Responses to the comments of reviewer 5

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 5:

In Liu et al. the paper focuses on describing the scattering function of a sample that was collected from the Chinese Loess Plateau and subsequently milled to change the physical properties of the particles. The major conclusion gleaned by the authors in the article is that the size of the particles affects the scattering properties. The paper does describe well the need for the research being performed on complex systems, but systematic experiments need to be performed to start to tease out some of that information instead of broad statements about size since that is what they were trying to control. The authors mention that the size distribution is the major factor, but refractive index and micro structure are not ignorable (line 237-240) and then seem to discount that the shape and refractive index have little effect (line 318). They additionally mention that the Refractive index is different (Table 1), but do not seem to try and account for the difference using any kind of modeling to show that it is primarily size. Or identify as to why these are different for the same material.

Response:

Thank you very much for reviewing our manuscript and all these constructive comments.

In our study, we investigated two loess samples with large difference in their particle size distributions from the perspective of long range transportation of dust, the effective size parameters of these two samples are 580 and 30, respectively. Optical simulation results of Gaussian spheres showed that, for particles with above two size parameter, the effects of size parameter and irregularity on scattering matrix elements are roughly opposite (Liu et al., 2015). And the effect of size was qualitatively confirmed by experimental results of these two loess samples, so we concluded that the difference in size distributions plays a major role in leading to discrepancies in measured scattering matrices. However, for particles with effective size parameter smaller than "pristine loess" (580) but larger than "milled loess" (30), optical simulations of Gaussian spheres showed that both size and irregularity have roughly similar effects on matrix elements F_{33}/F_{11}, $F_{34}/F_{11}$and F_{44}/F_{11}, it becomes impossible to identify the main factor of influence. In such cases, only qualitative analyses is not enough anymore, supports and cooperation from optical modeling experts are essential to further explore the reasons of discrepancies in matrix elements. Therefore, we did not investigate samples with more sizes, and only measured scattering matrices for two loess samples with large difference in their size distributions. We very hope that there are optical modeling experts interest in our preliminary experimental results and cooperate with us to investigate more samples, since only combinations of experimental measurements and optical simulations are more meaningful.

As mentioned above, with the assistant of qualitative analyses of simulations of Gaussian spheres, we think difference in size distributions is the main reason for these

discrepancies in measured scattering matrices for loess. However, even though it is hard to exactly quantify the change of particle morphology, its effect on scattering matrix is also obvious (Liu et al., 2015). Furthermore, difference of real part of refractive indices of loess samples may also have effects on scattering matrix (Muinonen et al., 2007). So based on qualitative analyses of optical simulation results, we can only draw conclusions that size distribution plays a major role in leading to different scattering matrices while differences in factors such as refractive index and micro structure have relatively small and recessive contributions..

As for the descriptions of refractive index, there is a clerical error in original manuscript, which should be "larger particles have relatively smaller real part of refractive index". For specific material, the refractive index is inherent and unique. SALD-2300 retrieves refractive index of particles by reproducing measured light intensity distributions based on Mie theory. The retrieved refractive index can be regard as a kind of optical equivalent refractive index, it is close to inherent refractive index of measured material but not necessarily the same.

Kinoshita (2001) retrieved refractive indices for alumina dust (whose inherent refractive index is known to be 1.76) with 1 μm and 5 μm diameter using the same method as SALD-2300, the retrieved real parts were 1.80 and 1.75 respectively, the difference was small but cannot be ignored, and this phenomenon was explained as the effect of nonspherical nature by Kinoshita. In our study, we also found larger particles have smaller real part of refractive index, so we think the difference in real part is real and can be explained using the same reason.

This paper does not show significant new data or a new approach to understanding the optical properties of aerosol particles that had not been published previously by the group. The technique has been described by the authors at least twice previously in prior publications and one of the 2 sample sets is already published elsewhere (Liu et al. 2019 and 2018). The paper itself needs to be edited further and reorganized as there are multiple sections that are very similar but spread out through the paper. This paper appears to be more of an addendum to the Liu et al 2019 article than a stand-alone article. Based on these above points, I would be hesitant to recommend this paper for publication as is since there is little information that is novel and there are some unsubstantiated claims throughout.

Response:

Thanks a lot for your comments, and we also appreciate your attention on our previous works.

