Response to Anonymous Referee #3

General Comments:

The authors describe a novel method to correct measurement errors which are inherent to the use of current commercial integrating nephelometers. This method based on machine learning sounds attractive since it is said not to "need additional observation data". Additional data are actually needed during the machine learning phase though. The scope of applicability of the relationship between the correction factor (CF) and the two selected variables derived from the nephelometer data (the scattering Angstrom exponent, SAE, and the hemispheric backscatter fraction, HBF) is a key question that is not addressed in the manuscript. In particular, HBF is said to depend on externally mixed fraction of black carbon (Rext), while it depends also on the particle number size distribution (PNSD). Can it be demonstrated that the rule learnt by the machine to determine CF will apply at a location and/or time where different Rext and PNSD combinations lead to the same HBF for the ensemble of the aerosol? Can it be said anything about the applicability of the CF determination method described in the manuscript to nephelometer measurements performed at much less polluted locations? Other missing elements as well as the overall organisation of the manuscript make it generally quite obscure, as described below.

Response: Thank you very much for your review of our manuscript. Your comments were very helpful and constructive for improving the work's coherence and logic. It is true that we have used the particle number size distribution and BC data during the machine learning phase, but the trained random forest models are saved for researchers' further use. That is to say, when applying this method, others could use the models to directly correct the nephelometer without additional in-situ measurements. The results of Fig.3 show that there can be possible relationships between CF and the two selected parameters (SAE and HBF). Since the three parameters all relate to particle size, the machine learning method is used to derive their relationships. HBF is calculated by the ratio of backscattering to total scattering, and then Rext is derived from the inversion of HBF: Only when the difference between nephelometer calculated HBF and the Rext-assumed HBF is minimal can we obtain the Rext value (Ma et al., 2012). Therefore, it is not accurate to state that different Rext and PNSD combinations lead to the same HBF. Even if this is the case, our machine learning method still works. The field sites we choose are all located in the Northern China Plain (NCP) where pollution is often occurred. The new Fig.2 shows that our field data can represent the background aerosol properties in NCP. Below we will respond to your comments one by one. Your comments are in bold italics, and my responses are in plain text. All the changes have been included in the newest version of our manuscript.

Specific Comments:

1. The method used to determine CF cannot be understood before reading steps (1) to (8) in lines 138 – 146. It would probably be useful to have an outline of Section 2.2 at the beginning of this section.

Response: Thanks very much for your comment. We have added the brief introduction of our new method in the very beginning of Sect.2.2.

2. Figure 2 shows intermediate results (indicating that satisfactory CF values cannot be obtained based on the SAE only), but there is no figure showing CF values eventually determined by the novel method vs CF values calculated using the Mie theory in dry conditions (a vast majority of nephelometers are operated in in dry conditions, in accordance with WMO-GAW recommendations).

Response: We put the verification result in the Sect.3.2. Under the dry condition, our new method of predicting CF performs well (Fig.11 in the new manuscript).

3. Simple processes are described in details (e.g. particle hygroscopic growth) while unobvious logical steps are not precisely explained (see examples in Technical Comments).

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

4. How much is the CF assessment learning depending on the assumptions about the aerosol mixing state, i.e. the fraction of purely scattering, purely absorbing, and mixed particles, including fully internally mixed aerosol?

Response: The parameters used for machine learning are derived from Mie calculation. It is demonstrated in Ma et al. (2015, Chinese journal) that both mixing states and the change of refractive index exert little impact on the Mie calculation results of scattering correction factor. As shown in the Table. S1 (translated from Ma et al., 2015), with the increase or decrease of refractive index, there are small relative differences of correction factors between the control group and experimental group, and it is the same case for change of mixing states. That is to say, although we set the refractive index of BC and non-absorbing as the constant value in our study, the results are still credible and accurate with small errors. Moreover, CF assessment learning barely depends on assumptions about the aerosol mixing state, too.

Table S1: Relative differences between CF_{550} calculated under different parameter assumptions and the reference value (translated from Ma et al., 2015).

