

Interactive comment on "Quantifying the sensitivity of aerosol optical properties to the parameterizations of physico-chemical processes during the 2010 Russian wildfires and heatwave" by Laura Palacios-Peña et al.

Anonymous Referee #1

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Palacios-Peña and co-authors have analyzed the sensitivity of simulated aerosol optical properties (aerosol optical depth, extinction and backscatter) to parameterizations of several physico-chemical processes. They did the analysis using the fully-coupled on-line WRF-Chem model and concentrating on the well documented Russian wildfire event in 2010. Their sensitivity studies showed some non-linear responses in optical properties which is a surprising result. Therefore, the manuscript is relevant to the readers of the ACP and it will likely be interesting to a wide audience. The methods are sound and described in enough detail, and the assumptions appear to be reasonable.

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My detailed comments are given below.

My main concern with this study is related to the comparability of the different sensitivity tests. For example, RH is changed by -10 %, +0.5 %, and +1 %, whereas dry deposition velocity is scaled by 0.5 and 2 for the Aitken mode and 0.1 and 10 for the accumulation mode. As these changes vary from 0.5 % to 1000 % it is quite hard to understand how these sensitivity tests compare with each other. And why didn't you simulate RH reduction of 1 % and 0.5 %? Then they could have been compared directly with the enhancements. I think, it would be good to explain in the text why these changes are thought to be representative, meaningful and comparable for the parameters. For example, do they represent similar portions of the total ranges of the parameters? Or do they map the uncertainty ranges of these parameters? In order to say that sensitivity of the optical properties to RH is more important than to dry deposition, the changes in the parameters should be somehow comparable. This could be the true for current the analysis but it is not clear to the reader.

As the sensitivity test were made for an exceptional event, it is not clear how easily they can be generalized. For example, the low amount of clouds around the fire regions makes it seem that wet scavenging is not that important. However, based on Figure 2, wet scavenging has the strongest impact in the low-AOD region as there are more clouds present. Could you please clarify in the text if the conclusions are limited to specific conditions or if there are other processes which might have a stronger effect in some conditions.

I would have also liked to see a bit more detailed discussion on the significance of these results. Lately, there have been some studies where identical anthropogenic aerosol fields have been used in different models. For example, Nordling et al. (2019) found significant differences in the aerosol forcing between the models and they concluded that differences in model circulation responses appear to dominate the differences in regional climate responses. So, I feel that it would be an interesting addition to discuss (and compare at some level) the significance of the processes analyzed in this

manuscript and uncertainties in simulated circulation.

Page 1, lines 10-11: Are these absolute or relative differences. Would be good to clarify.

Page 2, line 23: "larger uncertainty", larger than what? Please, clarify.

Page 2, line 33: "as aerosol optical properties" \rightarrow such as aerosol optical properties. Can optical properties be considered as a process?

Page 2, line 47: "high" \rightarrow highly

Page 3, line 80: "To achieve this objective", it is not entirely clear what is objective you are referring to.

Page 3, line 83: What are the wavelengths of the AOD, extinction and backscatter coefficients? Please, mention them in the text.

Page 4, line 89: This sentence is a bit confusing: " with monthly mean temperatures in the summer months 5–9âUeC higher than those for 2002–2009 due to a prolonged blocking anticyclone situation which triggered large wildfires". First of all, I'm not sure what you mean with the temperature comparison. Were the monthly mean temperatures 5-9 degrees warmer than the monthly averages for 2002-2009 or was the comparison done for individual months and the temperature range covers all these months? Secondly, I don't think it was the anticyclone situation which triggered the fires. I believe it was the people and the meteorological situation just made the thing worse.

Page 4, line 107: I wouldn't call 0.95 a very high single-scattering albedo as sulphate aerosols have SSA close to unity. Or do you mean that the SSA was high for smoke aerosols?

Page 4, line 116: There seems to be something missing from the end of the sentence ("in the "). Also the unit should be Wm-2.

Page 5, line 142: Temporal profile of what? It seems that this sentence is missing

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some words.

Page 6, line 156: "as nucleation, chemistry or uptake of water" \rightarrow such as nucleation, chemistry and uptake of water

Page 7, line 193: Is the modelled AOD in Figure 2 an average over the studied period? I'm just wondering if an average is the best way to present the data as there was a lot of variability in AOD during the episode and single outliers can have a big impact on averages. Did you check how the results would look if you would use medians instead of averages? It would also be interesting to see the variability of AOD during the period. It is likely higher in the MAX-AOD and Moscow points than in the MIN-AOD point, which might have an effect on the differences between the studied points.

