Atmospheric Measurement Techniques Discussions

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Comment on amt-2020-412

Anonymous Referee #3

Referee comment on "New correction method for the scattering coefficient measurements of a three-wavelength nephelometer" by Jie Qiu et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-412-RC2, 2021

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.

Specific Comments:

- 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.
- 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).
- Simple processes are described in details (e.g. particle hygroscopic growth) while

unobvious logical steps are not precisely explained (see examples in Technical Comments).
- 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?
- 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.

Technical Comments:

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

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"

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

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

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

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

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?

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.

Line 123: rather HBF is sensitive to Rext

Line 149: RF is not defined.

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.

Line 167-168: RH instead of RH’ in the denominator

Line 171: “f (RH) and fb(RH) values”

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

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?

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

Line 271: suggestion: “essential” rather than “significant”

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.”?

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.

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.

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