Responses to Reviewer 1:

The revised paper is an improvement over the previous version. There are still several items that need to be resolved before the paper is suitable for publication.

Line 420: Was the AMS run in a configuration so that Positive Matrix Factorization (PMF) could be performed to derive more speciated organic species which could be compared with the model? Typically, PMF can differentiate between biomass burning aerosols and other organics that may be of anthropogenic or biogenic origin.

Response: This is a very good point. m/z tracer data are available from the AMS for this campaign and this could be used to try and identify sources of the organic aerosol. We did not do this for our study, but the text has been changed to read:

Lines 418 – 421: "For example, the AMS can quantify mass concentrations of organic tracer species, but it is difficult to determine the fraction of these species associated with smoke versus the fraction associated with anthropogenic/biogenic emissions (that NAAPS-RA would place in the ABF category)."

Line 462. The statement regarding uncertainties in optical instruments is true, but some citations would be useful here.

Response: We agree this statement needs to be backed up with citations. The sentences following the line the reviewer is discussing (Lines 461 – 462: "Additionally, there are known challenges in using OPCs [such as the FCDP] to accurately quantify coarse particle mass concentrations.") elaborate on the challenges and include citations (lines 465 – 468).

Note that we did not make any changes to address this comment. We feel that the following sentences we provided in the previously submitted manuscript are sufficient to back up the statement about uncertainties in optical instruments:

Lines 464 – 469: "For example, Reid et al. (2003, 2006) found coarse mode OPCs to overestimate the size of coarse particles (e.g., sea salt and dust), while other works have found OPCs to underestimate coarse mass concentrations (Kulkarni and Baron, 2011; Burkart et al., 2010)."

Line 482-490. It would be useful to add a sentence or two regarding the scatter between observed and simulated AOT for small AOT. While the correlation is good and the overall bias is small, there is still a bit of scatter for small AOT so that the model could be a factor of 2 – 3 times of at a particular time.

Response: Great suggestion. We have added the following sentence to the manuscript:

Lines 482 – 484: "However, it is worth noting that there is scatter between observed and simulated AOT at low AOT where NAAPS-RA can be off by a factor of two to three in some cases."

Line 518-535: Another reason why AOT does not improve when using observed RH is that the simulated mass and/or composition may be off which is not mentioned here. The errors in mass and composition (or treatment of composition) will also affect water uptake.

Response: This is an excellent point. We have added the following sentences to include the reviewer's suggestion.

Lines 521 – 524: "First, errors in simulated mass concentrations and/or hygroscopicity for each of the four species will affect how NAAPS-RA simulates water uptake. These types of errors are almost guaranteed to prevent extinction agreement with observations even when NAAPS-RA is using corrected RH profiles."

Figure 3. Include text in b), c) and d) for the height ranges, similar to what was done in Figure 2.

Response: We have updated this figure to include information about the altitude range on each panel. The updated version of Fig. 3 appears like this:

