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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessment of natural and anthropogenic aerosol air pollution in the Middle East using MERRA-2, CAMS data assimilation products,and high-resolution WRF-Chem model simulations" *by* Alexander Ukhov et al.

Anonymous Referee #1

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This paper shows the dust and non-dust aerosol components over the Middle East that are available from two reanalysis products, MERRA-2 and CAMS, and WRF-Chem model simulations. It first compares the 10-m wind speed among the products, then compares AOD and size distributions with remove-sensing data (such as MODIS MAIAC and AERONET) and PM2.5/PM10 concentrations with ground-based measurements. With the results from WRF-Chem simulations and reanalysis products, the aerosol composition of PM2.5 and PM10 is presented and days of PM above the reg-

ulatory standard are estimated.

I found this paper is interesting in the sense that an evaluation of model and reanalysis products is specifically performed for the Middle East region with, albeit limited, remote sensing and ground-based measurements, and the seasonal and annual levels of PM are presented. I however have quite some comments regarding the presentation and understanding of the products used in the paper, and recommend substantial revision before accepting for publication on ACP. I believe that the revision is not difficult to deal with although it could be extensive.

For the presentation style in general: - Abstract should contain only one paragraph with acronyms spelled out (e.g., ME). - Introduction section is too long - should be more concise and more relevant to the point of the study. It is not a literature review. - Conclusion section is also too long and unfocused. It has 14 paragraphs! It should be consolidated with key points summarized and highlighted, not list everything you have done.

Aerosol composition: there are no data to evaluate the models. The surface measurement data are for PM, not chemical species. Besides, the models do not include nitrate and ammonium, and it seems they don't have the chemical mechanisms for producing secondary organic aerosols. Therefore, the chemical composition from the model omits some important components. The problem should be acknowledged at least. Is there any reference for the aerosol composition in the region?

Reanalysis products: It should be pointed out that the reanalysis products from MERRA-2 and CAMS-OA are the reanalysis of AOD, not the mass concentrations of individual aerosol species. The mass of individual aerosol specie is adjusted mostly proportionally according to the differences between the AOD from native model simulation and after the assimilation of satellite data. Also, in general, a better understanding of the reanalysis products and other products is needed.

Comparisons with data: The comparison with AERONET AOD is not an independent

evaluation of WRF-Chem and MERRA-2, because the WRF-Chem is "tuned" to match AERONET AOD and MERRA-2 assimilates AERONET AOD. This evaluation should be properly addressed. Also, for the AOD comparisons with both AERONET and satellite data, it is not clear if the comparisons were done under the same spatial and temporal conditions (e.g., models are sampled under clear-sky only condition or all-sky, if model and data are temporally matched).

To me, a major conclusion is that the PM2.5 concentrations over the Middle East (at least at the places the study was examined) almost never below the WHO standard because of the dominance of dust in PM2.5 which cannot or very hard to mitigate. Even if all anthropogenic emissions are shut down, the air quality in the Middle East will not improve. What is the implication for that? How to improve the air quality in the Middle East under such circumstance? This problem should be discussed.

Specific comments:

Page 1, line 4: Spell out "ME".

Page 2, line 26: "mass budget" – it should be "emission budget".

Page 2, line 55-56: AVHRR was not designed to measure column aerosol properties. It was designed to observe clouds, surface temperature, and vegetation but later was expanded to retrieve aerosols over the ocean.

Page 3, line 57: Change "CALIOPE" to "CALIOP".

Page 4, line 92: "we improve the latest. . .emission. . ." does not sound appropriate. You just use the new SO2 emission data set. What is the spatial resolution of the new SO2 emission data set from Liu et al. 2018? Does it match the WRF-Chem spatial resolution? Do you have to do "downscaling" or interpolation?

Page 6, line 123: Which wavelength is your chosen reference wavelength? What the wavelengths pair you used to calculate the Angstrom Exponent? Or did you use the Angstrom Exponent provided by AERONET?

Page 7, line 167-168: "MERRA-2 assimilates AOD at 550 nm from the AVHRR over the oceans": This was done before the MODIS observations. MERRA-2 assimilates the MODIS AOD over the oceans since 2000.

Page 7, line 169: "specially processed MODIS observations. . .": What product is that? Any references for such "non-standard" product?

Page 7, line 184: "CAMS-OA assimilates MODIS observations": Be more specific on what MODIS product(s) it assimilates.

Page 8, line 200: "wavelengths larger than 450 km"???

Page 9, section 4.1: SO2 is oxidized to form sulfate aerosol. It is described that gas phase SO2+OH reaction is done with the RACM, but it is not clear how the heterogeneous reactions are treated. Such description should be added.

Page 9, line 234: "the first bin appears to be very poorly populated": Why? The small particles should be transported by the winds more easily than the larger particles. Explain.

Page 10, line 260-261: What was the error that you are correcting? Simply saying it was corrected because it was incorrectly calculated does not help the readers/users.

Page 10, section 4.2: I don't think the bug-fix needs to be described in a devoted section. It can be summarized in a few sentences in the model description.

Page 12, Figure 3 caption: change "for" to "from".

