Review of Benedetti et al. Status and Future of Numerical Atmospheric Aerosol Prediction

This is a useful review of the status of an important, emerging field, and is sufficiently well developed to be posted as a discussion paper, in my opinion. With some refinement, I think it deserves publication in ACP. The review touches on many things, and it would be helpful if it also provided a bit more synthesis, i.e., if it identified more specifically which issues are currently the most important for certain applications, and which might be achieved most readily.

As this is a review paper, key references might be added in paragraphs where they are lacking, and in several places, text covering a topic in several places could be consolidated, making the presentation more concise and easier to follow. I have included below some specific suggestions for the authors to consider. As I don't have time to reread the paper for this review, I've given it a fairly careful review now. So this is my full review, and as such, it can be posted when the time comes.

1. Section 1.1, Point 1. You might add something along the lines of "reasonably well documented" to the data distribution requirements.
2. Section 1.1, Point 3, line 102. "… for verification and model refinement purposes, …"
3. Section 1.2, lines 118-119. Perhaps: "… data from an observational platform and simulations from a model." Just to distinguish measurements from model results.
4. Section 1.3, line 223. Perhaps: "… aerosol particle emission, secondary production, and removal."
5. Section 2.1. Could you say more about why "ensembles" in general seem to work so much better than individual models? For example, wouldn't the selection of ensemble members matter?
6. Section 3.1, line 278. Might be: "… whereas in remote regions, transports and aerosol processes control the uncertainty."
7. Section 3.1, line 288. By "DA" I assume you mean data assimilation, but this is not defined previously, as best I can tell.
8. Section 3.1, lines 295-296. Might be: "… temporal resolution, speciation, aerosol size distribution, and hygroscopicity." Wouldn't hygroscopicity matter for aerosol-cloud interaction and particle removal efficiency in the models? Perhaps aerosol light-absorption properties is a bit more removed from the considerations for forecasting, but I thought I'd just mention that as a factor that might also be worthy of mention here.
9. Section 3.2, around line 305. I'm a bit confused by this discussion. "Biogenic" aerosol often refers to the aerosol produced by secondary processes from gases emitted by natural vegetation. You seem to mention all these elements in this section, but as written, it is difficult to discern how they relate to each other.
10. Section 3.2.1. Regarding the accuracy of inventories, another issue is "small" sources. For satellite-based inventories, sources too small to be detected by satellite fall into this category. To take one example, this can be a big issue for smoke inventories of agricultural-burning regions.
11. Section 3.2.4. I'm wondering whether the requirements for particle vertical distribution, light-absorbing properties, and/or Mass Extinction Efficiency should be covered in this or one of the other subsections here.
12. Section 3.2.5, lines 377-378. I think there is a typo regarding the subscripts.