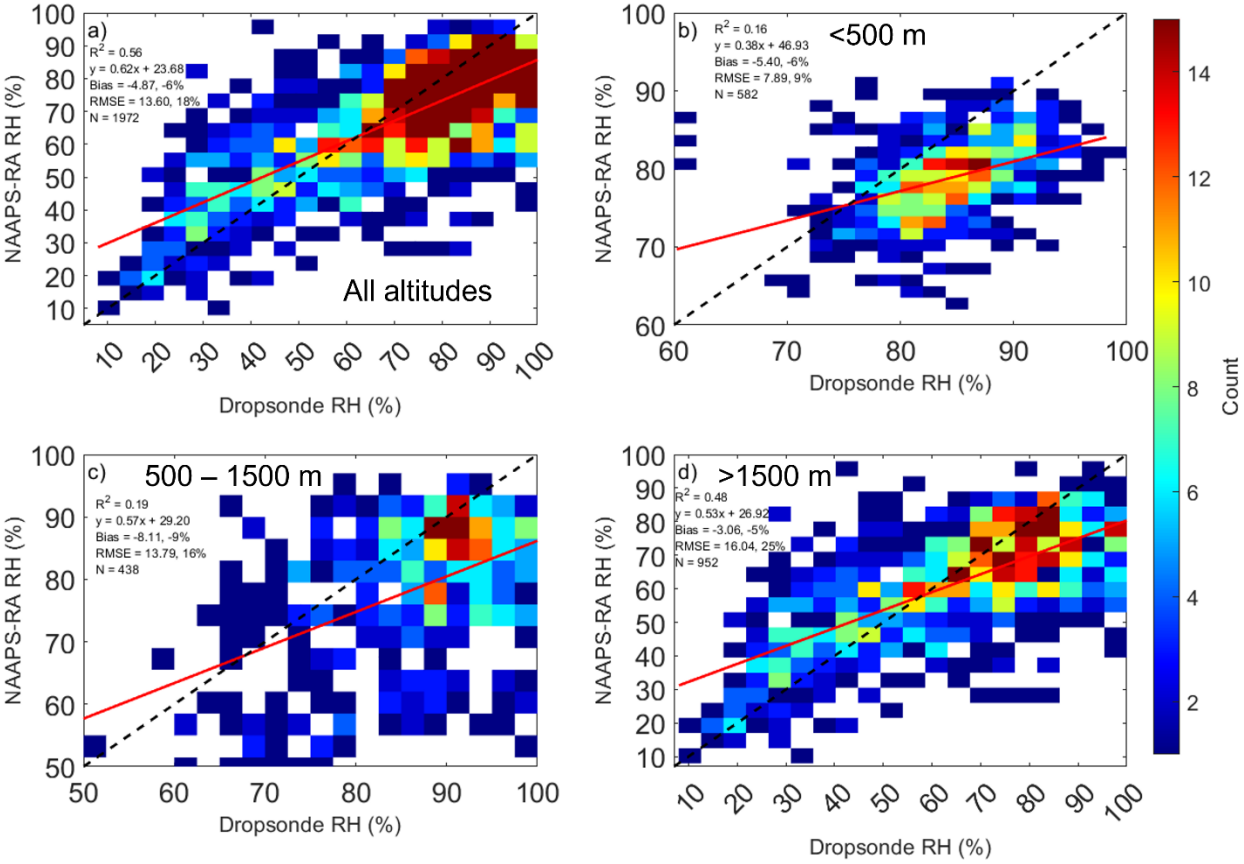

Fig. 3

Line 565: The authors say that mass concentrations are an issue (which is true), but that there is no data to examine this. However, this seems to contradict other statements (line 115 says

extensive data are available, mention AMS in other places, and compare observed and simulated mass later in Figures 7-10).

Response: Actually, line 115 does not say that extensive data are available. Lines 115 - 119 say:

"Extensive profiles of observed speciated particle mass concentrations, the particle hygroscopic growth parameter (γ), and RH collocated with HSRL-2 retrievals of extinction and AOT **are required** to thoroughly evaluate the model's outputs and identify sources of error. **Such collocated profiles of mass concentrations, γ values, and HSRL-2 retrievals are limited** for this campaign."

So we are acknowledging that we need the vertical profiles to robustly evaluate NAAPS-RA but we are saying that we do not have the collocated profiles of all the variables we need.

We do have AMS data, but that does not mean we have full vertical profiles at the same locations where we have full vertical profiles of extinction, RH, and gamma. That is the problem. We explain on lines 125 – 130 the assumptions we make to conduct the mass comparison we present for each case study. These assumptions are not ideal, and it is very true indeed that a lack of collocated vertical profiles of all the necessary variables was a big problem in this study.

Note that we have not made any changes to the manuscript regarding this comment.

The end of 3.3: Perhaps a better transition is to mention the issue of mass concentrations which have not been examined up to this point, but some evaluation will be done in the subsequent section.

Response: Good suggestion.

Lines 601 – 608 have been added:

"As mentioned above, NAAPS-RA extinction sensitivity to changes in RH also depends on speciated particle mass concentrations and/or the hygroscopicity assigned to each species. Sufficient data are not available to evaluate relationships between these parameters and extinction agreement between NAAPS-RA and HSRL-2 retrievals for the entire campaign. However, we confine our analysis to the ML (assumed to have homogeneous particle microphysical properties) for four case studies in the following section to provide some assessment of simulated hygroscopicity and particle mass concentrations."

