Comments on "A new parameterization scheme of the real part of the ambient aerosols refractive index" by Zhao et al.

This study deals with a very interesting and important issue, i.e., parameterization of aerosol refractive index, which is essential for the estimation of aerosol direct radiative forcing. I read the manuscript and the authors' response with great interest. However, after careful evaluation, I agree with the other reviewer that this study is not suitable for publication in ACP as "it needs further analysis, reorganization, discussion and clarification to (prove) improve the confidence of the results (reviewer 1)". I will expand a bit on these issues as detailed below.

Major Comments:

The parameterization scheme of the current study ($Re = (RRI^2-1)/(RRI^2+2) = 0.18\rho_{eff}$) is in principle a justification / an update of the scheme proposed by Liu and Daum (2008) ($Re = (RRI^2-1)/(RRI^2+2) = 0.23\rho_{eff}^{0.39}$) based on a new dataset measured at Taizhou in China for 7 days in June 2018. The major concern from the other reviewers is whether the new parameterization is universal and applicable in global and climate models as suggested by the authors. I am not convinced by the author's arguments because of the following reasons.

1. Is the new scheme universal and better than the one from Liu and Daum (2008)?

   If a scheme is universal, it should not only explain one dataset, but also be applicable and compatible for other datasets. To come up with their parameterization scheme, Liu and Daum (2008) studied the relationship of refractive index to mass density (index-density relationship) for over 4000 pure materials and for aerosol particles. Note that, in Liu and Daum (2008), the summarized pure materials include organics, and investigated aerosol data cover aerosol samples from Amazon (Guyon et al., 2003), which is expected to constitute significant fraction of organics. Thus, it is not appropriate for the authors to make a statement "the influence of organic aerosols components on aerosol RRI is not considered in their work (L270-271)".

   In Fig. C1, I compared the results of the current study with Liu and Daum (2008) by overlaying the data from the Taizhou site (small light blue dots) and from the PKU site (small pink dots) onto the original Fig. 3 of Liu and Daum (2008). Interestingly, the new datasets are not much different from the ones already summarized by Liu and Daum (2008), as they fit well into the data clouds within the same mass density range. For the whole data population, it appears that Liu and Daum's scheme (black solid line) is still the best approach to describe the overall index-density relationship. Because the new parametrization from the current study (blue solid line) is not able to represent the general trend in the existing dataset (over 4000 pure materials marked by small grey dots), especially it failed to explain the aerosol data from early field campaign/laboratory studies (marked by big black triangles).

   Depending on to which degree one would like the schemes to represent the variability, for the Taizhou site the predicted average RRI (~1.44) by Liu and Daum's scheme is in a reasonable agreement with the observed 28-day average RRI of 1.425, 1.435 and

1.47 for 200 nm, 300 nm and 450 nm particles, respectively, which is probably already good enough for global and climate model applications.

On the other hand, when speaking of explaining the detailed temporal and spatial variability, the prediction of the new parameterization at the PKU site is quite scattered with y = 1.0x and R^2 = 0.03 (see my Major Comments 2). For example, a prediction of RRI ~1.5 with the new parameterization scheme at the PKU site may correspond to a variability of real/observed RRI from 1.42 to 1.58 (Fig. C2-A).

Thus, it is unlikely that the new parameterization scheme from the current study is universal and applicable to global and climate models. In my opinion, the intrinsic scattering of the index-density relationship (Fig. C1) implies that a perfect parameterization may not be even possible. If a compromise has to be made, Liu and Daum's scheme still seems to be optimal choice in terms of universality.