Page 7, line 194: "The top-right figure shows the mean bias", do you mean the text in the top-left corner of the plots?

Page 7, line 208; "but less significant", compared to what? I'm surprised that there isn't more discussion on the HDDV_ACC simulation as it produces the largest mean change in AOD (-0.06).

Page 7, line 213 and 216: The indices of the subplots seem to have been mixed: Figure 2,j \rightarrow Figure 2,k, Figure 2,k \rightarrow Figure 2,j

Page 8, line 217: What do you mean with smooth differences? Please clarify.

Page 8, line 220: Here you could also mention that the mean AOD difference (0.04) for this simulation is the second largest even though there aren't many clouds in the studied domain. It implies that the wet scavenging is really important when there are clouds present.

Page 8, line 224: "where the spot where" \rightarrow the spots where. "claims to bring" \rightarrow aims to bring

Page 8, line 225: "time mean" \rightarrow temporal mean

Page 8, line 228: Please clarify in the text that these are profiles are temporal averages. Would the results look the same if medians were used instead of averages?

Page 8, line 230: " α and β present similar profiles" sounds a bit strange to me. Do you mean that the profiles of α and β have similar shapes?

Page 8, line 232: This and the following sentence are missing "for β " after the backscatter values.

Page 8, line 245: These low LR values are a surprising and interesting result. Especially, as the LR values over the MIN-AOD location are in the same range as reported by Mielonen et al. (2013). What could explain the large difference in the source and reasonable results farther away? I believe this would be an interesting point to discuss in the manuscript.

Page 9, line 251: Please, clarify in the text how you calculated the mean absolute error for the profiles in practice. Did you first calculate the errors for each model level and then average them for the whole profile?

Page 9, line 252: I didn't quite catch how you calculated the normalized error. Could you please clarify? Which values were used in the normalization and how was it done in practice? Did you use the pixel-wise mean values from the base case at each model level or averaged over the whole column?

Page 9, line 262: "optical properties profiles" \rightarrow profiles of extinction and backscatter coefficients

Page 9, line 277: "However, in the case with a reduction of the RH in a 10 % (L10RH), NO-3 displays a similar concentration as the base caseat surface levels and higher at levels above 800 hPa.", to me it seemed that the concentrations were similar only at surface levels and around 800 hPa. Did I read the figure wrong?

Page 10, line 288: "hidroxy" \rightarrow hydroxy

Page 10, line 294: "the shape of the NOx and SOA profiles are similar, and thus, at these vertical levels, variations in SOA concentrations may be due to the effect described by Sarrafzadeh et al. (2016): an increase in NOx concentrations at low-NOx conditions (less than 30 ppb or around 55 μ g m-3)", this is a bit hard to follow. Would something like this work better: the shape of the NOx and SOA profiles are similar, and thus, at these vertical levels, variations in SOA concentrations may be due to an increase in NOx concentrations at low-NOx conditions (less than 30 ppb or around 55 μ g m-3)", statement of the NOx and SOA profiles are similar, and thus, at these vertical levels, variations in SOA concentrations may be due to an increase in NOx concentrations at low-NOx conditions (less than 30 ppb or around 55 μ g m-3; Sarrafzadeh et al. (2016))

Page 10, line 297: "meanwhile in the L10RH case the positive variation of the concentration of SOA caused by the RH is limited.", I'm not sure what you mean with this. Could you please clarify?

Page 10, line 300: "provokes" \rightarrow provoke

Page 10, line 309: With "target area" you mean the MAX-AOD point? I find it interesting that in this NO_DD simulation the positive AOD change forms a similar arch as in the H05RH simulation whereas other simulation exhibit a more uniform blob around the MAX-AOD point (see Figure 2). Furthermore, the lowest values in the NO_WS simulation match approximately the "gap" in the blobs of the NO_DD and H05RH simulations. Could some specific process explain these common features in these simulations? Also, in this NO_DD simulation the AOD increased a lot more around the MAX-AOD point so would the conclusions have changed if the selected point would have been a bit more eastward (see Figure 2)? Based on the AOD changes shown in Figure 2, moving the point slightly eastwards would not affect the magnitude of the change much in most simulations. Maybe doing the profile analysis with averages calculated over a number of pixels would give more robust results as the AOD changes in all the simulation are not smooth around the MAX-AOD and Moscow points?