Figures 4 and 5: As with the previous figures, it would be helpful to add labels of height ranges in the figure panels.

Response: We agree. We have updated these figures to include information about the altitude ranges.

Figs. 4, 5, and 6 now appears like this, respectively:

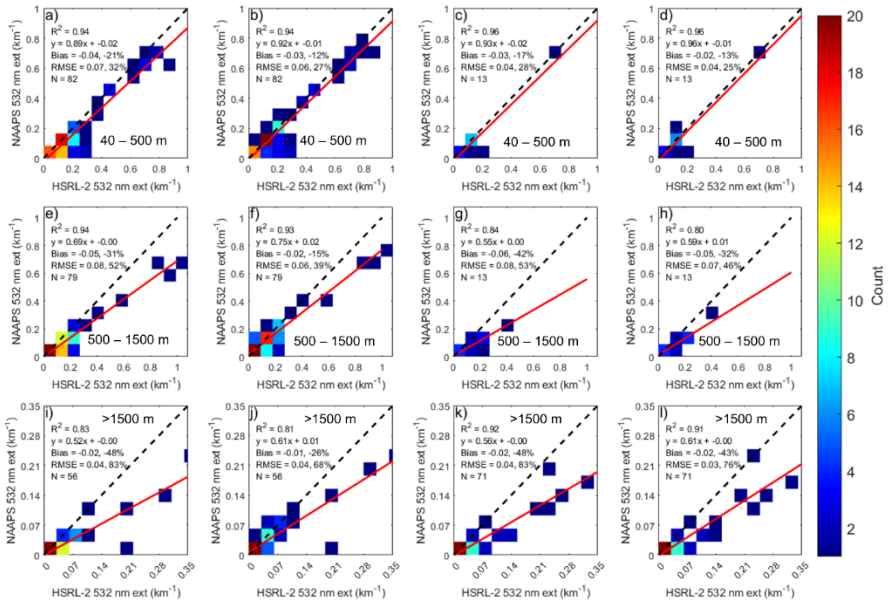
a)
R² = 0.94
y = 0.89x + -0.02
Bias = -0.04, -21%
RMSE = 0.07, 32%
N = 82
40 – 500 m
b)
R² = 0.94
y = 0.92x + -0.01
Bias = -0.03, -12%
RMSE = 0.06, 27%
N = 82
40 – 500 m
c)
R² = 0.96
y = 0.93x + -0.02
Bias = -0.03, -17%
RMSE = 0.04, 28%
N = 13
40 – 500 m
d)
R² = 0.96
y = 0.96x + -0.01
Bias = -0.02, -13%
RMSE = 0.04, 25%
N = 13
40 – 500 m
e)
R² = 0.94
y = 0.69x + -0.00
Bias = -0.05, -31%
RMSE = 0.08, 52%
N = 79
500 – 1500 m
f)
R² = 0.93
y = 0.75x + 0.02
Bias = -0.02, -15%
RMSE = 0.06, 39%
N = 79
500 – 1500 m
g)
R² = 0.84
y = 0.55x + 0.00
Bias = -0.06, -42%
RMSE = 0.08, 53%
N = 13
500 – 1500 m
h)
R² = 0.80
y = 0.59x + 0.01
Bias = -0.05, -32%
RMSE = 0.07, 46%
N = 13
500 – 1500 m
i)
R² = 0.83
y = 0.52x + -0.00
Bias = -0.02, -48%
RMSE = 0.04, 83%
N = 56
>1500 m
j)
R² = 0.81
y = 0.61x + 0.01
Bias = -0.01, -26%
RMSE = 0.04, 68%
N = 56
>1500 m
k)
R² = 0.92
y = 0.56x + -0.00
Bias = -0.02, -48%
RMSE = 0.04, 83%
N = 71
>1500 m
l)
R² = 0.91
y = 0.61x + -0.00
Bias = -0.02, -43%
RMSE = 0.03, 76%
N = 71
>1500 m
NAAPS 532 nm ext (km⁻¹)
HSRL-2 532 nm ext (km⁻¹)
Count

