Responses to the comments of reviewer 6

The authors really appreciate the valuable comments and constructive suggestions from the reviewer. The suggestions and comments of reviewer are listed in black font, and responses are highlighted in blue. The changes made in the revised manuscript are marked in red font.

Comments from reviewer 6:

General Comments: The manuscript by Liu et al. presents results from the light scattering matrices for the samples collected from Chinese Loess Plateau. Auxiliary analyses including particle size distribution, refractive index, chemical component, and microscopic appearance etc. were also done. Based on their results, the authors conclude that the size distribution play a major role in leading to different matrices. In general, the method developed by the authors is novel and fits the slope of the journal. However, some modifications are necessary before it can be considered for publication. One major comment is that the authors did not discuss the atmospheric implications of this novel method. The authors mentioned that the average scattering matrix changed due to the updated sample "pristine loess" compared to previous studies (Fig. 6), this is very interesting, but how meaningful this is to the atmospheric aerosols study? How accurate will it be if we use this new average scattering matrix in future studies? Also, it is necessary for the authors to ask a native English speaker to review the article.

Response:

Thank you very much for reviewing our manuscript and all these constructive comments. We have responded to the comments point by point and modified related descriptions in the revised manuscript. We also have invited native English speakers to review our manuscript to correct language mistakes. And we hope that you will reconsider our manuscript.

Optical modeling of dust particles with non-spherical shapes has been an essential subject. Dubovik et al. (2006) recommended a specific shape distribution of spheroids with different aspect ratios to be used in the retrieval of dust aerosol properties from remote sensing observations, because this shape distribution of spheroids had the best performance in reproducing measured scattering matrices for different kinds of mineral dust published by Volten et al. (2001). Since then, this shape distribution had been widely used in the properties retrieval of airborne dust particles. However, Li et al. (2019) found that shape distributions of spheroids have obvious effects of scattering matrices and further affect radiance distribution and polarization properties of sky light. Therefore, the application of above recommended shape distribution for all kinds of dust with different properties is somewhat unreasonable. Our study provided measurements of scattering matrices of "pristine loess" and "milled loess" with large difference in their size distributions, from the perspective of particle transportation. These measured results are useful for the refinement of shape distributions of spheroids for optical modelling of loess dust, and are finally useful to improve retrieval accuracy of dust aerosol properties over both loess source and downwind regions. Furthermore, the updated average scattering matrix for loess is more representative than before, because it contains measured results of coarse "pristine loess" sample that affects dust source regions. We think this average scattering matrix is also useful for the verification and development of more advanced morphological models for loess dust than spheroids. The improvement of retrieval accuracy of dust aerosol properties by using shape distributions of spheroids or more advanced models retrieved based on our average scattering matrix still requires specific studies to quantify.

We have added above atmospheric implications in the Introduction and Conclusions in the revised manuscript:

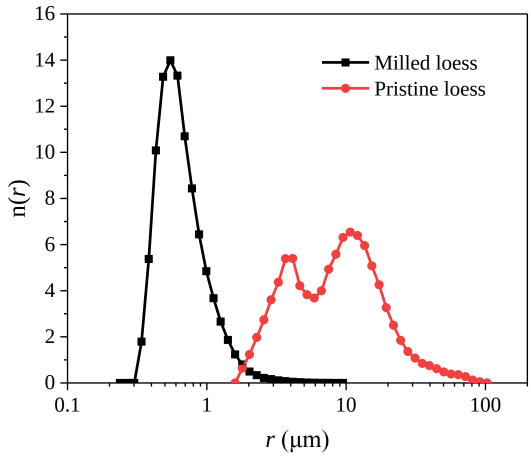
"It is well known that dust particles have distinct non-spherical shapes, thus retrievals of dust aerosol properties, like optical thickness, based on Lorenz-Mie computations will lead to significant errors (Herman et al., 2005; Mishchenko et al., 2003). Optical modeling of dust particles with non-spherical shapes has been an essential subject. Dubovik et al. (2006) employed a mixture of spheroids with different axial ratios as well as spheres to reproduce laboratory measured angular light scattering patterns of dust aerosols presented by Volten et al. (2001), and the best fitted shape distribution of spheroids was obtained and proposed. Subsequent studies on the retrievals of dust aerosol properties from space-based (Dubovik et al., 2011), airborne (Espinosa et al., 2019) and ground-based (Titos et al., 2019) remote sensing observations were all based on this shape distribution. However, the application of a same shape distribution of spheroids for different kinds of dust is somewhat too arbitrary (Li et al., 2019) and may not be suitable for simulating optical properties of loess dust with different size distributions. Furthermore, more precise optical models which are more complex than spheroids and similar to real dust morphology are still needed. Laboratory measurements of angular scattering patterns as well as basic physical features, like size distribution, refractive index and micro structure, of loess dust with different sizes are essential and beneficial to the development of more precise models for loess dust. These models will further useful for more accurate retrievals of dust aerosol properties over both source and downwind regions from remote sensing observations, and more accurate assessments of radiative forcing at different regions."