Page 10, line 315: Please note that Supplementary Figures 2 and 3 are not mentioned in the text at all.

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Page 10, line 316: "modifying the accumulation mode" \rightarrow modifying the deposition of the accumulation mode. And the same change for the Aitken mode on the next line.

Page 11, line 327: "However, those species which are not directly emitted but are products of atmospheric chemistry (secondary aerosols), as SOA (NMAE>0.8 and MAE 0.2283) and most of the secondary inorganic species have their concentrations peak higher than thosein the base case between 900 and 600 hPa", this sentence is hard to follow. Please, revise.

Page 11, line 329: "fires area" \rightarrow fire area, "optical properties profiles" \rightarrow profiles of optical properties

Page 11, line 333: The Greenfield gap may not be familiar to all readers so, please, provide a size range and a reference for it.

Page 11, line 340: "When the profiles are analyzed, the response differs between species. EC, POA and NO–3 shows a slight reduction in their concentration, and SOA exhibits a large reduction.", is this correct? Based on Figure 3, it seemed that the concentrations of NO–3 and SOA increased. Furhermore, the SEA concentration appeared to decrease and not increase as mentioned in the text and the the highest SO2-4 concentrations appear to be around 800 hPa, not near the surface. Did I read the figure correctly?

Page 11, line 348: " β profile is similar to the profiles of organic species (EC, POA and SOA)", to me it seems that the β profile is also similar to NH-4 and SO2-4 profiles.

Page 12, line 358: "scaled to 1.5" \rightarrow scaled by 1.5

Page 12, line 359: "For these species, the NO_CONV_TR experiment exhibits a concentration profile similar to the base case with slightly higher concentrations at surface levels and lower at higher levels", isn't it the opposite for the SEA concentrations? And the SOA, NO-3, and NH-4 concentrations appear to be constantly smaller than the base case? It would also be good to mention at the beginning of each section that

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which point is analyzed. I'm guessing this analysis is related to the MAX-AOD point.

Page 12, line 369: ""differently as" \rightarrow differently than

Page 12, line 376: "show a peak in their profiles around the PBL" \rightarrow show a peak around the PBL

Page 12, line 382: "at surface levels" \rightarrow at the lowest levels

Page 12, line 383: "below the PBL" \rightarrow below 800 hPa

Page 12, line 384: "highlights the high impact of organic species", please, clarify this statement. The concentrations of inorganics over PBL are also decreased so why are organics are more important? Is it related to the higher mass of POA (max 150 μ g m-3 vs. ~4 μ g m-3)

Page 13, line 387: "experiment is that with the strongest" \rightarrow experiment has the strongest

Page 13, line 391: "It should also be highlighted that over the MIN-AOD and Moscow spots, EC and POA profiles of the assessed experiments show larger differences between them than over the MAX-AOD. This fact could be explained because over these locations these species are not being directly emitted. Moreover, the farther the location is, the larger the differences are." This statement could be true in relative sense but what about in absolute values? The concentration scales in the figures for the different locations are quite different. For example, the POA concentration scale is up to 150 μ m m-3 at MAX-AOD, 30 μ m m-3 at Moscow and only 2 μ m m-3 at MIN-AOD. Therefore, based on figures 3, 5, 7 it is quite impossible to say which location has the largest changes in absolute sense. Could you please discuss this in more detail in the text?

Page 13, line 397: "In order to reduce", please explain how the uncertainties can be reduced based on the results presented in this study.

Page 13, line 400: "carried out during" \rightarrow carried out for

Page 13, line 412: What do you mean with "important supersaturation"?

Page 14, line 442: It would be good to mention in this paragraph that the simulated LR values were different only/mainly at the MAX-AOD location, not everywhere.

Figure 2: Please, consider using binned color scale for the base AOD plot as well. Currently, the different hues are quite hard to differentiate. A binned color scale would make it easier to see the differences between the regions.

Figures 3, 5, and 7: Currently the lines are quite hard separate from each other. Maybe thicker lines would make it easier to see the colors?

Figures 4, 6, 8: Please, consider using binned color scale for the NMAE as it could make it easier to compare the different cases.

References

Nordling, K., Korhonen, H., Räisänen, P., Alper, M. E., Uotila, P., O'Donnell, D., and Merikanto, J.: Role of climate model dynamics in estimated climate responses to anthropogenic aerosols, Atmos. Chem. Phys., 19, 9969–9987, https://doi.org/10.5194/acp-19-9969-2019, 2019.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-42, 2020.