Fig. 4

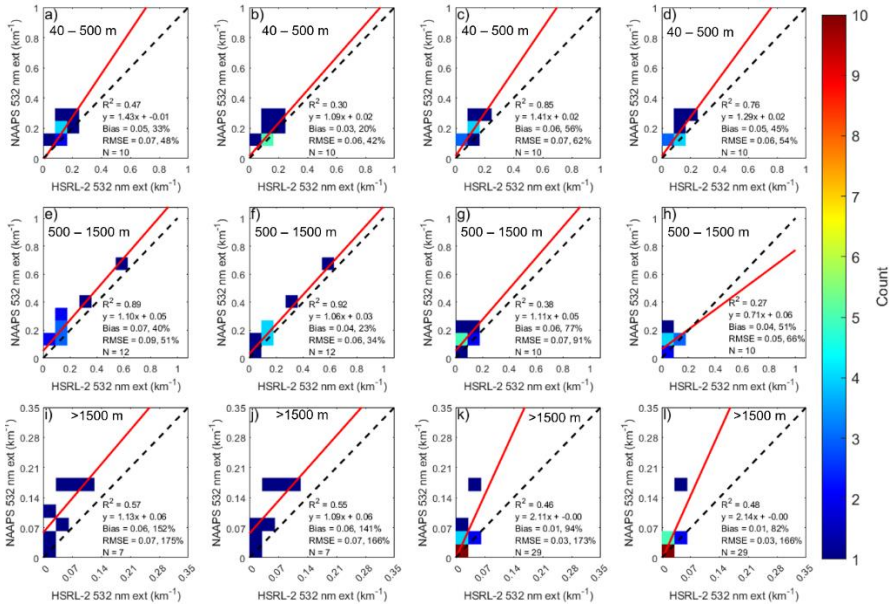
a)
40 – 500 m
R² = 0.47
y = 1.43x + -0.01
Bias = 0.05, 33%
RMSE = 0.07, 48%
N = 10
b)
40 – 500 m
R² = 0.30
y = 1.09x + 0.02
Bias = 0.03, 20%
RMSE = 0.06, 42%
N = 10
c)
40 – 500 m
R² = 0.85
y = 1.41x + 0.02
Bias = 0.06, 56%
RMSE = 0.07, 62%
N = 10
d)
40 – 500 m
R² = 0.76
y = 1.29x + 0.02
Bias = 0.05, 45%
RMSE = 0.06, 54%
N = 10
e)
500 – 1500 m
R² = 0.89
y = 1.10x + 0.05
Bias = 0.07, 40%
RMSE = 0.09, 51%
N = 12
f)
500 – 1500 m
R² = 0.92
y = 1.06x + 0.03
Bias = 0.04, 23%
RMSE = 0.06, 34%
N = 12
g)
500 – 1500 m
R² = 0.38
y = 1.11x + 0.05
Bias = 0.06, 77%
RMSE = 0.07, 91%
N = 10
h)
500 – 1500 m
R² = 0.27
y = 0.71x + 0.06
Bias = 0.04, 51%
RMSE = 0.05, 66%
N = 10
i)
>1500 m
R² = 0.57
y = 1.13x + 0.06
Bias = 0.06, 152%
RMSE = 0.07, 175%
N = 7
j)
>1500 m
R² = 0.55
y = 1.09x + 0.06
Bias = 0.06, 141%
RMSE = 0.07, 166%
N = 7
k)
>1500 m
R² = 0.46
y = 2.11x + -0.00
Bias = 0.01, 94%
RMSE = 0.03, 173%
N = 29
l)
>1500 m
R² = 0.48
y = 2.14x + -0.00
Bias = 0.01, 82%
RMSE = 0.03, 166%
N = 29
NAAPS 532 nm ext (km⁻¹)
HSRL-2 532 nm ext (km⁻¹)
Count