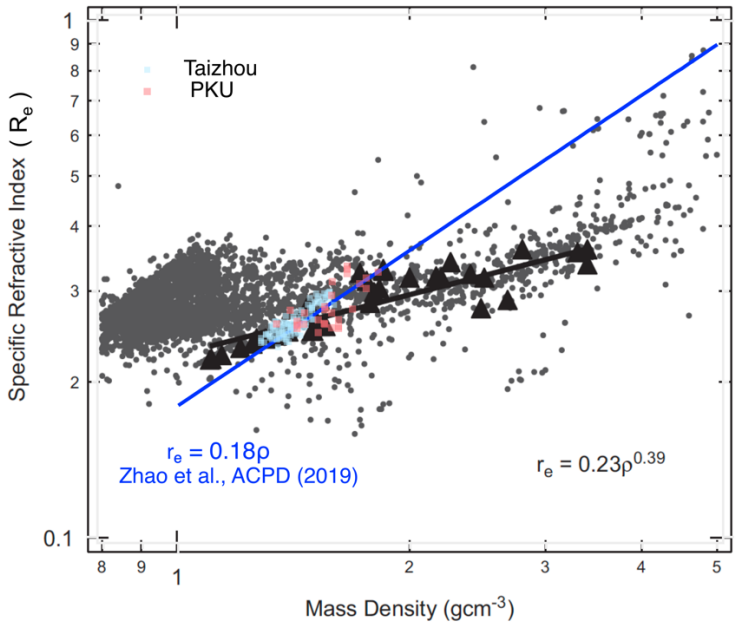

Fig. C1. Dependence of the effective refractive index (Re = (RRI^2-1)/(RRI^2+2)) on the effective mass density (ρ_{eff}) for pure materials and for aerosol particles. The new data sets from the current study (small light blue dots for Taizhou and small pink dots for PKU) overlay the original Fig. 3 of Liu and Daum (2008). The data summarized by Liu Daum (2008) includes over 4000 pure materials (small grey dots, including organics, inorganics and minerals, www.knovel.com), as well as ambient aerosol and lab generated surrogate with chemical compositions representing ambient aerosols (big black triangles) (Hänel, 1968; Tang and Munkelwitz, 1994; Hand and Kreidenweis, 2002; Guyon et al. 2003).

2. Consistence between the PKU and Taizhou sites?

When comparing the consistency of different dataset, I find that I cannot reproduce the results of Fig. 6 in the revised manuscript (also shown here as Fig. C2-B). While the authors provided a R^2 of 0.47 for the PKU site (Fig. C2-B), using the same fit function (y = ax by forcing intercept = 0), I received a coefficient of determination (R^2) of only 0.03 for the same dataset (Fig. C2-A), not sufficient to support the authors' argument about consistence between the PKU and Taizhou sites. Apparently, the authors have selected the "good" slope (1.0) of y = ax and the "better" R^2 (0.47) of y = ax + b to justify the advantage of their method in Fig. 6 and relevant text. This is misleading. Such a way of selectively presenting results is a serious issue and has to be corrected.

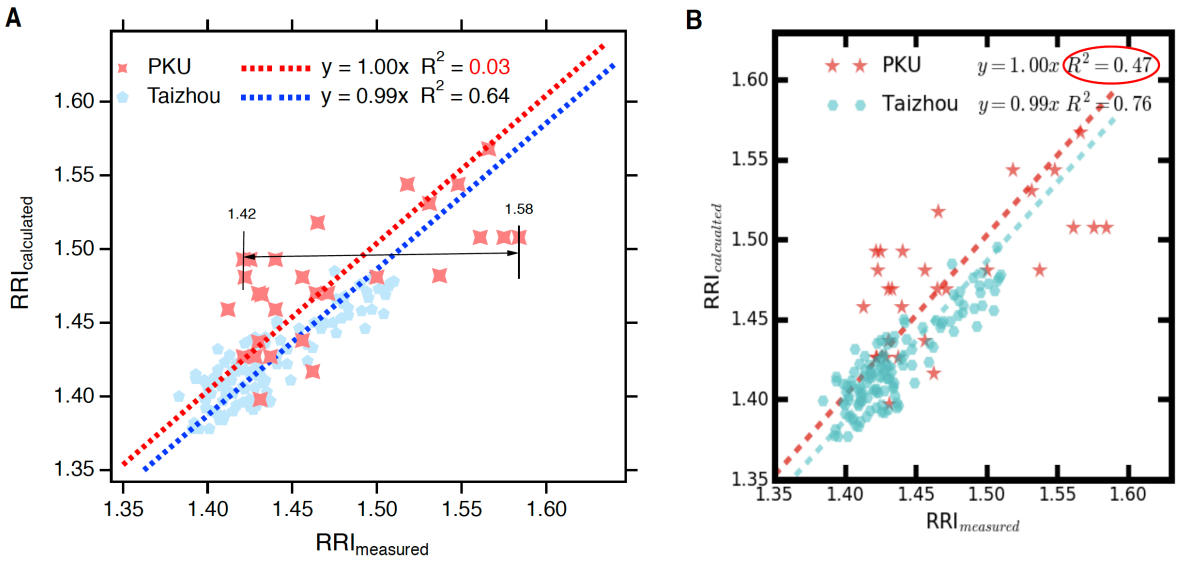

Fig. C2. Comparison of measured and predicted RRI by the parameterization scheme of Zhao et al. (ACPD). **A**, my re-calculation with data from Fig. 6 of Zhao et al. (ACPD). **B**, the original Fig. 6 of Zhao et al. (ACPD).

Other Comments:

1. Abstract: The retrieved RRI is for pure scattering aerosols (or may be extended for the coating materials when calculating the effective refractive index of mixed black particles?), while the effective density is measured for all aerosols (both scattering and absorbing aerosols). Direct comparison between the two measured quantifies may induce uncertainties, and should be justified or at least clarified.

2. Abstract and section 3.2: I suggest to remove "rather than the main chemical components" from "We find that the ambient aerosol RRI is highly related with the aerosol effective density (ρ_{eff}) rather than the main chemical components", or change 'related' to 'correlated'. This is because both refractive index and effective density are determined by main chemical components of aerosol particles. Even for the proposed application in global or climate model (calculation of RRI from ρ_{eff}), one would still need the simulated chemical compositions to calculate ρ_{eff} (see my Other Comments 3).