13. Section 3.3, Line 400 ff. In small-fire regions, the required factors can be much larger. See, e.g., *Petrenko et al*., JGR 2017, *doi:10.1002/2017JD026693*.
14. Section 3.3.1, lines 463-464. Overlying smoke opacity and fire emissivity are two additional factors that might be mentioned here (I know they are mentioned elsewhere in this section). A similar point applies to "small" fires (see point 10 above). More generally, the fire-emissions subsections seem a bit longer than necessary – a little reorganization could help consolidate and remove a bit of "rambling." This paper is quite long, which is fine, but it would be a service to readers to consolidate as much as possible.
15. Section 3.3.1. Another, empirical approach relating FRP directly with smoke emission should be mentioned here: *Ichoku, C., and L. Ellison*, 2014. Global top-down smoke-aerosol emissions estimation using satellite fire radiative power measurements. *Atmosph. Chem. Phys., 14, doi:10.5194/acp-14-6643-2014*.
16. Section 3.3.1, Line 501-506. Especially as this is a review paper, some references regarding inverste modeling would be appropriate here.
17. Section 3.3.3, line 564. Another appropriate reference here, for completeness, would be *Val Martin et al*. 2012, Space-based observations constraints for 1-D plume-rise models. *J. Geophys. Res. 117, D22204, doi:10.1029/2012JD018370*.
18. Section 3.4, lines 596-597. It would be helpful here to include one more sentence, saying what they accomplished.
19. Section 3.4, lines 603-604. Would pressure or wind sensors be most efficacious in this case?
20. Section 3.4, lines 629-633. Perhaps this reference would be useful here: *J. Li et a*l., 2016. Reducing Multi-sensor Monthly Mean Aerosol Optical Depth Uncertainty Part I: Objective Assessment of Current AERONET Locations. *J. Geophys. Res*. 122, doi:10.1002/2016JD026308.
21. Section 3.4 overall. I note that the desert dust section goes into much less detail about processes (e.g., surface roughness length, mobilization thresholds, etc.) than the corresponding discussion in the biomass burning section.
22. Section 3.6, line 685. It might be worth mentioning here that dry removal also depends on particle size, shape, density, and hygroscopicity.
23. Section 4.1, first paragraph. To some extent, by assimilating radiances rather than retrieved quantities, all the assumptions and issues treated in the satellite retrievals get pushed onto the model. The assumptions involved are likely to be more consistent, as you note, but not necessarily better, given the attention the modelers must pay to all the other components of the model.
24. Section 4.1, lines 750-757. Are there any appropriate references for this material?
25. Section 4.2, lines 771-777. This largely duplicates the discussion in Section 4.1.
26. Section 4.2.2, lines 797-804. One appropriate reference here might be: *Zhang, J., and J.S. Reid*, 2006. MODIS aerosol product analysis for data assimilation: Assessment of over-ocean level 2 aerosol optical thickness retrievals, *J. Geophys. Res. 111*, doi:10.1029/2005JD006898.
27. Section 4.2.2, Points 1 and 2. AeroCom and AeroSat are spearheading considerable work in the area of pixel-level uncertainties for satellite aerosol retrievals. One example publication: *Witek, M. et al*., 2018. New approach to the retrieval of AOD and its uncertainty from MISR observations over dark water. *Atmosph. Meas. Tech*. doi.org/10.5194/amt-11-429-2018.

28. Section 4.2.4. In practical terms, is there specific temporal sampling that would address specific data-assimilation needs?
29. Section 4.2.5, lines 842-848. Shouldn't this have been covered, actually in more detail, in the earlier, measurement sections? Also, obtaining pixel-level uncertainties on any retrieval-produced speciation is another issue about which something might be said.
30. Section 4.2.5, lines 865-866. The idea that dust and sea salt are "coarse mode," whereas pollution and smoke are "fine mode," is a gross oversimplification. Both dust and sea salt have size-distribution tails that extend into the fine mode, and often dominate the fine mode. If speciation really matters for the modeling applications under consideration, this needs to be clarified.
31. Section 4.2.5. How good would the particle size and AAOD information need to be to make a significant contribution to NWP?
32. Section 4.2.5, lines 873-874. Surface measurement will not get the transported aerosol, and where there are local aerosol sources, interpreting the results can be complex.
33. Section 4.2.6. MODIS aerosol observations are effectively continued by VIIRS. However, the data records for other instruments, such as CALIPSO and MISR, are at greater risk.
34. Section 4.2.7, lines 901-904. However, errors in a slope derived from two or more spectral AOD measurements can be large. And if there are several modes in the column, interpretation of AE is not straightforward.
35. Section 4. Aside from data assimilation, are there other aerosol forecast applications, and if so, could their requirements be summarized or at least mentioned?
36. Section 5.1, line 951. The residence time of aerosols is "short" compared to… This matters for the discussion here. Transported aerosol can stay aloft for days, even exceeding a week, in the troposphere. Please clarify what is meant here.
37. Section 5.1, lines 957-959. However, spatial and usually temporal sampling by commercial aircraft tends to be highly skewed. You might elaborate on how the limitations affect application to aerosol forecasting.
38. Section 5.1. This is a good summary of network capabilities. It would be helpful for the purposes of the current paper to summarize the strengths and limitations as they relate to aerosol forecasting in particular, e.g., desired site locations, coincident meteorological observations, etc. (Many networks also have a strong climate or air quality focus.)
39. Section 5.2, line 1012. Perhaps, in one place in the paper, you could clarify what is meant by "high temporal resolution" for the applications under consideration, and elsewhere refer to that section for clarification. I get the impression that 3-hourly temporal resolution is desired for most of the applications considered here, but maybe not all.
40. Section 5.3, lines 1044-1048. This seems like a fairly comprehensive list. Are some species higher priority than others, perhaps at different locations? Generally for this section, are some quantities higher priority than others?