Fig. 5

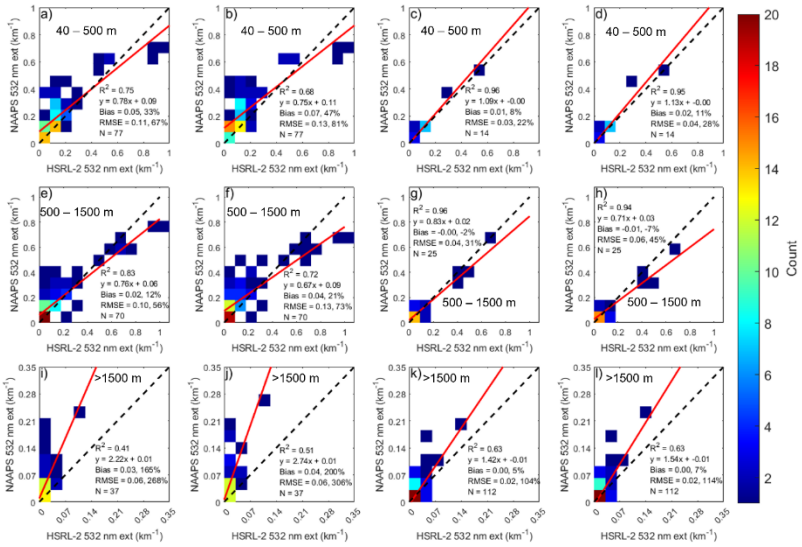

Fig. 6

Line 639: now state mass evaluation, but where is this done?

Response: We evaluate simulated and observed mass concentrations in the boundary layer for each case study. This is described in Section 2.6.3 and is mentioned in other parts of the paper (e.g., lines 125 – 132).

Note that we did not make any changes regarding this comment. We simply refer the reviewer to the section where the mass evaluation is discussed.

Line 628-644: It would be useful to point to each sub-panel that is being discussed, rather than just solely citing the entire figure at the beginning of the paragraph.

Response: We understand that this would be helpful since there is a lot going on in the case study figures. However, there are many multi-paneled figures throughout the paper, and we do not want to create "alphabet soup" by referencing each panel (e.g., Fig. 4 has 12 panels). To maintain consistency throughout the paper, we simply refer to a figure once and not to each panel.

Thus, we did not make any changes regarding this comment.

Line 636-372: The authors say that increasing the mass will improve extinction, but Figure 7d already shows that the model mass concentrations are slightly too high. So increasing the simulated mass will make the agreement poorer, while the simulated extinction improves.

Response: We put very little stock in our mass concentration comparison because the methods to derive in situ particle mass concentrations are subject to uncertainty. On the lines the reviewer is referencing, we focus primarily on what would need to happen to achieve *extinction agreement*. In this case, NAAPS-RA underestimates extinction when in situ RHs and gamma values are used. The only thing left to do to achieve extinction agreement is to increase dry extinction,

which is either an increase in particle mass concentrations or an increase in mass scattering/absorption efficiency.

Because we focus the analysis on *extinction* agreement in this section, the text is correct as written and we did not make any changes to address this comment.

Figure 7a. Gamma in legend is very, very small and barely legible.

Response: We agree. The font for this legend is now larger in Figs. 7 – 10. These figures now appear as follows:

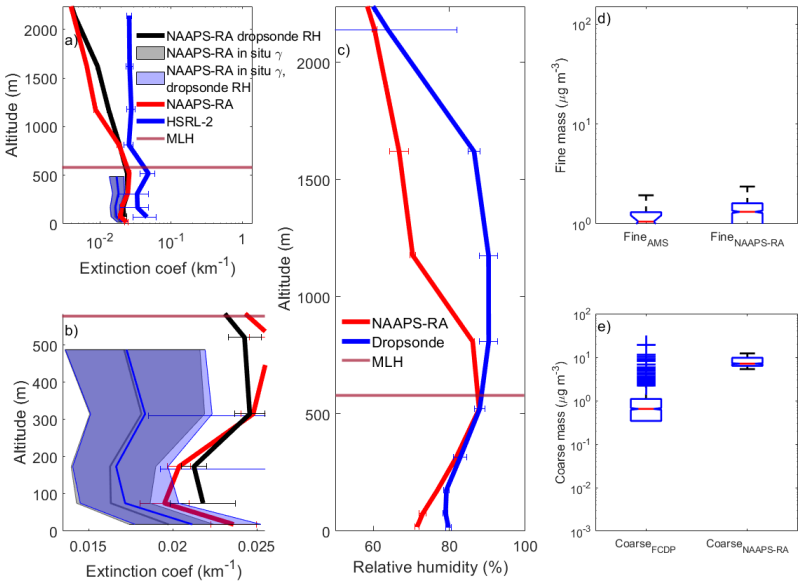

Fig. 7

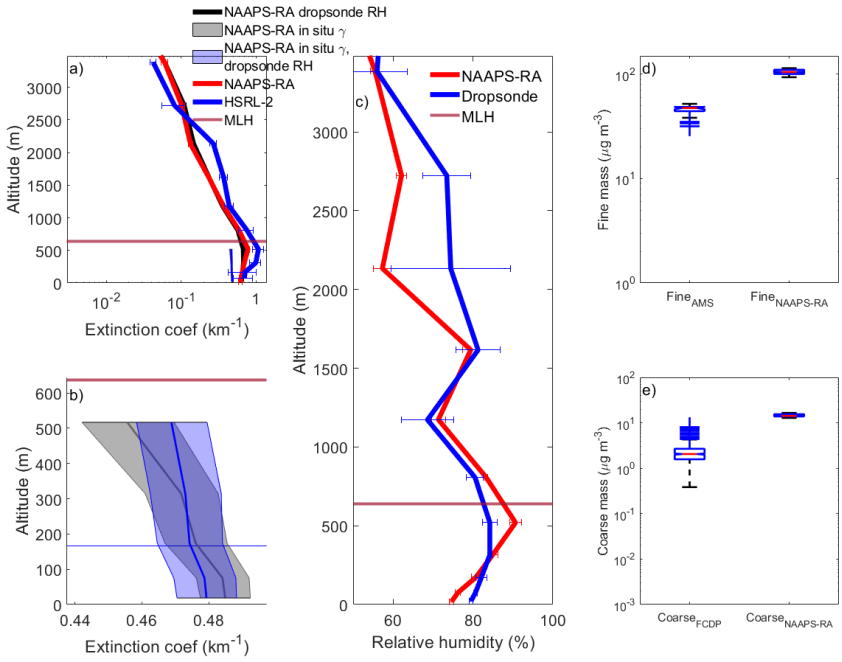
NAAPS-RA dropsonde RH
NAAPS-RA in situ γ
NAAPS-RA in situ γ, dropsonde RH
NAAPS-RA
HSRL-2
MLH
a)
b)
c)
d)
e)
Altitude (m)
Extinction coef (km-1)
Relative humidity (%)
NAAPS-RA
Dropsonde
MLH
Fine mass (μg m-3)
Coarse mass (μg m-3)
FineAMS
FineNAAPS-RA
CoarseFCDP
CoarseNAAPS-RA

