Author Response to Reviewer Comments:

Reviewer 1:

- **The reviewer wrote that the figures are very small and details are difficult to read in print version and asked for the labels to be increased in size.**
    - **Response:** we updated the figures to try and make them bigger and easier to read, particularly Figure 7 and 14 which are the central results of the paper. Many of the other figures were increased in size as well. We hope that this makes them more useful.
- **The reviewer asked to introduce the definition of ABF on page 6.**
    - **Response:** We added to line 198, "where γ_i is an empirical species-dependent exponent (anthropogenic/biogenic fine (ABF) ".
- **The reviewer asked to clarify the timeframe for appearance of the negative correlation in spring/summer months (page 11). Is this boreal spring/summer?**
    - **Response: We agree it is important to clarify the time frame. In order to address this, we added the months we are referring to in parentheses on line 383: "**Likewise, stronger European AOD and PW correlations are found in the summer months (JJA), in Eastern Asia in the winter season (DJF), and the Middle East in the fall (SON)." And also added on line 400-401 "During all seasons, negative correlations are found in the Sahel region in both AERONET and the NAAPS-RA with the negative relationships extending further northwards in the boreal spring and summer months."

Reviewer 2:

- **Comment on Page 1:** Some types were highlighted in the abstract.
    - **Response:** the highlighted typos were corrected in the abstract accordingly.
- **Comment on Page 1:** The final sentence of the abstract was highlighted and the reviewer wrote "This is also very important for retrievals of PM from space"
    - **Response:** we agree with this statement and updated the final sentence to reflect this: **"**Given these results, PW can be exploited in coupled aerosol and meteorology data assimilation for AOD and the collocation of aerosol and water vapor should be carefully taken into account when conducting particulate matter (PM) retrievals from space and in evaluating radiative impacts of aerosol, with the season and location in mind. " Also, given the word count limit, we went back through the abstract and updated the wording a little to maintain the word count.
- **Comment on Page 4 (line 143): the reviewer suggested putting a comma instead of "and" in this sentence.**
    - **Response:** We disagree here and kept "and" as aerosol microphysical and radiative properties are grouped together (i.e. aerosol microphysical properties and aerosol radiative properties).
- **Comment on Page 4 (Section 2.1.1 AERONET AOD): The reviewer asked if MAN data was included in the evaluation.**
    - **Response:** We did not include MAN data due to the nature of that data not fitting into our existing analysis, although we agree it would be a useful dataset. We added a sentence at the end of section 2.1.1 to clarify this point: **"**It should be noted that the AERONET-Maritime Aerosol Network (MAN) data is not included in this analysis as MAN data is shipborne and available on a periodic basis, and thus is not consistent with the long-term evaluation at fixed points that is conducted in this work."

- **Comment on Page 5 (Section 2.1.3 NAAPS Reanalysis):** The reviewer asked in AERONET AOD data is assimilated in the NAAPS-RA.
    - **Response**: It is not. I sentence to clarify this point was added to the manuscript on line 181: "AERONET AOD is not assimilated in the NAAPS-RA".
- **Comment on Page 9 (Section 3.1 Global Patterns of AOD-PW Correlation):** The reviewer suggested adding an additional figure in the appendix to support the region by region discussion.
    - **Response:** Since the regions discussed in this work are consistent with designated verification regions used in previously published work (Figure 4 in Lynch et al. 2016 and Figure 1 Rubin et al. 2016), we chose to add a reference to these papers on line 302-303: "A more in-depth discussion of the data by region, which is consistent with verification regions presented in Lynch et al. (2016) and Rubin et al. (2016), is below:"
- **Comment on Page 12 (Section 3.1 Global Patterns of AOD-PW Correlation):** The reviewer highlighted a sentence "While NAAPS and AERONET are in general agreement in the locations of negative correlations, this discrepancy is likely related to meso or small scale features that are not captured in a global, 1 degree model." And wrote "That is true, particularly for mountainous sites where the difference in model orography and the actual height of the station can be large"
    - **Response**: We took this as the reviewer agreeing with the point we were making. Just for clarification, we added to line 421: "While NAAPS and AERONET are in general agreement in the locations of negative correlations, this discrepancy is likely related to meso or small scale features, including orography, that are not captured in a global, 1 degree model. "
- **Comment on Page 14 (Section 3.3 Slope Evaluation):** The reviewer highlighted the sentence **"**This terrain/altitude influence is likely a factor in the discrepancies." And wrote "Definitely, it is hard to make meaningful model-obs comparisons at mountain sites due to coarse model resolution and differences in orography".
    - **Response:** We appreciate the reviewer agreeing with our point here. Nothing was changed.
- **Comment on Page 16 (Section 3.5 Vertical Evaluation of the AOD and PW Relationship):** The reviewer highlighted the sentence "This indicates that the model may be transporting too much dust aerosol and water vapor higher into the atmosphere and this transport is well correlated" and wrote "Is this supported by vertically resolved observations such as CALIPSO for aerosol extinction and MW sounders for PW? This is beyond the scope of the paper, but could be interesting for future study.
    - **Response:** We are currently using lidar and sounding data to evaluate these relationships vertically (we do see too much vertical diffusion in the model), but as the reviewer

commented, this is beyond the scope of the paper. We hope to show some of that work in future publications.

- **Comment on Page 17 (Section 3.6 Impact of Hygroscopic Growth): The reviewer highlighted the sentence "**In this highly correlated region, hygroscopic growth is expected to be a significant driver in AOD and PW relationships when dust is not the dominant aerosol type" and wrote "In urban areas this is extremely important as AOD is not a good proxy for dry PM".
    - **Response:** We agree with this point. The hygroscopic growth evaluation helps highlight areas where AOD is not a good proxy for dry PM. We didn't update anything at the highlighted sentence, but we did add a point regarding this in the conclusions, writing on line 812-813 "These findings are also valuable in identifying locations with potential for PM retrieval from space in which hygroscopic growth was found not to be an important factor in the AOD and PW relationship."
- **Comment on Page 22 (Conclusions):** The reviewer highlighted the sentence: "This indicates that PW is a good tracer for AOD, but not necessarily aerosol mass" and wrote "this is an extremely important point.
    - **Response:** We agree and appreciate the reviewer highlighting this as an important point in the paper. We added to this sentence "This indicates that PW is a good tracer for AOD, but not necessarily aerosol mass. This finding has relevance for data assimilation applications as well as PM retrievals." This point is also included in the abstract: "The importance of hygroscopic growth in these relationships indicates that PW is a useful tracer for AOD, or light extinction, but not necessarily as strongly for aerosol mass."