almost every flight within the CONCERT 2011 field experiment. In order to provide more information in Table 2, we added the rBC mass concentrations in the SP2 size range for all observed aerosol layers. We also think that showing the rBC mass concentrations observed in other aerosol layers helps to put the results of the Pagami Creek fire into context with other measurements during the CONCERT 2011 field experiment.

Q: Page 28764, Line 21: Figure 5 could be moved to the supplement.

A: We decided to keep this figure in the main part of the paper, because we think that it is important to show that such smoke aerosol layers can be visible to the human eye. This is also important in the context of the "visible ash" (= volcanic ash which is visible to the human eye) discussion in aviation (see also Weinzierl et al., 2012).

References:

Weinzierl, B., D. Sauer, A. Minikin, O. Reitebuch, F. Dahlkötter, B. Mayer, C. Emde, I. Tegen, J. Gasteiger, A. Petzold, A. Veira, U. Kueppers, and U. Schumann (2012b), On the visibility of airborne volcanic ash and mineral dust from the pilot's perspective in flight, J. Phys. Chem. Earth, doi:10.1016/j.pce.2012.04.003.

Q: Page 28766, Line 2: Please add that CALIPSO is a space borne lidar.

A: Done.

Q: Page 28766, Line 8: Why was the section between segment 12 and 13 not used (especially the second half of this intermediate segment looks like a clear signal to me)?

A: Between segment 12 and 13, we sampled aircraft exhausts and contrails which were mixed with the Pagami Creek smoke plume. Since we focus our analysis on the properties of the aged Pagami Creek smoke plume, we did not consider this sequence in the manuscript.

Q: Page 28766, Line 13: Please precise that Fig. 9 only shows the lognorm-fits of the measured size distributions (besides the median of all the distributions; red points). Error bars for the FT background are missing as well.

A:. The text has been rephrased. We plotted error bars for the FT background case. However, the 15% uncertainty lies within the diameter of the circles of the data points.

Q: Sect. 3: It would ease the reading and improve the structure of the paper, if the discussions follow the description of the individual results. For example, add the discussion of the other measured size distribution from the literature shown in Fig. 10 right after the paragraph of Sect. 3.3.2. (same for all the other subsections).

A: We restructured the paper as recommended and moved large parts of the former discussion section into the results parts thereby avoiding redundancies. However, we prefer to keep a separate "Results" and a "Discussions" section, because we find it important to separate the presentation of the results and their interpretation.

Q: Sect. 4: It looks to me that the HYSPLIT model results miss the lidar observation in Leipzig (Fig. 11g). Please clarify and precise the sentence on page 28770, line 14.

A: We rephrased the paragraph.

Q: Page 28771, Line 20: Are these values measured for plumes with a comparable age?

A: These values are measured for comparable ages. We modified the sentence.

Q: Page 28777, Line 14: Please add the DrBC-value.

A: Done.

Q: Figure 13 and within text: Please give the time unit in pure seconds and not increments of 0.2ms.
A: Done.

Q: Page 28785, Line 19: Please replace “height” by “thickness” or “depth”.

A: Done.

Q: Page 28805: Figure 1 could be removed (shown already elsewhere), moved to the supplement or combined with Fig. 2.

A: Done. We combined Figures 1 and 2.