Dear Editor:

Thank you and all the reviewers for the quite constructive and helpful comments! All these comments raised by the referees have been explicitly replied point by point and incorporated into the revision. All authors consent to the revisions and the responses. Thank you very much for your attention and consideration.

Sincerely Yours

Chunsheng Zhao

Responses to anonymous referee #1

Comment: 1. Lines 265-271: I am confused which dataset the authors used for training the random forest model. It seems that the authors used the measurements of TSI 3563, however, the campaign F6 used Aurora 3000 (table 2). Meanwhile, the authors claimed that good agreement between measured σsp and that calculated based on measured PNSD and BC with Mie theory was found in the campaigns F1, F4, F5 and F6 (section 3.1). so why you use dataset from field campaigns F1 to F4 and F6. Please check and explain.

Response: Thanks for your comment. To avoid that the measurements uncertainties are involved in the training processes of the random forest model. Datasets of PNSD and BC from field campaigns F1 to F4 and F6 are used to calculate $V_a(\text{dry})$ and simulate six optical parameters corresponding to measurements of TSI 3563. Calculated $V_a(\text{dry})$ and simulated six optical properties are then used as inputs for training the random forest model. This process is independent of simultaneously measured aerosol optical properties from the nephelometer, and therefore not relevant with what nephelometer has been used. The reason that datasets of PNSD and BC from field campaign F6 are also used, is that we want the trained random forest model can cover different aerosol loadings as much as possible. Also, the closure results between modelled and measured σ_{sp} do not affect whether the PNSD and BC should be used in the training process. However, the closure results are important for deciding if the simultaneously measured PNSD and aerosol optical properties (scattering and backscattering coefficients) should be used for validating the machine learning model.

Comment: 2. Figure 5. This figure shows the schematic diagram of step 1 of the proposed ALWC calculation method. Is it possible to add the information of step 2 in this figure?

Response: Thanks for your comment. This figure shows how to train the random forest model. It is difficult to add the information of step 2 in this figure. However, we think it is important to show the flowchart of calculating the ambient aerosol liquid water contents based on measurements of the humidified nephelometer system and this flowchart is shown in Fig.8 of the revised manuscript.

Comment: Line 192: diameter

Response: Thanks for your comment, we have revised this word.

Comment: Line 196: equation (5)? I think it should be equation (1).

Response: Thanks for your comment. We have revised equation (5) to equation (1)

Comment: Line 344: Figure 7 came first than figure 6.

Response: Thanks for your comment. This problem is solved by adding a figure in the supplement which describes the used average size-resolved κ distribution.

Comment: Figure 8: please add units.

Response: Thanks for your comment. The unit of $V_a(\mathrm{dry})$ is added in the figure caption.

Figure 9: I think the unit of ALWC is wrong, please revise.

Response: Thanks for your comment. The unit of ALWC is revised.

Responses to anonymous referee #2

Comment: Dust is pervasive in the NCP. If not from the Loess regions, then roads and construction dust are present. This may be a factor influencing differences between the PM2.5 and PM10 aerosol in this study.

Response: Thanks for your comment. In Sect.4.4 of the revised manuscript, we emphasized that cautions should be exercised if using the proposed method to estimate the ALWC when the air mass is significantly influenced by sea salt or dust.

Comment: You need to stress in the Introduction that this study uses data from multiple sites to characterize a regional aerosol. Otherwise, mixing data from multiple sites over different time periods would give erroneous results. The quantity of data from multiple sites used in the machine learning code is large enough to infer the results apply to a larger data population and region.

Response: Thanks for your comment. The following sentence is added in the last paragraph of the introduction of revised manuscript: "In this study, datasets of PNSD and BC measured from multiple sites are used in the machine learning model to characterize a regional aerosol and these datasets have covered a wide range of aerosol loadings."

Comment: Title: Change "contents" to "content"

Response: Thanks for your comment. We have revised the title accordingly.

Comment: Page1: Change “so far” to “before now”

Response: Thanks for your comment. We have revised the abstract accordingly.

Comment: Page 9, line 182: Did you mean “either or” instead of “neither nor” ?

Response: Thanks for your comment. We mean “either or”, and the manuscript is revised accordingly.

Comment: Line 183… view of this, Qscat at 550 nm, as a function of particle diameter for four types of aerosol particles, is simulated…

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Page 15, line 300: “Kelvin”

Response: Thanks for your comment. We have revised this word accordingly!

Comment: Lines 302-304: You show in Figure 7a a large size-dependence to k with aerosol size. Use of a single k volume parameter doesn’t assume a constant k value with particles size. Rather it implies that a kappa(volume) can be expressed as a single value that perhaps is proportional to a weighted average of the size-dependent kappa (diameter) values. Such that $k\ vol = w * k(D)$. This assumption allows Vg(RH) to be expressed as a linear function.

Response: Thanks for your comment. The sentence “which means that if κ values of aerosol particles of different sizes are the same” is rewrote as “If a constant κ which represents the overall aerosol hygroscopicity of ambient aerosol particles, is used as the κ of different particle sizes”

Comment: Page 16, line324: replace “consolidate” with “validate”

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Page 18, lines364-366: rewrite “Figure 6 shows the influence of aerosol size and chemistry on Rvf. For Angstrom exponents less than ~1.1, Rvf varies strongly with ksca. However for Angstrom exponent values greater than ~1.1, the Rvf relative standard deviation exhibits a higher variability with the Angstrom exponent; thus showing the sensitivity of Rvf to changes in aerosol size for small particles.”

Response: Thanks for your comment. This part is revised as “Simulated values of R_{Vf} range from 0.8 to 1.7 with an average of 1.2. Overall, R_{Vf} value is lower when value of Ångstr$\ddot{o}$m exponent is larger. The percentile value of standard deviation of R_{Vf} values within each grid divided by its average is shown in Fig.6b. In most cases, these percentile values are less than 10% (about 90%) which demonstrates that R_{Vf} varies little within each grid shown in Fig.6a. Figure 6 shows the influence of aerosol size and chemistry on R_{Vf}. For Ångstr$\ddot{o}$m exponent less than ~1.1, R_{Vf} varies strongly with κ_{sca}. However, for Ångstr$\ddot{o}$m exponent values greater than ~1.1, the R_{Vf} relative standard deviation exhibits a higher variability with the Ångstr$\ddot{o}$m exponent. Thus, showing the sensitivity of R_{Vf} to changes in aerosol size for small particles. In general, results shown in Fig.6 imply that results of Fig.6a can serve as a look up table to estimate R_{Vf} and thereby κ_{Vf}, such that these values can be directly predicted from measurements of a three-wavelength humidified nephelometer system."

Comment: Line 370:rewrite " ..thereby kf , such that these values can be directly…" As a note, the high variability of Rvf at high Angstrom exponents may result from differences between monomodal and bimodal size distributions.

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Line 379: rewrite "… is highly variable yet has no apparent correlation with aerosol loading."

Response: Thanks for your comment. We have revised the manuscript accordingly.

Comment: Page 23, line 478: rewrite "On average, when ambient …"

Response: Thanks for your comment. We have revised the manuscript accordingly.