Response to reviewer#2

Thanks for the reviewer's helpful suggestions! The comments are addressed point-by-point and responses are listed below.

**Comment:** General comments: The real part of the refractive index is surely still uncertain and its impact on the aerosol radiative forcing (ARF) is large. The scope of this manuscript is important. The logic of this manuscript is generally clear, but the following three points should be clarified.

**Reply:** Thanks for the comments.

**Comment:** Firstly, the title is "A new parameterization scheme of the real part of the ambient aerosols refractive index", so the proposed parameterization must be evaluated in the manuscript, but the evaluation is not enough. The parameterization is based on the measurements at one Chinese site during May-June of the specific year. Generally, the parameterization must be universal, so the proposed one should be tested under various conditions using other measurements at different places and seasons or using a numerical model. Otherwise, I suppose other people do not tend to use the proposed parameterization

**Reply:** Thanks for the comment. The objective of this article is to bring up a novel idea of parameterization scheme of real part of the refractive index (RRI) for ambient aerosol. Traditionally, RRI is parameterized by the measurement of ambient aerosol main inorganic components (Han et al., 2009). The influence of organic compositions is ignored. In this work, we found that the ambient aerosol RRI was highly related with the aerosol effective density ($\rho_{eff}$) rather than the chemical components. Thus, a new parameterization scheme of the RRI using the effective density was proposed.

To validate the universality of this parameterization scheme, we conducted another measurement in the campus of Peking University (PKU) (N39°59′, E116°18′), in China, where the aerosol effective density and real part of the refractive index are measured concurrently at 16th, December in 2018. The RRI were also calculated using the parameterization scheme, $\frac{RRI^2-1}{RRI^2+2} = 0.18\rho_{eff}$. Comparison of the measured and calculated RRI is shown in fig. R1. Results show that the calculated and measured RRI show good consistence.

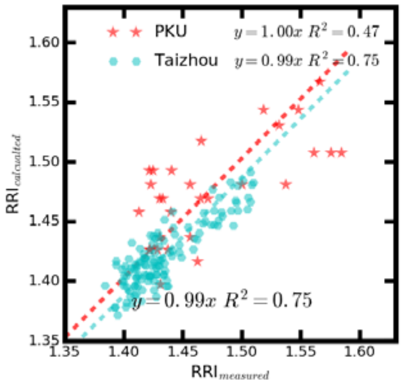


**Fig. R1.** Comparison between the measured and calculated RRI at PKU and Taizhou.

**Comment**: Also, an introduction how to use the parameterization in numerical models, i.e., what is the input and required parameters, may be required.

**Reply:** Our parameterization scheme is simple and easily used in numerical models because the effective density is the only parameter as input. We have demonstrated that the traditional method of calculating the RRI using aerosol main chemical components can have significant bias because the effects of organic aerosol is not considered. We added some discussions in the manuscript correspondingly.

**Comment:** Second, the main conclusion can be led from Figure 4. However, Figure 4 only indicates that Equation (1) is applicable for the effective particle (I understand this is also one of the findings in this study). I expect the clear evidence of the relationship between measured-RRI and calculated-RRI, as shown in Figures S8 and S9.

**Reply:** Thanks for the comment. We have replotted the figure 4.

**Comment:** Finally, in the result and discussion of section 3.4, the authors estimated the ARF, but the objectives of this section may be side tracked. Here, the authors should discuss the impact of the parameterization on the ARF, but the conclusion is "the real-time measured RRI be used rather than a constant RRI when estimating the ambient aerosol optical and radiative properties". This conclusion confuses me. When the proposed parameterization is applied to numerical models, is the real-time measured RRI still required? If so, this parameterization is not attractive to modelers. In addition, the experimental conditions of the ARF calculation is unclear (see the below comment).

**Reply:** Thanks for the comments. Traditionally, a constant RRI is used when estimating the DARF. As shown in section 3.4, large uncertainties may arise when estimating the DARF using a constant RRI. The real time measured RRI should be used rather than a constant RRI in order to estimate the ambient aerosol optical and radiative properties with high accuracy. However, the real-time measurement of ambient aerosol RRI is not available for most of the conditions. Our proposed parameterization scheme can act as a substitute for real-time RRI.

We added some descriptions of method for ARF calculation in section S3 in the supplementary material.

**Comment:** In overall, the manuscript would be acceptable for publication if these comments can be satisfactorily addressed.

**Reply:** Thanks for the comment.

**Comment:** Specific comments: L23 (and L233): Only correlation coefficient is not enough to evaluate the relation. Please add the other statistical metrics.

