

Review of “Cross-polar transport and scavenging of Siberian aerosols containing black carbon during the 2012 ACCESS summer campaign” by Raut et al.

General comments

This paper presents a thorough and detailed model investigation of the factors affecting transport of several plumes of BC&CO to the Arctic, comparing the influences of different pollution sources and the spatial distribution, amount and types of precipitation. There is a limited comparison to aircraft measurements that shows the model is doing a reasonable job of capturing the general features of the observations, though not doing a perfect job of simulating the exact concentrations or resolving the finer structure of the plumes (and I would not expect it to). Overall I think the work is of a very high scientific quality, and the main issue I have is that it is too long. However, I think it is probably not suitable to be split into 2 papers, nor are there any sections that could easily be cut without detracting from the rigour of the analysis, so I think it will just have to remain long. Possibly, parts of Section 2 might work as an appendix/supplement. Additionally, some of the written English is phrased in strange ways and some sentences do not quite make sense, so it could do with some work. I recommend publication in ACP subject to the following minor revisions.

Specific comments

P1L13-15 The way this is phrased makes it sound like the source determines the APT. I think this is because you have said “is” rather than “was”, which makes it sound more like a general statement rather than a specific statement about the plumes studied in this paper. This type of error occurs throughout the paper and is mostly benign but can sometimes be confusing, such as in the example above. As a general rule of thumb, the work you have done is in the past (e.g. “the campaign took place” or “we ran the model”) but things you do in the paper itself should be in the present (e.g. “in this study we describe...” or “Figure 5 shows ...”). It gets more tricky, for example when you come to the conclusions- the specific plumes you studied *were* affected by precipitation but plumes in general *are* affected by precipitation.

P3L8-9 “Schwarz et al showed....” I’m not sure what you are trying to say with this sentence, other than to mention the study by Schwarz et al. It seems like you do a better job of saying the same thing in the next sentence, so this one isn’t really necessary

P3L15 What results from Koch and Hansen? Do they confirm them or do they just agree with them? (i.e. both could be wrong)

P5L6-10 Please give details of how the SP2 was calibrated. Actually, you should also say how the CO box was calibrated. Just one sentence for each would probably be sufficient if a standard method was used.

P5L14-15 “Absolute uncertainty of BC particle mass is within 10%, the uncertainty of the derived total BC mass mixing ratio is about 30%.” I am not sure I follow the logic here. Also 10% is not an absolute uncertainty, it is a relative uncertainty- a percentage is relative by definition. Given you do not mention particle size in your analysis, the only relevant errors are A) The systematic uncertainty in your BC calibration and B) The statistical uncertainty in the derived BC mass concentrations.

A) is down to a combination of the sampling time, concentrations and flowrate, but is easy as you can just pick a time when you think your concentrations are constant and look at the variation in your time series. You could express this as a relative error (e.g. +/- 10%) or absolute (e.g. +/- 2 ng/kg)

B) is more difficult as you have two factors- firstly the random variation in your calibration slope (in other words how accurately does your particular slope recreate the mass of the calibration material, if you repeated your calibration exactly how much would the slopes differ?) (see Laborde et al. (2012b)) and secondly how well does your calibration material represent the instrument response to the BC you measure in the atmosphere? As Laborde et al. (2012a) showed, the SP2 responds differently to different BC types. I understand the observations present a fairly minor part of your paper but if you are going to present them and quote an uncertainty it should be done correctly, which at present I don't think it is.

P7L23 You note that the height of the emissions injection is very important- how good a job does the plume rise model do? How does it work out the buoyancy of a particular fire? Please provide a brief summary

Figure 2a is there a reason why potential temperature is more useful than just temperature?

P10L20 wrong OH and transport- the way this is written it is not quite clear what you mean. Wrong transport? Vertical or horizontal? Or just transport? Do these factors and the studies you reference explain why CO is underestimated specifically between 6 – 9km?