As we mentioned in the manuscript, the effect of particle size on scattering matrices of mineral dust is still not clear enough, and there is no published research on the effect of size on scattering matrices for loess. Loess dust originating from Chinese Loess Plateau usually forms sand storms in spring season, affecting its source and downwind regions in East Asia. In this study, we experimentally investigated scattering matrices for loess dust with large discrepancy in the size distributions, which is meaningful from the perspective of dust long range transportation. Measured scattering matrices for both coarse "pristine loess" and fine "milled loess" samples are meaningful for the refinement of shape distributions of widely used spheroids as well as the validation and development of more advanced models (Dubovik et al., 2006; Li et al., 2019; Liu et al., 2015). These models will help to improve the retrieve accuracy of aerosol properties

from remote sensing observations over both dust source regions and downwind remote regions.

Until now, the scattering matrix measurement method, synthetic matrix and average matrix construction method have been rigorous enough. Therefore, we just followed the previous methods, and we think that measurement results of samples with atmospheric implication are more important than the improvement of these methods to some extent.

During the revision of the manuscript, we re-measured scattering matrices for both "pristine loess" and "milled loess" samples using an improved experimental apparatus, the maximum backscatter angle coverage of which was extended from 160 ° to 175 °. Newly measured results were in good agreement with measurement results using the original apparatus in the range of 5-160 °. The extension of scattering angle made the polynomial extrapolation of matrix elements $F_{11}(\theta)/F_{11}(10°)$ and $F_{22}(\theta)/F_{11}(\theta)$ at backscattering angles more rigorous when constructing synthetic matrix, and calculated backscattering depolarization ratios were also more reliable.

We have modified related descriptions about apparatus as well as measured, synthetic and average scattering matrices. We also have re-organized the manuscript and added more descriptions about atmospheric implication. And we hope that you will reconsider our manuscript.

Specific Comments:

Line 26: Please specify what this % is, from written it appears to be total aerosol loading worldwide.

Response:

Thanks a lot for pointing this out. We have specified the meaning of "%" in the revised manuscript:

"During aerosol characterization experiments ACE-Asia, mass balance calculations indicated that 45-82 % of atmospheric aerosol mass at observation sites in China were attributed to Asian dust (Zhang et al., 2003)."

Line 36: Please specify what 'r' refers to specifically.

Response:

Thanks a lot for your comments. The 'r' stands for aerodynamic diameter, which is used to characterize transportation and deposition ability of particle. In our study, we measured optical diameter of dust, because, to our best knowledge, there is no instrument available to measure aerodynamic diameter of particles larger than 20 μm, while measurement range of SALD-2300 covers optical diameter from 0.017-2500 μm.

As for the relationship between aerodynamic diameter and optical diameter, Chen et al. (2011) showed that the ratio of aerodynamic diameter to optical diameter is about 0.94-1.21 for Asian dust. Furthermore, according to Li et al. (2018), the ratio of aerodynamic diameter to optical diameter is about 1.15. In our study, we did not make a strict distinction between these two kinds of diameter, because this did not affect the assessment of whether the two loess samples are capable of long range transportation.

Line 42: Remove “Without a doubt”

Response:

Thank you for pointing this out. We have remove these words in the revised manuscript:

“Optical properties of dust particles vary with changes of their size distributions.”

Line 55: ‘Furthermore ... scattering matrices’. This sentence is not completely coherent and needs to be rewritten.

Response:

Thanks for your suggestion. We have rewritten this sentence in the revised manuscript:

“Most published literatures of experimental measurements of scattering matrices focused more on similarities and discrepancies between different kinds of mineral dust, or between the same kinds of dust sampled from different sources. Furthermore, some researches paid more attention to the effect of particle size distribution on scattering matrices.”

Line: 99-100: What was the injection type for the laser particle sizer? Were they injected in solution or dry?

Response:

Thank you for the comments. During the measurement of size distribution, dry loess particles were injected into the measurement unit of laser particle sizer. We have added related descriptions in the revised manuscript:

“The size distributions of “pristine loess” and “milled loess” were determined by a laser particle sizer (SALD-2300; Shimadzu) using dry measurement method, dry loess particles were injected into the measurement unit of laser particle sizer, and three independent repeated measurements were conducted for each sample.”

Line 101: Size comparison can be difficult between the two samples due the fact that the original dust sample has a bimodal distribution. This distribution itself will lead to very different scattering properties, whereas the milled sample is a more uniform size. What is the cause of the bimodal shape? Could this be due to a heterogeneity of mineral types being different sizes and having large differences in scattering properties that are then not comparable to the milled sample?

