Anonymous Referee #1

<General>

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This paper describes measurements of the real part of the refractive index and density of aerosol particles having different chemical compositions using simultaneous scattering measurements with time of flight aerosol mass spectrometer measurements. Though the basic concept of the method is not new, the authors developed a measurement instrument and applied to the observation in Guangzhou, China. They compared their results with previously reported measurements and discussed characteristics of aerosols in the observed region. The paper is well written, and the content is suitable for publication in ACP. One general comment, which may be beyond the scope of this paper, is that the method is not sensitive to the imaginary part of refractive index as described in the paper. However, the imaginary part of refractive index or the single scattering albedo of aerosol particles is an essential parameter determining radiative characteristics. Are there any possible extensions of the single particle aerosol mass spectrometer techniques for detecting some signals related to absorption, for example, by detecting infrared radiation at the ionization with the pulsed laser?

We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript. We agree with the comment that the method is not sensitive to the imaginary part of refractive index, which is an essential parameter determining radiative characteristics. The absorption properties of particles are typically obtained through the measurements of the attenuation of light that passes through the aerosols (e.g., Arnott et al., 2005; Petzold and Schonlinner, 2004). This procedure requires some amount of samples collected in at least a few *minutes to accurately obtain the absorptive efficiency of particles. The single particle and highly time-resolved measurement limits the ability of SPAMS to collect enough information on absorptive properties. However, it is noted that this limitation only poses effects on absorptive particles, i.e., soot-containing particles.*

Regardless of this limitation, Moffet and Prather (2009) have developed a method to investigate the absorptive properties of soot-containing particles. They applied Mie core-shell model, assuming soot as cores with a constant refractive index (1.8 + 0.71i) and non-absorbing components as shells. The model adjusts the core diameter to obtain best fit between the scattering curve to model result. In this way, the scattering and absorbing of soot-containing particles could be retrieved. It is necessary to note that since the refractive index for soot varies, and thus uncertainty would not be avoided in the model.

Anonymous Referee #2

Received and published: 8 January 2016

This manuscript describes the characterization of aerosols in Guangzhou, China with a single particle mass spectrometer. The analysis included using the collected mass spec data to determine the density and the real part of the refractive index. This manuscript is well-written, the topic is relevant for ACP, and provides important new results. The results are put into context with previous studies. I would recommend that the manuscript is accepted. I do have a couple minor points on the manuscript that I would recommend are addressed before the final submission.

In section 2.2, the discussion of the use of the "upper limit" is a little unclear as written. I would recommend either on page 3452 line 27 adding the exact "data" that are being referred to in the sentence ": : :we only used data that lie at the 90th percentiles." I believe this is referring to LSS. Alternatively, if the discussion of how the calculation was performed (page 34653 starting at line 6) was moved towards the beginning of the paragraph, might help to make the description of the upper limit more clear. Also, I would recommend in this section to include a line that says something about how in the figures that follow in the results section these points are referred to as "upper limit," as that term was not specifically used often in the methods section, and thus was a little confusing in the results section.

We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript. We agree with the comment, and we have revised this paragraph as recommended to make the description of the upper limit more clear.

The paragraph has been revised to "The methodology is briefly summarized herein. Firstly, theoretical response is firstly compared with the response of scattered light measured by SPAMS from PSL particles with sizes ranging from 150 to 2000 nm. Then a calibration curve is constructed to transform the LSS (at the 90th percentiles, i.e., upper limit) measured by SPAMS to the PSCS, enabling a quantitative comparison between the measured and theoretical PSCS. Before performing scattering calculation, the detected particles were grouped into 17 particle types on the basis of chemical compositions. Finally, a series of n and peff were used as input in Mie theoretical calculation to find the best fit (i.e., a global minimum of the sum of squares due to error, SSE) between the measured and theoretical PSCS with least square method, thus enabling the estimation of n and peff. A detailed description of the methodology for the SPAMS and the results of the test on the known aerosol samples (i.e., NaNO3 and (NH4)2SO4) is available in our previous publication (Zhang et al., 2015a). During the sizing detection, the particles diverge from each other and thus a wide range of light scattering intensities from nearly zero to some upper limit are obtained for similar particles. This is due to the uneven distribution of laser beam energy, and also to the relative position of the laser beam and the pathway of particles. In order to avoid the effect of high intensity outliers in our study, we used only LSS that lie at the 90th percentiles of the collected data in 20 nm size bins, regarding the upper 10% as outliers. Satisfied results were obtained for calibration on PSL and test on NaNO3 and (NH4)2SO4). Other percentile data (such as values *lie at 95th and 99th percentiles) was also tested, however, the PSL calibration cannot be improved, which is possibly attributed to much more outliers lying at the upper 10% percentile of the collected data. Therefore, we use 90th percentiles of LSS as their upper limit values in the following discussion."*

Please refer to Lines 113-134 in the revised manuscript.

Section 3.1.2, starting line 14 of page 34656, it is stated that EC that has been exposed to water can change its shape towards more spherical. In this study the particles were dried. Would it be expected that EC particles, if they started as spheres, would keep their shape after they are dried?

It is hardly to show the variations of particle shape after they are dried in this study. With a Transmission Electron Microscopy, Pagels et al. (2009) illustrated that fresh soot is with a highly irregular structure, coated soot with a considerable compaction. They also showed that considerable compaction of the particles had occurred when they were heated or dried. Consistently, our study showed that there were still spherical particles when particles are dried as indicated in Figure 3.

Page 34650 line 7, should read ": : :various compounds: : :"

It has been corrected as suggested.

Page 34650 line 15 should read ": : :measurements into aerosol mass: : :"

It has been corrected as suggested.

Page 34650 line 20, should read "..which have served as important parameters: : :"

It has been corrected as suggested.

Page 34651 line 25, I would recommend "related" instead of "corresponded" in "the velocity is corresponded to: : :"

It has been corrected as suggested.

Page 34652 line 23, include cite of previous publication that explains methodology.

The citation (Zhang et al., 2015) has been included in the revised manuscript, please refer to Line 124.

Zhang, G., Bi, X., Han, B., Qiu, N., Dai, S., Wang, X., Sheng, G., and Fu, J.: Measurement of aerosol effective density by single particle mass spectrometry, Science China Earth Sciences, 1-8, doi:10.1007/s11430-015-5146-y, 2015a.

Page 34686, line 6, should read ": : :there metal rich types are mainly: : :"

It has been corrected as suggested.

Figure 5, it is hard to distinguish between black and green line. Perhaps would lighter color for the green line work better? Or even just a different symbol (i.e. square, cross) would help.

The line color has been revised to light green as suggested, please refer to Fig.5 in the revised manuscript.

Reference

Arnott W.P., Hamasha K., Moosmuller H., Sheridan P.J., Ogren J.A., 2005. Towards aerosol light-absorption measurements with a 7-wavelength Aethalometer: Evaluation with a photoacoustic instrument and 3-wavelength nephelometer. Aerosol Science and Technology 39, 17-29.

Moffet R.C., Prather K.A., 2009. In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates. Proceedings of the National Academy of Sciences of the United States of America 106, 11872-11877.

Pagels J., Khalizov A.F., McMurry P.H., Zhang R.Y., 2009. Processing of Soot by Controlled Sulphuric Acid and Water CondensationMass and Mobility Relationship. Aerosol Science and Technology 43, 629-640.

Petzold A., Schonlinner M., 2004. Multi-angle absorption photometry - a new method for the measurement of aerosol light absorption and atmospheric black carbon. Journal of Aerosol Science 35, 421-441.