"In this study, scattering matrices for Chinese loess samples with large difference in their size distributions are investigated. Based on all the measurements, suitable shape distributions of spheroids can be obtained respectively, which are useful for the retrievals of airborne loess dust properties at both source and downwind areas in China or even East Asia. On the other hand, the updated average scattering matrix for loess are meaningful for the validation of exiting models and the development of more advanced morphological models suitable for loess dust, which are also useful to finally improve the retrieval accuracies of dust aerosol properties."

Specific comments:
Pg4, line 100: Figure 1: What is the meaning of r in y-axis? Please explain in the figure caption.

Response:

Thanks a lot for your comments. In Figure 1, x-axis is radius of particle "*r*", and y-axis is normalized number fraction "n(*r*)" for particle with radius "*r*". We have redrawn Figure 1 and modified descriptions in figure caption in the revised manuscript:

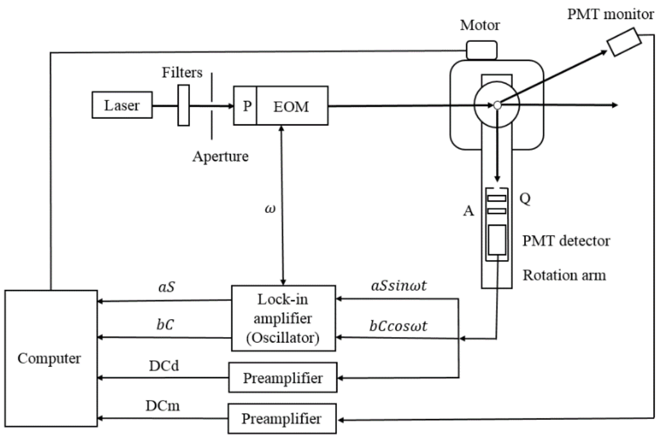


"**Figure 1.** Normalized number size distributions n(r) of "pristine loess" and "milled loess". Radius r is plotted in logarithmic scale, and error bars are small and covered by symbols."

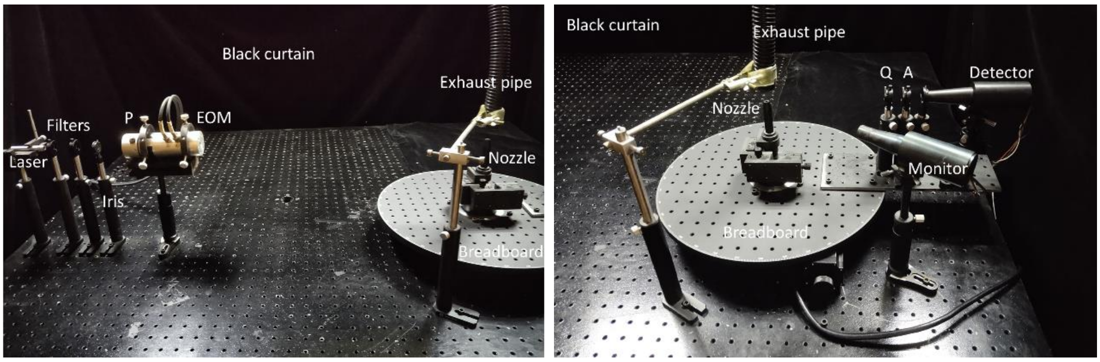
Pg6, Fig. 3 is not so clear. Please draw a schematic of the experimental setup.

Response:

Thank you very much for the comments. The schematic of our experimental apparatus can be seen below, and it had been published in our previous paper (Liu et al., 2018), so we did not add it to the revised manuscript.