Response:

Thanks for your comments. We agree that it is difficult to compare bimodal distribution with unimodal distribution. That is why we employed effective radius and effective standard deviation, with the help of these two parameters, we can compare

particles with different size distributions. Volten et al. (2001) showed that the directly sampled and unprocessed red clay, loess, volcanic ash and Sahara sand have different size distributions. This may be because these samples contain different mineral components, these components have distinct size distributions and finally lead to different size distributions of these samples. In our study, the bimodal distribution of "pristine loess" may also be explained using the same reason, and after ball milling, size distributions of different mineral components tend to be the same unimodal distribution. The difference in size distributions can be reflected by measured scattering matrices to some extent.

We have modified related descriptions in the revised manuscript:

"As can be seen from Figure 1, the size of "pristine loess" shows a distinct bimodal distribution, after ball milling, particle size of "milled loess" becomes a unimodal distribution."

Line 103: It is stated that the majority of the particles are larger than 5 microns, but there is a peak at 3 and 10 microns. Please reword this section because you use a cutoff of 5 microns earlier for local vs. long range transport.

Response:

Thank you very much for the comments. As we mentioned in manuscript, particles with radii larger than 5 μm cannot be transported over long distances. However, this does not mean that all airborne particles over source regions have radii larger than 5 μm, there are still a part of fine particles. In our study, the number fraction of particles with radii larger than 5 μm are more than 70% in "pristine loess" sample, so we think this sample can be used to represent airborne loess dust over source regions. We have modified some descriptions in the revised manuscript:

"From the viewpoint of atmospheric particle transportation, the majority (number fraction more than 70%) of "pristine loess" particles have radii larger than 5 μm with peaks at about 3.9 and 10.7 μm, thus this sample can be used to represent coarse dust that only affect source regions, like Xi'an City (Yan et al., 2015)."

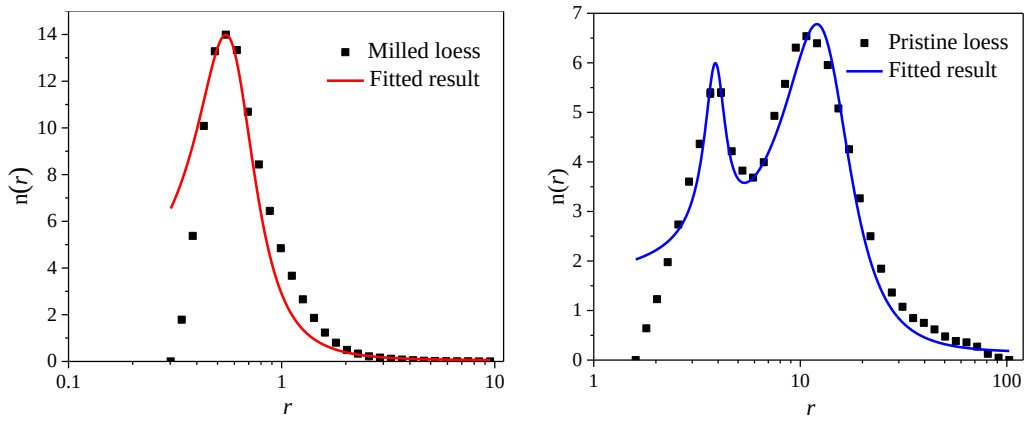
Line 105: Please define the peaks more clearly for both samples, with a peak maximum and additional parameters to describe the spread.

Response:

Thanks a lot for the comments. We tried to use Origin Software to fit the measured size distributions of loess samples, and we found only Lorentz function have relatively good fit results. The Lorentz function can be written as:

$$y = y_0 + \frac{2A}{\pi}\frac{w}{4(x - x_c)^2 + w^2}$$

where x_c is peak center, A is peak area, w is full width at half maximum, and y_0 is offset of y-axis. Fitted results for "milled loess" (left panel) showed that the peak center x_c is 0.55 μm and full width at half maximum w is 0.46.And fitted results for "pristine loess" (right panel) showed that the peak centers x_c are 3.87 and 12.05 μm, and full widths at half maximum w are 1.11 and 12.21.



However, we think the fitted results for both loess samples are not satisfactory, especially for small radius values. Therefore, we used the peak radii of measured results only in the revised manuscript rather than the fitted results:

"From the viewpoint of atmospheric particle transportation, the majority (number fraction more than 70%) of "pristine loess" particles have radii larger than 5 μm with peaks at about 3.9 and 10.7 μm, thus this sample can be used to represent coarse dust that only affect source regions, like Xi'an City (Yan et al., 2015). On the other hand, almost all particles of "milled loess" sample have radii smaller than 2 μm with a peak at about 0.55 μm, and can be used as a representative of fine dust that can be transported over long distance and affect regions far away from dust sources."