Page 11-13, section 5.1: Why not compare soil moisture and precipitation, since you mentioned on page 11 that dust emission and deposition are sensitive to the soil moisture and precipitation.

Page 14, line 321: What is "the lower atmospheric layer"? i.e., what is the altitude range the dust is emitted into? Or is it emitted into the lowest atmospheric layer? Please clarify.

Page 14, line 324-325: “But because. . .” this sentence has been said in the WRF-Chem description section. It does not belong here anyway.

Page 14, line 328-329: “WRF-Chem underestimated. . .” What is the evidence for that? Is there any reference or from your own simulation describing that problem? This contradicts the findings by Kok et al. that global models overestimate the fine mode aerosols but underestimate the coarse mode aerosols.

Page 14, line 330: How do you know that the total emitted dust mass is overestimated, since there is absolutely no measurements of dust emission?

Page 14, line 331, adjusted sp fraction: The first size bin represents clay and the rest four bins represents silt. The 0.1, 0.25, 0.25, 0.25, 0.25 fractions is based on the assumption that 10% of clay will be emitted but 100% of silt is subject to be emitted to the atmosphere based on the early work in the 1990s from Tegan. Even though these numbers are arbitrary, but the sum of adjusted silt fractions (0.15, 0.17, 0.38, 0.1) is only 0.8. Please explain why you do not account for the rest of 0.2 fraction in the silt group.

Page 16-18, Section 5.2.2: As I mentioned at the beginning, the comparison with AERONET AOD is not an independent evaluation of WRF-Chem and MERRA-2, because the WRF-Chem is “tuned” to match AERONET AOD and MERRA-2 assimilates AERONET AOD.

Page 17, Figure 6: There are several very large spikes of AOD from the WRF-Chem simulations in Mesaira and Sede Boker in 2016. What causes these spikes?

Page 19, Figure 7: How are the models sampled when compared to satellite data? Are they temporally matched (i.e., model results are concurrent with the satellite data, or model results are averaged for the clear sky only during the season)?

Page 20, line 388-389: “. . .in good agreement with. . .”: What is your criteria for “good agreement”? In general, such subjective statement should be avoided. Instead, you

could say something more quantitative, such as “with xx%” or “correlation coefficient within xx-yy”.

Page 20, line 406: “. . .good agreement. . .” again! See my comments above.

Page 21, line 420: sulfate ion: So for PM2.5 you only consider the mass of sulfate ion, not neutralized sulfate that exists in the atmosphere, such as ammonium sulfate? The mass of ammonium sulfate is 37% more than just sulfate ion.

Page 21, line 422: again, what is the reason that “the first sea salt bin is poorly populated”?

Page 21, line 426-429, PM calculations: It should be noted that all models do not include nitrate and ammonium when calculating the PM mass. Associated error/uncertainty should be estimated.

Page 24, 3rd line from the bottom: “As we have shown, WRF-Chem provides reliable estimates. . .”: What is the criteria for "reliable"? From Fig. 8, WRF-Chem underestimates PM2.5 at Jeddah and Riyadh by a factor of 2 and overestimates PM2.5 at Dammam. Its performance for total PM2.5 is inferior to CAMS. In addition, its chemical composition of PM have not been evaluated at all.

Page 27, Figure 10: The labels and legends on this figure are way too small to be legible.

Page 28, line 516-517: Is this a “drift of sulfate”? What is the emission patter of SO2?

Page 28, line 517-521: I don’t understand what the relevancy is to refer the sulfate concentration over the US.

Page 28, line 523, MERRA-2 underestimates the SO2 emission: Do you know if indeed sulfate is too low or SO2 emission is too low in MERRA-2? Several issues here to challenge such statement. First, sulfate mass in MERRA-2 is not necessarily corresponding to SO2 emission because the aerosol masses (including sulfate) are adjusted

after the AOD simulation, which has nothing to do with SO2 emission. Second, van Donkelaar's work "retrieved" PM2.5 based on the satellite AOD and the GEOS-Chem model such that the sulfate (and other aerosols) concentration is adjusted based on the adjustment of model AOD to satellite total AOD. As a result, the sulfate from van Donkelaar's work is not necessarily representative of the "true" sulfate concentrations.

Page 28, line 538-539: Again, I don't understand what the relevancy of the US-EPA standard being applied here. The Saudi Arabia's standard should be used. And in line 539, now you use the WHO guidelines as reference. This is confusing.

Page 30, line 554 and 556-557: I would not emphasize "for the first time" to elevate the significance of the paper. Simply state what you've done and found is more appropriate.

Page 30, line 564: "The air pollution in the major Middle Eastern cities is evaluated" sounds overstatement. The evaluation is rather limited to only three cities and only with PM2.5 and PM10, not all major cities and not all pollutants.

Page 30, line 576: " improve calculation of sulfate aerosol": there is no approve that sulfate simulation is improved because there is no data to evaluate it.

Page 31, line 582-583: CAMS-OA deficiency has been corrected: Then why don't you use the latest version that is available in 2019? What is the point to evaluate the results from an obsolete model version?

Page 31, line 589: "quite well" – again! Please avoid using such subjective statement.

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