Group	$m_{r,NBC}$	$m_{i,NBC}$	$m_{r,BC}$	$m_{i,BC}$	Mixing states	CF_{550}	Relative differences of CF_{550} (%)
Control	1.53	10^{-7}	1.75	0.55	Internal+External	1.1174	0
Experiment 1	1.50	10^{-7}	1.75	0.55	Internal+External	1.1208	0.30
Experiment 2	1.55	10^{-7}	1.75	0.55	Internal+External	1.1150	-0.21
Experiment 3	1.53	10^{-7}	1.50	0.55	Internal+External	1.1190	0.14
Experiment 4	1.53	10^{-7}	2.00	0.55	Internal+External	1.1157	-0.15
Experiment 5	1.53	10^{-7}	1.75	0.44	Internal+External	1.1169	-0.04
Experiment 6	1.53	10^{-7}	1.75	0.66	Internal+External	1.1177	0.03
Experiment 7	1.53	10^{-7}	1.75	0.55	Internal	1.1178	0.04
Experiment 8	1.53	10^{-7}	1.75	0.55	External	1.1171	-0.03

5. It is not stated if the CF determination method described would apply to TSI 3563 instruments, nor how measurements performed with a TSI 3563 (at least in campaign #5) were used in the machine learning process regarding the Aurora 3000 instrument.

Response: Our new method works for three-wavelength nephelometers, and hence it can be applicable to both Aurora 3000 and TSI 3563. This paper takes the Aurora 3000 as an example to introduce this method, and researchers can follow the method to figure out how TSI 3563 measurements perform. As for the TSI 3563, the truncation angle and the angular intensity distribution are different from those of Aurora 3000. That is to say, the machine learning process should be redone with the parameters calculated under the condition of TSI 3563 nephelometer's light source. It is not reasonable to use the random forest model trained for Aurora 3000 to predict the correction factor for TSI 3563. In our group, there is something wrong with TSI 3563 and Aurora 3000 is the only nephelometer that we use for now. Therefore, this paper takes Aurora 3000 rather than TSI 3563 as an example.

Technical Comments:

1. Line 23: "··· the aerosol direct radiative forcing varies ···" across what, as a function of what? Is this a range of uncertainty or variability?

Response: Sorry for the ambiguity. This is a range of uncertainty and we have added "the uncertainty of" in the new manuscript.

2. Line 25: Aerosol direct radiative forcing also depends on the aerosol HBF and vertical profile (or at least the integrated aerosol optical depth). The 4 variables (aerosol single scattering albedo, extinction coefficient, aerosol scattering coefficient, and absorbing coefficient) are equivalent. Knowing 2 of them is enough. Since this manuscript is about integrating nephelometers (which measure scattering), I would suggest to stick to "scattering and absorbing coefficients"

Response: Thanks for pointing this out. We have revised this part.

3. Line 50: suggestion: "Bond et al. (2009) found that SAE is also affected by the particle refractive Index."

Response: Thanks for your suggestion. Revised as suggested.

4. Line 50 – 56: please consider streamlining: the sentences referring to Bond et al (2009) are redundant.

Response: We have revised this part in the new manuscript.

5. Line 70: suggestion: "our number size distribution measurements cover a wide range of 10-1000 nm, ···"

Response: Thanks for your suggestion. Change made.

6. Line 71: Table 1: which nephelometers were used in campaigns 1-4?

Response: We did not own nephelometers at that time, and hence no nephelometers were used in campaigns 1-4.

7. Line 88: "···three types of particles:". The composition (i.e. chemical composition) controls the refractive index. What was the refractive index selected

for the absorbing material?

Response: We selected black carbon as the absorbing material and the refractive index is set to be 1.80-0.54i (Ma et al., 2012). Information has been added in the new manuscript.

8. Lines 97-100: this section is confusing. The variations in the SAE as a function of the particle diameter directly result from the Mie theory, and does not support the last sentence starting with "Therefore" (line 99): on which basis are particles in the size range 100-200 nm stated not to contribute to the overall SAE values? Is it meant that they do not contribute much to SAE variations?

Response: Yes, it is certain that SAE varies with particle diameter according to the Mie theory. We aim to demonstrate that, for particles in the size range 100-300 nm, the SAE variations are rather small. For instance, 100 nm particle can have the similar SAE value as 150 nm particle. If we use SAE values in this size range of 100-300 nm, particle diameters cannot be well represented. Sorry for this unclear statement. We have revised this part in the new manuscript.