Line 110/Table 1: Why is there a difference in the refractive index if they are still the same material? Please provide the error associated with the measurements and propagate through the rest of the calculations.

Response:

Thank you for the comments. As we mentioned above, refractive index of particles retrieved by SALD-2300 is optically equivalent value, and it is not necessarily the same as inherent refractive index of measured material.

The reason for the small difference 0.05 in retrieved real parts of refractive index for our loess samples is because of the nonspherical nature of particles, Kinoshita (2001) also found similar phenomenon for alumina dust with different sizes. The smallest available calculation steps of real and imaginary part of refractive index in the retrieval are 0.05 and 0.01, respectively. All three repeat measurements obtained the same refractive indices for both "pristine loess" and "milled loess". We have added necessary descriptions of the retrieval of refractive index in the revised manuscript:

"During size distribution measurements of loess samples, the retrieval ranges of real part $Re(m)$ and imaginary part $Im(m)$ of refractive index were preset as 1.45-1.75 and 0-0.05, respectively (Volten et al., 2001). The smallest calculation steps of $Re(m)$ and $Im(m)$ are 0.05 and 0.01, respectively. As shown in Table 1, the optimal refractive indices are $1.65+0i$ for "pristine loess" and $1.70+0i$ for "milled loess", larger particles have relatively smaller real part of refractive index, which is similar to the results of Kinoshita (2001) and is caused by the nonspherical nature of loess dust. Retrieved

refractive index of particles based on measured light intensity distribution is a kind of optically equivalent refractive index, which is close to the inherent refractive index of the measured particles."

Line 120: how are the samples for SEM prepared? Are they impacted on the surface or collected some other way?

Response:

Thank you very much for the comments. During sampling process, we sprayed particles vertically onto copper grids through airflow, particles impact and attach on the surfaces of copper grids. We have added necessary descriptions in the revised manuscript:

"Some particles of each loess sample were sprayed into vessels or sprayed onto copper grids for subsequent size distribution measurements or SEM analyses."

Line 129: What is the detection limit of this instrument? You quote down to 0.0001 wt% in Table 2. This is mainly of interest since I do not know the limits of XRF.

Response:

Thanks a lot for your comments. For the instrument XRF-1800, the detection limit is 0.0001 wt%. We have added descriptions about the detection limit in the revised manuscript:

"For the purpose of detecting whether the chemical compositions of loess samples were changed, the oxide compositions of samples before and after milling process, that is the "pristine loess" and "milled loess", were determined using a X-ray fluorescence spectrometer (XRF-1800, Shimadzu), the detection limit of which is 0.0001 wt %."

Table 2: add an additional column with the difference between the pristine and milled samples. Also include that the characterization was performed by XRF in the caption.

Response:

Thank you for the comments. We added repeat measurements of chemical components of our loess samples using XRF-1800, and obtained the experimental errors from three measured results for each sample. Comparisons of chemical components for "pristine loess" and "milled loess" showed that the differences between these two samples are small and negligible when experimental errors were taken into consideration. Therefore, we did not add column of component differences between these two loess samples. But we have added columns of experimental errors to Table 2 in the revised manuscript. In addition, we also have added descriptions of XRF in table caption and modified descriptions of sample differences in the revised manuscript:

"As can be seen in Table 2, the largest change of content occurs for SiO_2, but this change is less than 2.5 % and even smaller than the errors between repeat measurements for "pristine loess" sample, and the change of ZrO_2 is only about 0.03 %. It can be concluded that the composition differences between these two samples are very small, and milling process has little effect on chemical compositions for loess samples."

"**Table 2.** Chemical components of "pristine loess" and "milled loess" measured by XRF-1800."

Components	Pristine loess (wt %)	Pristine loess error (wt %)	Milled loess (wt %)	Milled loess error (wt %)
SiO_2	63.8278	3.0237	66.2128	2.0900
Al_2O_3	12.3091	0.3772	11.6487	0.2018
CaO	9.2943	0.9455	7.8286	0.6450
Fe_2O_3	5.5260	0.8817	5.6390	0.7411
K_2O	3.3971	0.3004	3.3574	0.2358
MgO	2.7536	0.4522	2.4843	0.2665
Na_2O	1.2802	0.0243	1.3470	0.0214
TiO_2	0.8017	0.0595	0.7939	0.0579
P_2O_5	0.3340	0.0452	0.2549	0.0018
SO_3	0.2370	0.1056	0.1687	0.0721
MnO	0.1240	0.0294	0.1196	0.0120
ZrO_2	0.0583	0.0104	0.0846	0.0122
SrO	0.0348	0.0064	0.0299	0.0059
Rb_2O	0.0177	0.0041	0.0174	0.0040
Co_2O_3	NT*	-	0.0159	0.0049
Y_2O_3	NT*	-	0.0061	0.0025

Line 124: The aggregated particles are all on the large size of the size distribution, would this affect the scattering properties greatly or are they artefacts from particle collection for SEM analysis?

