Reviewer #2

We highly appreciate the valuable comments on our manuscript and the great editing. The comments were quite helpful and we have incorporated them when possible into the revised paper. We hope that you will be satisfied with our responses and the corresponding revisions for the original manuscript. Please find below the comments/suggestions (bold blue color) and our responses (red color, with manuscript changes indicated in red italic).

P4953, L5-6. AOD ang g are definitely important but I would not forget SSA when you evaluate the importance of aerosol optical parameters

We definitely agree with the reviewer and appreciate his/her pointing out our inadvertent omission of SSA. The revised manuscript includes the corrected statement.

(page 3-4, lines 89-94): *"Moreover, it is well-known that the aerosol optical depth (τ_a), asymmetry factor (g) and single scattering albedo are the most important aerosol optical parameters for numerous climate-relevant applications, including the Earth's radiation budget (e.g., McComiskey et al., 2008). The role of single scattering albedo is more pronounced for highly absorbing aerosols (e.g., Bond et al., 2009 and references therein)."*

P4954, L20. Why not talk about submicron and supermicron particles?

Particles with the moderate sizes (about 0.5-1 μm) represent a fraction of submicron particles. This fraction is modified during shifting of the APS spectra. In contrast, the fraction of particles with small sizes (<0.5 μm) represents the SMPS spectra and remains unchanged. This is the main reason why we use the more specific term "particles with the moderate sizes (about 0.5-1 μm)" instead of more general expression "submicron particles" for a case considered here. The terms "particles with large (>1 μm) sizes" and "supermicron particles" are interchangeable.

In response to this comment, we replaced "moderate sizes (0.5-0.7 μm)" with "moderate sizes (about 0.5-1 μm)" (page 5, line 131).

P4955, L5- Give a more detailed description of how the modeled scattering and backscatter fractions were calculated. Which code was used? What was the resolution of the angular integration?

This is great suggestion and we have added a new appendix (Appendix A) in response. In particular, the revised manuscript includes the requested details clarification.

(page 19, lines 504-505 and 509-512): *"The calculations of aerosol sub-micron and sub-10-micron optical properties (σ_s and β) are based on Mie codes developed by Barber and Hill (1990)…. To resolve accurately the angular distribution of the scattered light, variable angular resolution is applied: from 0.05° (near-forward direction) to 1° (near-backward direction). Selection of the angular resolution is driven mostly by a trade-off between the speed and accuracy of Mie calculations."*

P4954-4955 In the whole method description, give formulas where the relationship between scattering and density and backscatter fraction and density are explicitly given. It would make it easier to understand the method.

We added the corresponding equations (please see Appendix A, Equations (A1) and (A2)).

P4955 L12 it is written "the near orthogonal isolines". I don't understand this. Orthogonality is the relation of two lines at right angles (90°) to one another. Both in fig1b&1c I see nearly parallel isolines.
To clarify the intent of Figures 1b and 1c, we remove this confusing phrase and changed the wording.

(page 6, lines 150-153): *"The $\sigma_{s,\text{mod}}$ isolines increase as the effective density increases (Figure 1b). The opposite is true for β_{mod} isolines (Figure 1c). The observed opposite trends of $\sigma_{s,\text{mod}}$ isolines and β_{mod} isolines (increase versus decrease) suggest that a unique solution for n and ρ can be obtained."*

P4956, L12-13 In this sensitivity study, is the size distribution time series a real measured one from some selected day? Or all simulated, both SMPS and APS?
The clarification is added to the revised manuscript:

(page 7, lines 182-185): *"To illustrate the performance of our method, we devise a sensitivity study using the SMPS-APS size distributions. In particular, results shown in Figures 2 and 3 represent the SMPS-APS size distributions measured during a selected day (July 17). We treat assumed aerosol characteristics, n_{mod} and ρ_{mod}, as reference values."*

P4957, L2 Here you just say that there are diurnal cycles. What has been observed? Explain observations and the reasoning for using the sinusoidal variations of both n and density in the simulation.
We include the required explanation. Here is the revised section:

(pages 7-8, lines 193-202): *"Although microphysical and chemical properties of aerosol do not exhibit systematic diurnal changes (section 4), we assume that temporal changes of the reference values (n_{mod} and ρ_{mod}) can be approximated by simple functions with sine terms: … The focus of these simulations is to reproduce large temporal changes of observed aerosol optical properties (section 4) using a simple representation. A sinusoidal representation with properly selected amplitudes is one of several potential ways to capture the observed variability. The amplitudes ρ_0 and n_0 are selected is such way that the span of modeled optical properties (Figure 2a,b) matches roughly the wide range of optical properties observed during the TCAP study. Note that in our sensitivity study we generate "observations" for a 24-hour period."*

P4961, L17 "For these events, the relative contribution of large (1 µm < D_p < 10µm) particles to the light scattering can be substantial." The word "can" is strange here – the contribution of large particles IS substantial if f < 0.5
Agree. The word "can be" is replaced by "is" (page 12, line 318).

Fig 2e&f. I don't understand. If the "observation" is purely modeled from the size distribution, what is the difference between the "original" and "retrieved" characteristics? Shouldn't that be zero?

The clarification is added.

(page 9, lines 237-241): *"On average, these differences (Δ_ρ and Δ_n) should be zero for "error-free" conditions (α=0). The averaged difference found under "error-free" conditions is close to zero (Figure 2e,f). The small deviation (< 0.01) of this difference from zero suggests that our simple retrieval reproduces the "original" characteristics reasonably well under favorable "error-free" conditions. Under unfavorable "noisy" conditions, differences …."*

Fig 3. Why were scattering coefficients and backscatter fractions not calculated at the same density?

We do not fully understand your question. Hopefully, the added explanation (please see below) together with the related clarification for Figure 1 (we repeat our previous reply for your convenience) addresses the apparent confusion over our approach.

(page 6, lines 150-153): *"The $\sigma_{s,\mathrm{mod}}$ isolines increase as the effective density increases (Figure 1b). The opposite is true for β_{mod} isolines (Figure 1c). The observed opposite trends of $\sigma_{s,\mathrm{mod}}$ isolines and β_{mod} isolines (increase versus decrease) suggest that a unique solution for n and ρ can be obtained."*

(Caption of Figure 3): *"Note that the crossing isolines are related to the 2D diagrams of $\sigma_{s,\mathrm{mod}}$ and β_{mod} (e.g., Figure 1b,c) calculated for assumed n and ρ."*