Fig. 8

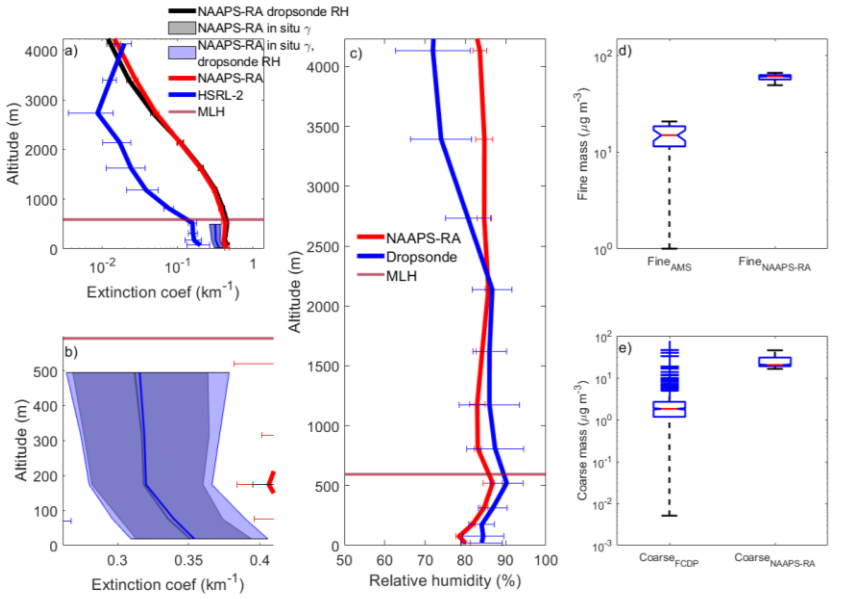
NAAPS-RA dropsonde RH
NAAPS-RA in situ γ
NAAPS-RA in situ γ, dropsonde RH
NAAPS-RA
HSRL-2
MLH
a)
b)
c)
d)
e)
Altitude (m)
Extinction coef (km-1)
Relative humidity (%)
NAAPS-RA
Dropsonde
MLH
Fine mass (μg m-3)
Coarse mass (μg m-3)
FineAMS
FineNAAPS-RA
CoarseFCDP
CoarseNAAPS-RA

Fig. 9

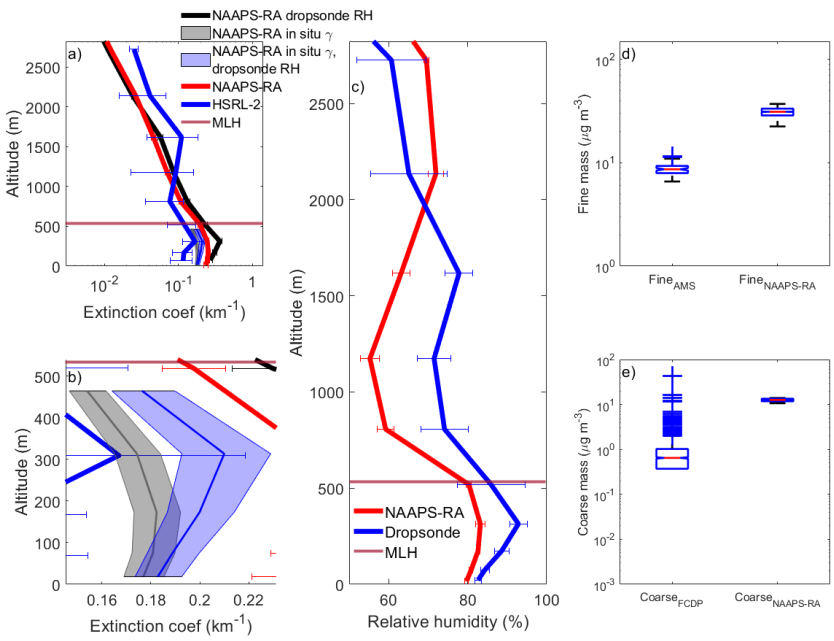

Fig. 10

Figure 7d and 7e: why not pick a more appropriate y axis range to compare the mass? I assume that the authors choose to use the same range as Figures 8-10, but this does not seem necessary since the figures are not side-by-side.

Response: We understand the point the reviewer is making. However, we wish to keep the axes the way they are so that readers can quickly flip between case studies to compare different air masses without having to readjust their eyes to a new y scale.

We did not make any changes in the manuscript regarding this comment.

Line 688: Again the author hypothesize that NAAPS-RA is underestimating mass, but panel d) seems to indicate the model is already too high.

Response: Similar to the response above, the text in this section is focused on the types of changes in NAAPS-RA that would lead to *extinction agreement*. Panel a) shows that when in situ RHs and gamma values are used, NAAPS-RA extinction is still too low. This necessary increase in modeled extinction could come from increasing particle mass concentrations and/or increasing mass scattering/absorption efficiencies. The in situ mass concentrations shown in panel d) are subject to great uncertainty so we cannot say for sure that NAAPS-RA is overestimating mass concentrations for this case study.