Response:

Thanks a lot for your valuable comments. We think these aggregated large particles are more likely artefacts of the sampling process using copper grids, since we spay particles onto grids directly. Therefore, we resampled particles for SEM analyses and obtained more representative images for "milled loess".

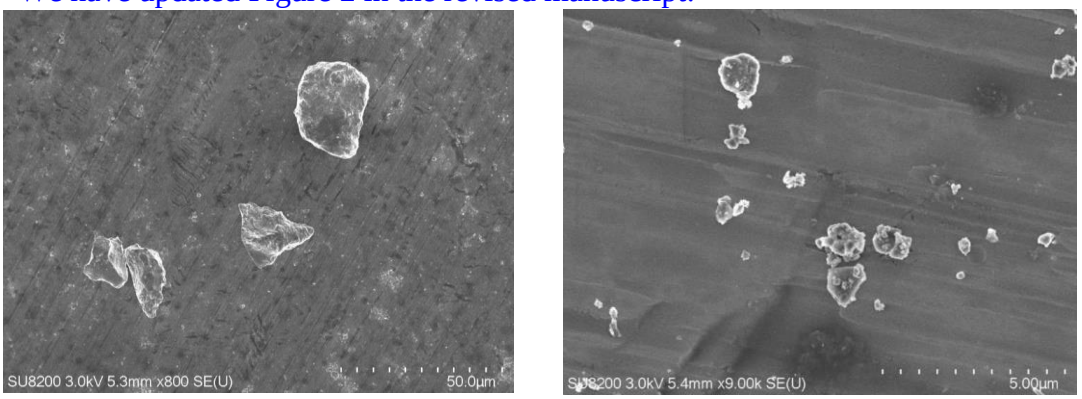
Figure 2: The SEM image for the pristine loess only shows particles in the 10s of microns, it is not a representative image of what the particles actually would look like since the peaks are at ~3 and 10 microns. Additionally, the image for the 'milled loess'

is the same as previously published in the prior manuscript. Please provide representative and comparative SEM images.

Response:

Thank you very much for pointing this out and thanks for your attention on our previous work. The peaks of radii of "pristine loess" particles are about 3.9 and 10.7 μm, corresponding particle diameters are about 7.8 and 21.4 μm. We resampled loess particles and performed SEM analyses again. Figure 2 in the revised manuscript has been updated using a more representative image of "pristine loess", particle sizes in which are much closer to peaks measured by laser particle sizer, as well as a new image of "milled loess". In addition, we think optical equivalent diameter measured by laser particle sizer are similar to but not exactly equal to geometric size in SEM images.

We have updated Figure 2 in the revised manuscript:



"**Figure 2.** SEM images for "pristine loess" (left panel) and "milled loess" (right panel)."

Line 126: What size ZrO2 ball were used and were they milled wet or dry?

Response:

Thanks a lot for your comments. The "milled loess" sample was prepared by dry ball milling method, and ZrO_2 balls with 6 mm diameter were used. We have added related descriptions in the revised manuscript:

"During the dry milling process, non-metal grinding balls with 6 mm diameter were used, the main component of which is ZrO_2."

Line 153-160: I like that the detectors are defined differently, but it would be better to have a different description that 'monitor' and 'detector' as they are both the same pmt detectors just with different functions.

Response:

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

"A photomultiplier named as the "detector", a 532 nm quarter-wave plate *Q* as well as a polarizer *A* are fixed on a rotation arm, rotation center of which is coincides with the center of aerosol nozzle. Before scattered light is detected by the "detector", it successively passes through *Q* and *A*. The dark cassette used to encapsulate the "detector", *Q* and *A* in previous apparatus is removed, which facilitate the adjustment of orientation angles of *Q* and *A*. The "detector" is controlled by an electric rotary table and is able to scan scattering angles from 5 ° to 175 °. Another photomultiplier named as the "monitor" is fixed at 30 ° scattering angle to record variations of dust aerosols."