   Along this line, section 3.2 should be substantially revised by including more detailed and thorough evidences and discussions. See my concerns below.

   The major argument/results presented in section 3.2 to support the conclusion "the ambient aerosol RRI is highly related with the aerosol effective density (ρ_{eff}) rather than the main chemical components" are Fig. 4, Fig. 5 and Fig. S6 (in the revised manuscript). However, RRI and chemical compositions in these comparisons were not taken from the same aerosol group. The RRI were taken from aerosol of a certain size (i.e., 300 nm) while the chemical compositions were taken from $PM_{2.5}$ either for direct comparison (Fig. 4 and Fig. S6) or for calculating the RRI (Fig. 5) (The water-soluble ions were from $PM_{2.5}$; whether ECOC measurements were from $PM_{2.5}$ or PM_{10} are not clear in the text of section 2.1). In this case, even the same parameter may differ from each other. For example, one can clearly see multiple modes in the comparison of main aerosol components and RRI in Fig. S6.

Such comparison can be misleading because there is a danger that the readers might get an impression that the commonly used mixing rules in calculating refractive index wouldn't work for ambient aerosols, e.g., volume linear mixing rule, Maxwell-Garnet and Brüggemann mixing rule, partial molar refraction mixing rule, Lorentz-Lorenz mixing rule etc. (those requires information of chemical compositions in a mixed system). A direct consequence has already been shown in Fig. 5, where the authors delivered a message that Stelson's approach (Stelson, 1990) of calculating refractive index with partial molar refraction mixing rule did not work for the Taizhou case. This is unfair because the mismatch of different aerosol population in this comparison may to some (large) extend lead to the very scattered data points of the Stelson's approach in Fig. 5.

3. The authors' response about "how to use the parameterization in numerical models, i.e., what is the input and required parameters, may be required" is not adequate. Modelers understand "the effective density is the only parameter as input", but the real question is how to determine the effective density in the model, which hasn't been answered. I guess that one would still need to calculate ρ_{eff} from densities of individual simulated chemical composition. This procedure however, may be hampered by the lack of density information of organic carbons and mixing state of black carbon, etc.

4. Concerning Reviewer 2's comment "Zhao et al. (2018b) seems to be still under discussion. The readers cannot trust the method only from the explanation in this manuscript.", the authors may want to refer to the work of Zhang et al. (JGR, 2018), where the method of combining DMA and SP2 to retrieve the real part of the refractive index of pure scattering aerosol particles has been proposed and published.

5. L50: "main aerosol" is duplicated.

6. L198-199: could it be that statistics of RRI at 200 and 300 nm is better that at 450 nm? Because the scattering signal of SP2 may become saturated for a large fraction of particles at 450 nm, and reduce the sample size. How were the double charged particles treated?

7. L147-151: "SSA is defined as the ratio of σ_{sca} to σ_{ext}, which reflects concentration of the absorbing aerosol (Tao et al., 2014) to some extent. The *g* expresses the distribution of the scattering light intensity in different directions (Zhao et al., 2018a). The σ_{ext}, SSA and *g* are the most important three factors that influence the aerosol radiative properties in radiative calculation (Kuang et al., 2015)." L169-170: "… the difference between fn↓ and fn↑ (fn↓ - fn↑) is the downward radiative irradiance flux for aerosol-free conditions (Kuang et al., 2016)."

   It does not seem to be appropriate to cite these references here, because such statements are rather classical textbook knowledge.

8. L192: change "The RRI and ρ_{eff} vary…" to "The RRI varies".

9. L201-202: change "at about 15:00 in the morning" to "at about 15:00 in the afternoon" and change "at around 9:00 in the afternoon" to "at around 9:00 in the morning".

10. L214-215: "…Thus, the effective tend to increase with the increment of aerosol diameter." sounds like a broken sentence.

11. L254: "one day" instead of "one days".

12. Fig. 5: The caption is confusing and needs to be revised. "Comparison between the measured RRI and calculated RRI using the main aerosol chemical component from Stelson (1990) (in red star)…" Do the authors mean using the same aerosol chemical species those are needed for applying the Stelson's method, but the concentrations of the chemical components are still the measured ones from this study? If yes, please revise the caption. This is related to reviewer's comment about "Why do the authors compare a result with other at different time series and measurement site?"

Reference

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N. Tang, H. R. Munkelwitz, Water activities, densities and refractive indices of aqueous sulfates and sodium nitrate droplets of atmospheric importance. J. Geophys. Res. 99, 18801-18808 (1994).

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A. W. Stelson, Urban aerosol refractive index prediction by partial molar refraction approach, ES&T, 24, 1990.