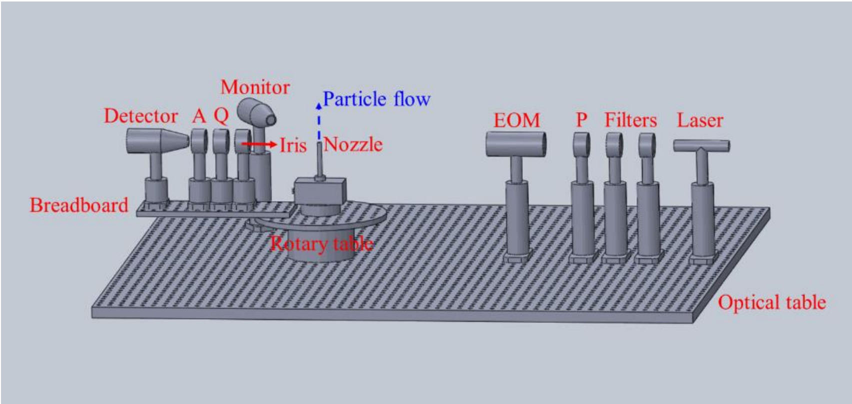


During the revision of manuscript, we improved the experimental apparatus by extending its maximum coverage of backscattering angle from 160° to 175°, and we re-measured scattering matrices for these two loess samples. In this way, during the construction of synthetic matrices, extrapolated values of scattering matrix elements $F_{11}(\theta)/F_{11}(10°)$ and $F_{22}(\theta)/F_{11}(\theta)$ at 180° scattering angle were more reliable. For the extension of angle coverage of apparatus, mechanical structures of scattered light detection part were adjusted and optimized. The dark cassette used to encapsulate the "detector", Q and A in previous apparatus is removed, which also facilitate the adjustment of orientation angles of Q and A. We have updated Figure 3 in the revised manuscript using a simple layout diagram of the improved apparatus, and the photograph of apparatus in the following figure had been shown in our another work (Liu et al., 2020).



We also have modified descriptions about Figure 3 in the revised manuscript:

"Figure 3 shows a layout diagram of the improved scattering matrix measurement apparatus."



"**Figure 3.** Layout diagram of the experimental apparatus after backscattering angle expended."

Pg6, line 155: How do you inject the dust aerosols into the setup? Please clarify.

Response:

Thanks a lot for pointing this out. We have added more detailed descriptions in the revised manuscript:

"Subsequently, the modulated incident light is scattered by particles in the scattering zone, which are dispersed using an aerosol generator and are sprayed upwards to scattering zone through a nozzle."

"A dust generator (RBG 1000; Palas) was applied to disperse loess particles (Liu et al., 2018). Re-aerosolized dust aerosols were transported to scattering matrix measurement apparatus using conductive tube and sprayed upwards to scattering zone through nozzle."

Pg8, line 222: The last sentence "while other. . . " is not clear.

Response:

Thank you very much for the comments. We have modified related descriptions in the revised manuscript:

“Therefore, it is reasonable to suspect that distinctions in angular distributions of measured scattering matrix elements for two loess samples may be mainly caused by different size distributions (effective radii differ by more than 20 times), while differences in other factors such as refractive index and micro structure have relatively small contributions in leading to different scattering matrices.”

Pg11, line 313-314: The authors should indicate what are these small “milled loess” compared to.

Response:

Thanks a lot for pointing this out. We have modified related descriptions in the revised manuscript:

“More specifically, for small “milled loess”, relative phase function $F_{11}(\theta)/ F_{11}(10°)$ as well as ratios $-F_{12}(\theta)/F_{11}(\theta)$ and $F_{22}(\theta)/F_{11}(\theta)$ are smaller than that for coarse “pristine loess”, while ratios $F_{33}(\theta)/F_{11}(\theta)$, $F_{34}(\theta)/F_{11}(\theta)$ and $F_{44}(\theta)/F_{11}(\theta)$ are larger than that for coarse “pristine loess”.”

Pg10-11, In the conclusion section, the authors should explicitly explain what is “novel” in the new average scattering matrix compared to their previous study, and the significance of this study.

Response:

Thank you very much for the valuable comments. We have added related descriptions in the revised manuscript:

“Synthetic scattering matrices for both “pristine loess” and “milled loess” were defined over 0°-180° scattering angle, and the previously presented average scattering matrix for loess was updated with new coarse “pristine loess” sample included. The phase function $F_{11}(\theta)$ in updated average matrix has larger forward scattering peaks and smaller values at side and backward scattering angles than that in previous average matrix. Compared to previous average matrix, updated average matrix has larger $-F_{12}(\theta)/F_{11}(\theta)$ at side scattering angles, has smaller $F_{33}(\theta)/F_{11}(\theta)$ and $F_{44}(\theta)/F_{11}(\theta)$ at backscattering angles. $F_{22}(\theta)/F_{11}(\theta)$ experiences the largest change before and after update, whose values are enlarged at almost all scattering angles.”

“In this study, scattering matrices for Chinese loess samples with large difference in their size distributions are investigated. Based on all the measurements, suitable shape distributions of spheroids can be obtained respectively, which are useful for the retrievals of airborne loess dust properties at both source and downwind areas in China or even East Asia. On the other hand, the updated average scattering matrix for loess are meaningful for the validation of exiting models and the development of more advanced morphological models suitable for loess dust, which are also useful to finally improve the retrieval accuracies of dust aerosol properties.”

References

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