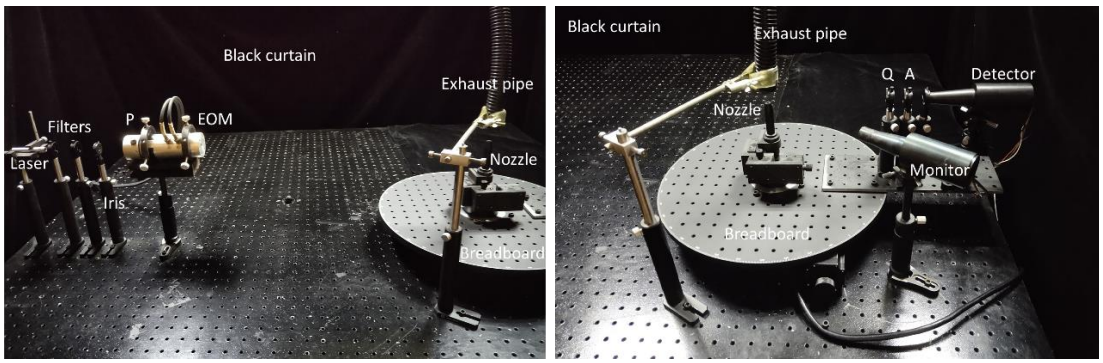
"Fluctuations of dust aerosols can be eliminated by normalizing measurements of the "detector" using *DC*(30°) measured by the "monitor"."

Figure 3: This does not seem necessary as the technique has been described twice previously.

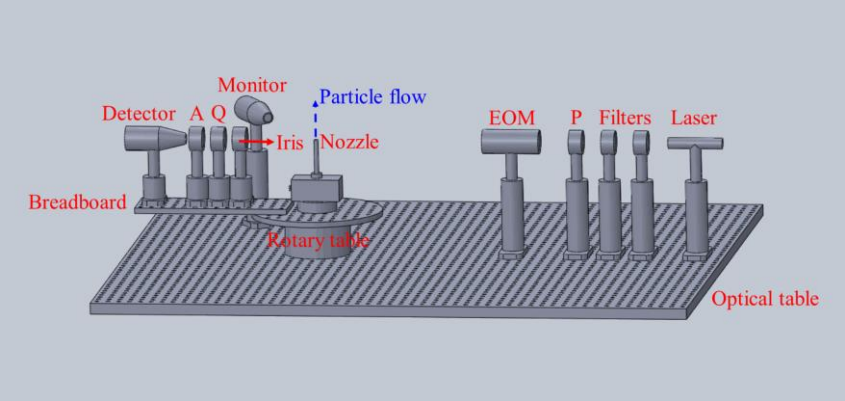
Response:

Thanks a lot for the comments.

During the revision of the manuscript, we improved experimental apparatus by extending coverage of the maximum backscattering angle from 160 ° to 175 °, and we re-measured scattering matrices for both "pristine loess" and "milled loess" samples. In this way, during the construction of synthetic matrices, the values of matrix elements $F_{11}(160°)/F_{11}(\theta)$ and $F_{22}(\theta)/F_{11}(\theta)$ at exact backscattering angle 180 ° obtained by extrapolations were more reliable. For the extension of angle coverage of apparatus, mechanical structure of detection part of scattered light was 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*. Therefore, we still showed a simple layout diagram of the improved apparatus in Figure 3 in the revised manuscript, and the photograph of improved apparatus in the following figure and detailed validation results using water droplets had been shown in our another work (Liu et al., 2020).



We have updated Figure 3 in the revised manuscript:



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

Lines: 215-223: this paragraph is in an odd place as it references past tables and figures.

Response:

Thank you very much for the comments. In this paragraph, we summarized the differences in fundamental characteristics of these two loess samples, and attempted to infer the main reason for the discrepancies in measured matrices. This provides general guidance for the analyses of literatures focusing on particle optical modeling in the next paragraph in the manuscript. Therefore, we think it is necessary to keep this paragraph, but we have modified and simplified it in the revised manuscript:

"In this study, several fundamental properties of loess dust samples were characterized for auxiliary analyses. As shown in Table 1, effective radii for "pristine loess" and "milled loess" are 49.40 μm and 2.35 μm, respectively. The real part of refractive index for "pristine loess" is 1.65 and that for "milled loess" is 1.70. Table 2 shows that the changes of chemical components are negligible. 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."