**Reply:** Thanks for the comment. We have added the slope in the manuscript.

**Comment:** In abstract, the correlation coefficient is 0.75, but the value is 0.76 in Figure 4. Which is right?

**Reply:** Thanks for the comment. This is a typo and we corrected it.

**Comment:** L36: Which wavelengths are used?

**Reply:** Thanks for the comment. The wavelength range between 0.2 and 5 um is used for calculating the radiative forcing (Marshall et al., 1995; Moise et al., 2015). We have added the description in the text.

**Comment:** L103: Zhao et al. (2018b) seems to be still under discussion. The readers cannot trust the method only from the explanation in this manuscript.

**Reply:** Thanks for the comment. We added some discussions about this method in the manuscript. Before the measurement, this system is calibrated with ammonia sulfate (RRI=1.52). After calibration, ammonium chloride is used to validate the method of deriving the RRI from SP2 for different aerosol diameters. The RRI value of ammonium chloride is 1.642 (Lide, 2006). The retrieved RRI of ammonium chloride is in the range between 1.624 and 1.656. Therefore, this measurement system can measure the ambient aerosol RRI with high accuracy.

**Comment:** L144-145: RI of BC is set at 1.8+0.54i. Do the authors consider a dependence of RI on wavelength?

**Reply:** Thanks for the comment. The RI value of 1.8+0.54i is frequently used in estimating the radiative effects of BC particles (Bond et al., 2013; Zhao et al., 2018). The dependence of RI on wavelength for BC particle is not well studied yet (Bond and Bergstrom, 2006). Therefore, a constant RI of BC at different wavelength is used in estimating the DARF.

**Comment:** L159: Please clarify "parameterization aerosol vertical distributions". This information is very important to estimate the ARF.

**Reply:** Thanks for the comment. We have added some descriptions in the section 3 of the supplementary material to introduce the method of calculating the aerosol vertical profiles.

**Comment:** L198-200: The RRI was measured at three different wavelengths (200nm, 300nm and 450nm). Here the measured RR is expressed as "1.34-1.56". Can the measured RRI at different wavelengths be combined? Do the authors consider the difference of RRI among the different wavelengths? In addition, is the focusing wavelength consistent to those proposed by the previous studies?

**Reply:** Thanks for the comment. The light scattering is measured by SP2 at the wavelength of 1064 and the measured RRI corresponds to the wavelength of 1064 nm. This system is no capable of measuring the RRI among different wavelengths. However, the measured RRI of ambient inorganic aerosols has little variation among different wavelengths. The RRI for $(NH_4)_2SO_4$ varies by 0.02 and less than 0.01 for wavelengths between 400 nm and 700 nm (Cotterell et al., 2017).

We conducted optical closure studies to demonstrate that the measured RRI at 1064 nm is applicable at other wavelength. First, the scattering coefficients ($\sigma_{sca}$) at wavelengths of 450, 525 and 635 nm were calculated using the measured refractive index at 1064 nm and Mie model (Bohren and Huffman, 2007) using the measured aerosol particle number size distribution and the BC mixing states. Then the calculated $\sigma_{sca}$ are compared with the measured $\sigma_{sca}$ by an nephelometer (Aurora 3000, Ecotech, Australia) (Müller et al., 2011). The Aurora 3000 is capable of measuring the $\sigma_{sca}$ at 450, 525 and 635 nm. The scattering truncation and non-Lambertian error was corrected using the same method as that of Ma et al. (2011). The comparison of measured and calculated $\sigma_{sca}$ are shown in fig. R2. The measured and calculated $\sigma_{sca}$ show good consistence, demonstrating the measured RRI using our measurement system is applicable in other wavelength.

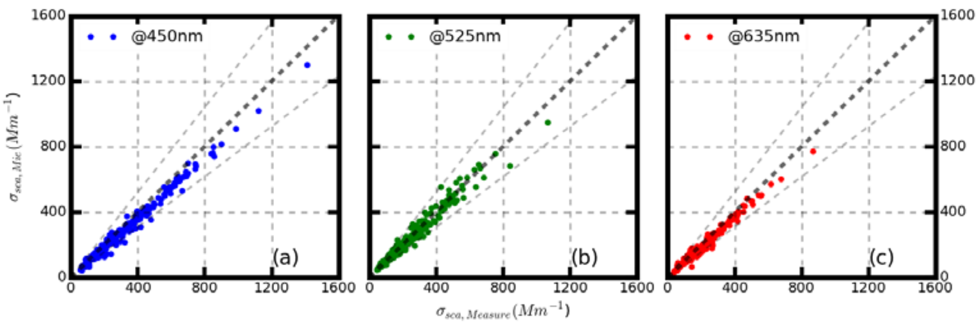

**Figure R2.** Comparison between the measured scattering coefficient and calculated scattering coefficient at (a) 450 nm, (b) 525 nm and (c) 635 nm.