Based on the explanation above, the text is correct as is and no changes were made regarding this comment.

At the end of each case section, the authors state that more work is needed to understand the sources of uncertainties. This gets repetitive. Instead, perhaps grouping this though together into a short final section 3.5 that discusses common issues among the 4 cases.

Response: We understand the reviewer's perspective. We do not mean for it to sound repetitive, but each of the four case studies are subject to the same uncertainty when deriving in situ fine and coarse particle mass concentrations. It is true that we could regroup the text, but we choose to leave it as is so that each case study can stand alone.

Thus, we did not make any changes regarding this comment.

Line 790-800. Maybe also point out to the limit of the NAAPS-RA model when comparing with observations. The authors allude to this earlier when describing the simple representation of aerosol composition in the model.

Response: Thank you for the great suggestion. We have added the following text to the manuscript:

Lines 806 – 807: "However, it is likely that the model's simple representation of speciated particle composition, hygroscopicity, and size contribute to these errors in some part."

Response to Reviewer 2:

We have accepted all edits presented in this reviewer's attached PDF. Thank you for the careful edit you gave this paper.

Responses to Reviewer 3:

The authors have considered many of my suggestions from the first round of reviews and I am very happy to see that the methodology for data analysis and the presentation of their work has much improved. Nevertheless, further revisions are needed to get this manuscript ready for publication.
Here are some issues that should be addressed:
- Do you really need the case studies to make your point? As stated during my first review, they still leave more open questions then they answer. It is very unsatisfying for the reader that all case studies end with "this will be addressed in future work" after all the issues are described. It might be more elegant to trim the case studies to the AOT/extinction comparison and summarize the problems in an outlook section?

Response: We understand why the reviewer has these feelings about the case studies as it is mildly frustrating that we do not have the data to answer all the questions we would like to answer. However, we wish to leave the case studies as we feel they provide valuable insight about individual air masses that the bulk analyses cannot reveal. For example, the case studies allow us to see that NAAPS-RA overestimates the hygroscopicity of an air mass transported from East Asia (called “Asian pollution” in the manuscript) as well as biomass burning particles transported from countries in the Maritime Continent to the southwest of the Philippines. These case studies will (and have been) important cases examined in independent projects and we feel keeping these in the paper will help other investigators in their separate (but potentially related) pursuits linked to these specific flights we look at.

We have not made any changes to the manuscript regarding this comment.

- Please shorten the Abstract and the last three paragraphs of the Introduction.

Response: We apologize that this reviewer finds these sections too long. We prefer to leave them at their current lengths as we feel they portray necessary and relevant information throughout.

We have not made any changes to the manuscript regarding this comment.

- I find the use of grid very confusing. It might be better to replace grid with grid CELL.

Response: Every instance where we used “grid” has now been replaced with “grid cell.”

- x, z, z are not needed in the equations

Response: We feel that these are necessary to describe the 3-D space that NAAPS-RA simulates optical properties. Providing/mentioning z in the equations is especially useful as it facilitates a discussion about how NAAPS-RA reports parameters at the midpoint of each pressure layer. This is relevant to how we grouped and averaged our remotely-sensed and in situ data to match the vertical resolution of the model.

We have not made any changes to the manuscript regarding this comment.

- line 299: in-situ data or remote-sensing data?

Response: Good catch.

Lines 299 – 300 have been changed to read: "Remotely-sensed data were averaged first vertically and then horizontally to match the resolution of the NAAPS-RA model."

- I am somewhat concerned about Figures 4 to 6. Stratifying the data into so many categories leaves you with cases with as little as 10 data points. Please be careful about drawing conclusions from such small samples. This should be commented in the text.

Response: We feel that we did already address how low sample sizes should be taken into consideration when viewing these figures (particularly Fig. 5 where many sample sizes are around 10).

Lines 572 – 575 state, "Note that different sample sizes should be taken into consideration when comparing R^2 values between these categories (e.g., there is a relatively low number of points in the second category, i.e., pressure layers where NAAPS-RA overestimates both extinction and RH)."

We have not changed the manuscript regarding this comment because we feel we have already addressed the reviewers concern in the original text.