218: "loess dust become more irregular after milling process" How is this defined? If you are saying that they become more irregular, then you will need to actually do analysis of the particles themselves to show the change in the shape parameters. Based on the images seen, this statement cannot be made.

Response:

Thanks a lot for your valuable comments. We agree that this statement is not rigorous. To our best knowledge, it is still very hard to use several morphological parameters to adequately describe the real morphologies of irregular dust particles. Therefore, we removed the related descriptions in the revised manuscript.

In addition, we think the best way to evaluate the change of particle irregularity at present may be employ shape models with different parameters, which may not be fully representative of true morphology of dust, to reproduce measured scattering matrices for these two loess samples, the best fitted shape distributions can be retrieved, then the change of particle irregularity can be roughly evaluated. For such evaluation, we definitely need to cooperate with optical modeling experts.

Line 241-253: This paragraph could be combined with the conclusion, it is very repetitive.

Response:

Thank you very much for the comments.

In the previous paragraph in the manuscript, we found that optical simulation results of Gaussian spheres with different size parameters can qualitatively explain the measured discrepancies in scattering matrices for our loess samples, effective size parameters of which differ by 20 times, and Gaussian spheres may be promising in simulating scattering matrix for loess dust. In this paragraph, we further tried to use Gaussian spheres to explain differences of scattering matrices for other kinds of particles with different sizes, such as olivine and forsterite, and found that simulation results of Gaussian spheres cannot explain these differences. The reason may be that the effects of micro structure and refractive index become more obvious when the difference in size are relatively small, or it may be that Gaussian spheres cannot be used for other kinds of particles.

Therefore, in order to prevent readers from mistakenly thinking that Gaussian spheres may be universal for optical simulation of different kinds of particles, we think it is necessary to keep this paragraph in the revised manuscript. In addition, since our work focuses on loess dust, so we mentioned these statements here rather than in the Conclusions section. According to the comments, we have removed and modified repetitive descriptions in this paragraph in the revised manuscript:

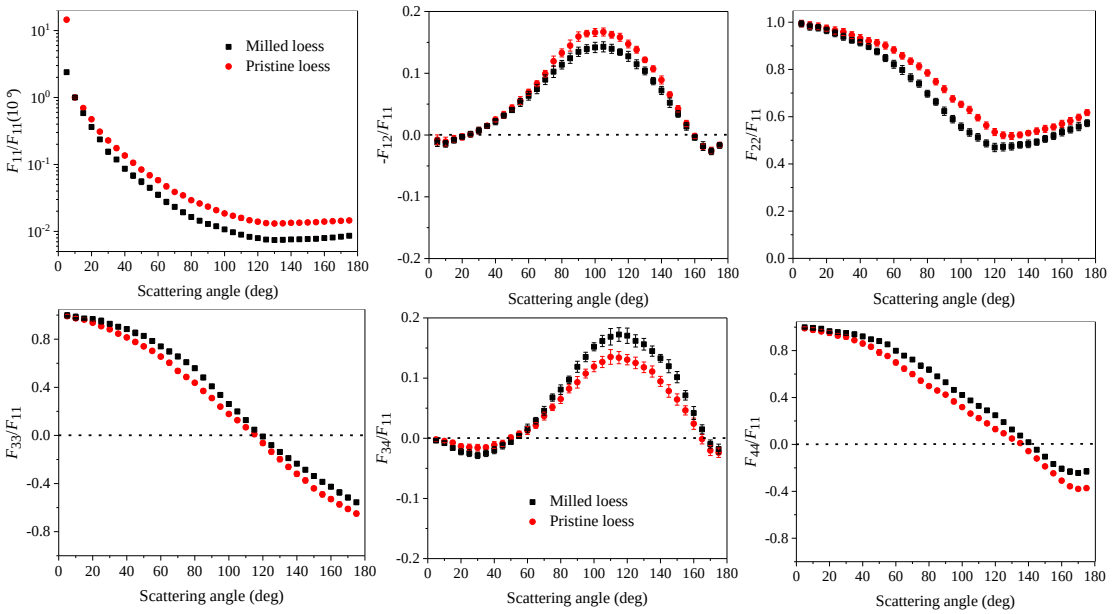
"In this work, a relatively good case is presented to show the effect of size distribution of loess dust on scattering matrices because effective radii of "pristine loess" and "milled loess" differ by more than 20 times. The influence of loess particle size is roughly verified through qualitative analyses of simulation results of Gaussian sphere, which deepen the understanding of this effect. For more detailed explanations, quantitative analyses are still needed based on much more optical simulations of Gaussian spheres. However, besides size distribution, physical properties such as refractive index and micro structure also play important roles in determining scattering matrices of dust particles. When the difference in particle size distributions or effective radii is relative small, the influences of other factors may become dominant or un-ignorable. This may be the reason why the effect of size distribution on measured scattering matrices for olivine samples cannot be concluded clearly (Muñoz et al., 2000). And this may also be the reason why effective radii cannot be used to explain all the