Section 3.2 I think you are overselling the agreement between model and measurements. For example "The two profiles are well correlated with maximum CO values of 200 ppbv at 7–8 km, associated with elevated BC values reaching 25 ng kg⁻¹." Actually the maximum in the model CO is ~150ppb at 6.5km. Compare to P32L10- this is a better way of describing the model/measurement comparison. The general features were well captured.

P11 L 1 "the influence of flaring emissions in this area is insignificant". Insignificant for what? At what scale? Later on in the paper you talk about some flaring plumes so it can't have been insignificant within those plumes.

P11L7-8 "The model shows appreciable skill in capturing the vertical profile of BC, but overestimates the BC mixing ratio between 2 and 3 km of altitude." Looking at the median BC concentrations, the 40km model overestimates between 1.5 – 7.5km then underestimates higher than 7.5km. Additionally, if you just went with a flat BC concentration of around 6 or 7 ng/kg the medians would probably show similar agreement. Now, I am not saying that the agreement is terrible- actually it OK, perhaps reasonably good, and it seems to capture the general features of the observations without doing a perfect job. But you read the text and it sounds like the model is doing an amazingly good job, which figures 3 and 8 show it isn't, it's just doing a reasonable job. So I think you should just tone down how well you claim the model and observations agree.

P11L9 The 30% error in the SP2- please relate this back to the previous comment on statistical vs systematic error. Also please give numbers for the model biases.

P11L13 Here you say the CO between 6-9km is due to biomass burning emissions, but previously you said the underestimation in CO was due to wrong OH concentrations and transport. Could it not be

that the model is underestimating the biomass burning CO at this altitude? Is that what you mean by transport? Please clarify

P13L11 You say the AOD underestimation is due to simplified SOA, but you haven't given any details of how the AOD is calculated. I don't think you can say this is due to missing SOA when you don't have a good handle on even the size distribution, let alone composition or refractive index.

P18L13 are the BC enhancements you are talking about in the model or in the observations? I assume model but it's not clear

P18L23 The agreement between model and measured BC is "very good" - Again I would say it's reasonable but I wouldn't say it's "very good". The model plumes in figure 5 are too diffuse and some are missing, such as the smaller amounts of BC associated with the CO plume at ~8km at 0915. You get the general features. For me, "very good" would be if you could plot model vs measurement for each grid box as the aircraft passed through it and get something approaching a 1:1 line, though I doubt that could happen in a study like this.

Figure 8 You could do with a longer time average of the measurement data as currently it's difficult to see the structure when the markers all overlap. I also wonder if there is a way to make the aircraft easier to see as the parts that stand out are the parts where it disagrees with the model. Also it might help if the x-axis was north/south or east/west as several points in the discussion relate to spatial location and this is difficult to see in this and subsequent plots

P21L18 If the aerosol from flaring had been removed by precipitation, wouldn't you still see the CO?

P21L25 Didn't you say in the previous section that the flaring plumes didn't exist?

Section 4.4 The discussion may be easier to follow here if you circled the plumes on one of the figures

P23L2 you say the fire injection lofted the plumes to 6km, but doesn't fig 10d shows that the emissions from the 8th initially remained below 4km. I'm not really sure I follow what figure 10d adds to the analysis

P24L20 How does figure 9 show European influence?

P24L24 Please define KFCuP in the text. You refer several times to KFCuP as if it is a process itself. In the real atmosphere it is actually "convective clouds" that do things that the model is trying to represent.

P26L4 It's frowned upon these days to refer to it as a coating, as that may not represent the actual morphology of the particle. Maybe say BC in BB plumes is more internally mixed or something like that. I also saw another point where you referred to coatings, please also change this.