**Comment:** L204-205: Can the authors explain the mechanism of the relationship between effective density and particle size?

**Reply:** Thanks for the comment. The difference of the effective density among different particle size should be resulted from the different chemical compositions. Based on the previous measurements of the size-resolved chemical compositions using a MOUDI, the mass fraction of OM decreases with the increment of aerosol diameter (Hu et al., 2012). At the same time, the effective density of OM is lower than the other inorganic compositions. Thus, the effective density increases with the increment of aerosol diameter.

**Comment:** Figure 5: Is the instant value or mean? Which wavelength do the authors calculate? Please clarify them.

**Reply:** Thanks for the comment. The calculated DARF from SBDARF is an instant value. The instant DARF is calculated over the wavelength range between 0.25 μm and 4 μm. We have added the descriptions in the text.

**Comment:** Figure S8 and S9: They are very interesting. I strongly recommend they are moved to the main text. Can the authors show the same figures estimated from the current study?

**Reply:** Thanks for the comment. Fig. 4 is replotted. Fig. S8 and fig. S9 were merged into figure 5.

**Comment:** Technical comments" L34: prat –> part

**Reply:** Thanks for the comment. We have revised it.

**Comment:** L46: It is better to add “n: refractive index” to the explanation of Equation (1).

**Reply:** Thanks for the comment. We have changed the equation as

$$RRI_{eff} = \sum_{\mathrm{i}} (\mathrm{f_i} \cdot \mathrm{RRI_i})$$

Where $\mathrm{f_i}$ and $\mathrm{RRI_i}$ are the volume fraction and real part of refractive index of known composition *i*.

**Comment:** L52: ne –> neff is suitable.

**Reply:** Thanks for the comment. We changed the "n" into $RRI_{eff}$ in the manuscript.

**Comment:** Figure S1 (a), S4, S5: Better to be moved to the main text.

**Reply:** Thanks for the comment. Fig. S1 is moved into Fig. 2 in the text and part of fig. S4 is moved to the main text.


Bohren, C.F., Huffman, D.R., (2007) Absorption and Scattering by a Sphere, Absorption and Scattering of Light by Small Particles. Wiley-VCH Verlag GmbH, pp. 82-129.

Bond, T.C., Bergstrom, R.W. (2006) Light Absorption by Carbonaceous Particles: An Investigative Review. Aerosol Science And Technology 40, 27-67.

Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J., Flanner, M.G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., Zender, C.S. (2013) Bounding the role of black carbon in the climate system: A scientific assessment. Journal Of Geophysical Research-Atmospheres 118, 5380-5552.

Cotterell, M.I., Willoughby, R.E., Bzdek, B.R., Orr-Ewing, A.J., Reid, J.P. (2017) A complete parameterisation of the relative humidity and wavelength dependence of the refractive index of hygroscopic inorganic aerosol particles. Atmospheric Chemistry and Physics 17, 9837-9851.

Han, Y., Lü, D., Rao, R., Wang, Y. (2009) Determination of the complex refractive indices of aerosol from aerodynamic particle size spectrometer and integrating nephelometer measurements. Applied Optics 48, 4108-4117.

Hu, M., Peng, J., Sun, K., Yue, D., Guo, S., Wiedensohler, A., Wu, Z. (2012) Estimation of size-resolved ambient particle density based on the measurement of aerosol number, mass, and chemical size distributions in the winter in Beijing. Environ Sci Technol 46, 9941-9947.

Lide, D.R. (2006) Handbook of Chemistry and Physics, 86th Edition Edited(National Institute of Standards and Technology). Journal of the American Chemical Society 128, 5585-5585.

Ma, N., Zhao, C.S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y.F., Deng, Z.Z., Liu, P.F., Xu, W.Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildenberger, K., Henning, S., Yu, J., Chen, L.L., Zhou, X.J., Stratmann, F., Wiedensohler, A. (2011) Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study. Atmos. Chem. Phys. 11, 5959-5973.

Marshall, S.F., Covert, D.S., Charlson, R.J. (1995) Relationship between asymmetry parameter and hemispheric backscatter ratio: implications for climate forcing by aerosols. Applied Optics 34, 6306-6311.

Moise, T., Flores, J.M., Rudich, Y. (2015) Optical properties of secondary organic aerosols and their changes by chemical processes. Chemical Reviews 115, 4400-4439.

Müller, T., Laborde, M., Kassell, G., Wiedensohler, A. (2011) Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer. Atmos. Meas. Tech. 4, 1291-1303.

Zhao, G., Zhao, C., Kuang, Y., Bian, Y., Tao, J., Shen, C., Yu, Y. (2018) Calculating the aerosol asymmetry factor based on measurements from the humidified nephelometer system. Atmospheric Chemistry and Physics 18, 9049-9060.