9. Line 107-109: "aerosol particles show a noticeable feature of HBF decreasing with the increment of particle size. However, when the particle becomes larger than 300 nm, the HBF is almost unchanged." is again a direct consequence of the Mie scattering theory. And again the logical connection with the last sentence "HBF can represent the size information of particles smaller than 300 nm" is unclear. It was probably meant that HBF variability is mostly sensitive to the concentration of particles in the 100-300 nm size range.

Response: Yes, that is what we try to demonstrate. We have revised this part to avoid misunderstanding.

10. Line 123: rather HBF is sensitive to Rext

Response: Thanks for pointing this out. Change made.

11. Line 149: RF is not defined.

Response: Thanks for your comment. We have defined it in the new manuscript.

12. Figure 5: the diagram omits to mention that Mie calculations are performed

using both the actual nephelometer light source characteristics and an ideal light source, which is essential for determining CF.

Response: Both diagrams are updated based on your suggestion.

13. Line 167-168: RH instead of RH' in the denominator

Response: Sorry for the ambiguity. It should be a comma. We have deleted it in the revised manuscript.

14. Line 171: "f (RH) and fb(RH) values"

Response: Thanks for your comment. Revised as suggested.

15. Line 175: C(RH) is not precisely defined. It can be guessed afterwards that C = CF.

Response: Thanks for pointing this out. C means CF actually and C(RH) is the hygroscopic growth of CF. We have rephrased this sentence in the new manuscript.

16. Line 231: information is missing to support the statement "which improves the accuracy of the CF estimation in the dry state": "improves" compared to what?

Response: We stated at the beginning of the sentence "compared with the traditional correction method".

17. Line 249-250: this is quite obvious since increasing RH and increasing hygroscopicity have the same effect on the particle sizes (increased diameters).

Response: Yes, this sentence is obvious to some extent, but we need one conclusion at the end of the paragraph. We have rephrased the sentence according to your comment.

18. Line 271: suggestion: "essential" rather than "significant"

Response: Thanks for your suggestion. Revised as suggested.

19. Line 273: The sentence "The scattering correction factor (CF) relating to the aerosol size and chemical composition is thus put forward" is unclear. Is it meant: "The correction factors (CF) to be applied depend on the aerosol particle number size distribution and chemical composition."?

Response: Sorry for the unclear statement. What we want to stress is that the scattering correction factor (CF) is thus put forward. That CF relates to the aerosol size and chemical composition is the additional information used as postpositive attributes. This sentence is rephrased as “The scattering correction factor (CF) is thus put forward and it depends on the aerosol size and chemical composition.”

20. Line 274-285: The description of the Mie calculation method is obscure. However, a clear and concise description is needed, since it is the basis for the machine learning process.

Response: We have deleted some sentences to make this part more concise.

21. Line 286: Suggestion: “SAE and HBF provide information on the aerosol particle size distribution for different size ranges (···)”. The size ranges should be specified in brackets.

Response: Thanks for your suggestion! We have added “(300-1000 nm for SAE and 100-300 nm for HBF)” in the revised manuscript.

22. Line 293: The first sentence should state that this paragraph regards “humidified nephelometer measurements”.

Response: We have added “the humidified nephelometer system is utilized” in the revised manuscript.

Reference:

Ma, N., Zhao, C. S., Müller, T., Cheng, Y. F., Liu, P. F., Deng, Z. Z., Xu, W. Y., Ran, L., Nekat, B., van Pinxteren, D., Gnauk, T., Müller, K., Herrmann, H., Yan, P., Zhou, X. J., and Wiedensohler, A.: A new method to determine the mixing state of light absorbing carbonaceous using the measured aerosol optical properties and number size distributions, Atmos. Chem. Phys., 12, 2381-2397, doi:10.5194/acp-12-2381-2012, 2012.

Ma, N., Zhou, X. J., Yan P. and Zhao, C. S.: A method of correcting the measurements of TSI 3563 nephelometer, Journal of Applied Meteorological Science (Chinese journal), 26(01), 15, doi: 10.11898/1001-7313.20150102, 2015.