P26L11-14 I don't know what these last 2 sentences add to the analysis

P27L19 The removal efficiencies may be low for large rain drops but not for drizzle

P29L6 The 5th percentile of the measured or the modelled CO concentration? Also why not do this for the BC as well? The average lifetime of aerosols is of the order of a week because of deposition

processes, mostly wet deposition. So the lifetime of aerosol that escapes wet deposition is longer. If you mean you looked at figure 3 and saw that the minimum BC in the observations was basically zero at all levels then say that. But if that was the case I still don't see the harm in taking the 5th percentile like you do with the CO.

P29L22 Perhaps this is not the case in the model but in the real atmosphere BC would only be lost if the cloud precipitated. If it evaporated the BC would still be there.

P32L16 Can you suggest how the discrepancy/difference might be resolved? Is it just because there is less precipitation in this study?

P33L30 "found to be more important" It is not clear what you mean by this. If you mean the cumulus clouds remove more of the low-level aerosol by scavenging or washout than by uplifting it then say that. It's better to say what is actually happening.

P34L1 This last sentence is an odd way to end a paper. It sounds like you are using the model to validate the measurements when actually it was the other way round. Also this wasn't the main focus of your analysis. It seems to me that the main important conclusions were

- 1) BC is transported more efficiently into arctic from high-latitude BB sources than from east-Asian anthropogenic sources because it rains less at higher latitudes
- 2) The ways in which the large-scale vs subgrid convective clouds affected the BC distributions differently.

It would be good if you could highlight these more in the abstract/conclusions in terms of the physical processes your results suggest are actually taking place in the real atmosphere, rather than abstract terms like grid-scale and APT

Finally, given you have actual observations it seems like there is a missed opportunity to calculate TE_{BC} based on the measured values of BC and CO. You have hinted at your reasons for not doing so in the comparison to previous studies calculating TE_{BC} but I'm not sure it's clear exactly why not.

Technical corrections

P3L25 "lead to in the" remove 'in'

P3L27-31 This is a very long sentence, please split it into at least 2.

P8L4 add "resolution" before "to adequately"

P11L12 remove "but"

Figure 3 please make the plots slightly taller to show the detail better

P11L19 This is a very long sentence, please split it into at least 2.

P12 L4 remove “that”

Figure 4 the scale is between 0 – 1 but the text mentions values up to 2.5

Figure 5 are (a) and (b) really the same grid? Panel (a) looks much more blocky. Also label (%) on panel (c) colorscale

P15L1 “than in altitude”, do you mean “that at higher altitudes”?

P18L6 “plumes transported in altitude” do you mean “at altitude”?

P18L20 “shifted a bit”- how far? Saying “a bit” is probably *a bit* too colloquial for a paper (but not a review!)

P21L17 “not due to” do you mean “did not lead to”?

P27L34-35 This sentence doesn’t make sense

Figure 13 caption Maybe rephrase to “The points are colored by the time (in hours) before the release of the trajectories in FLEXPART-WRF)”

P30L25 You may consider not using the word “septentrional” as it is not a commonly used word.

P32L4 Again change “a bit” to “slightly”

P33L4 Remove “very”

Finally, thank you for your interesting (but long!) paper

References

Laborde, M., Mertes, P., Zieger, P., Dommen, J., Baltensperger, U. and Gysel, M.: Sensitivity of the Single Particle Soot Photometer to different black carbon types, *Atmos. Meas. Tech.*, 5(5), 1031–1043, doi:10.5194/amt-5-1031-2012, 2012a.

Laborde, M., Schnaiter, M., Linke, C., Saathoff, H., Naumann, K.-H., Möhler, O., Berlenz, S., Wagner, U., Taylor, J. W., Liu, D., Flynn, M., Allan, J. D., Coe, H., Heimerl, K., Dahlkötter, F., Weinzierl, B., Wollny, A. G., Zanatta, M., Cozic, J., Laj, P., Hitzenberger, R., Schwarz, J. P. and Gysel, M.: Single Particle Soot Photometer intercomparison at the AIDA chamber, *Atmos. Meas. Tech.*, 5(12), 3077–3097, doi:10.5194/amt-5-3077-2012, 2012b.