discrepancies in matrix elements for forsterite samples based on simulation results of Gaussian spheres (Volten et al., 2006b). Another reason may be that Gaussian spheres are not suitable models to reproduce scattering matrix for forsterite dust, as optical modelling of irregular mineral dust is still a challenging subject."

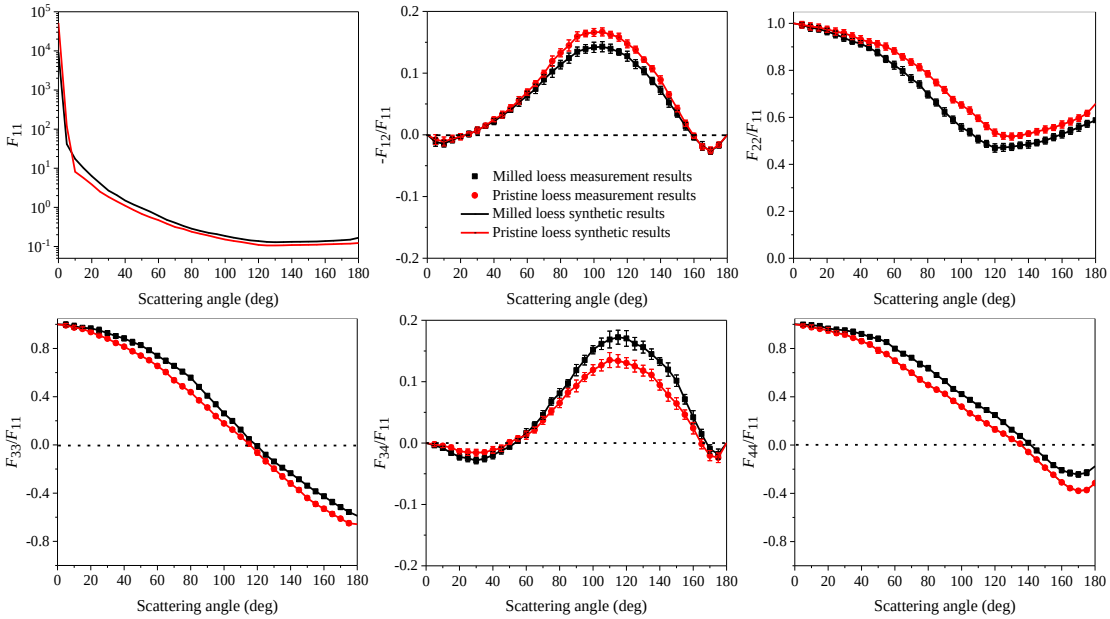
Figure 4/5: Could these be combined? You could have the synthetic scattering matrix as a different color and a line. It took me a while to see what the difference was between the 2 figures.

Response:

Thanks a lot for your comments. We have redrawn the synthetic scattering matrices in Figure 5 and added necessary descriptions in its caption in the revised manuscript:



"**Figure 4.** Measured non-zero scattering matrices for "pristine loess" and "milled loess". It should be noted that "milled loess" is the same sample as the "Luochuan loess" in Liu et al. (2019)."



"**Figure 5.** Synthetic scattering matrices for "milled loess" and "pristine loess". Lines are synthetic matrices and plots are measured values."

Figure 6: Could you specify all the samples that were used in this figure? Either here or in the text.

Response:

Thank you very much for the comments. We have added descriptions of the samples used to construct average matrix for loess dust in the revised manuscript:

"At last, the previously published average scattering matrix for loess, which consists of results for Hungary loess, milled Yangling loess and milled Luochuan loess (the latter two were sampled from CLP), was updated using new sample "pristine loess" from Luochuan, by averaging synthetic matrices for different loess samples."

318-319: "other factors ..." this is misleading, since there was no discussion on how the difference in RI affected the sample and no experiments were performed to single these factors out from the size effect. This is also in contrast to earlier where it is stated in line 239-240 "while other factors are also not ignorable"

Response:

Thanks a lot for your comments. It is very hard to separate single factor from others. In our study, qualitative analyses of simulation results of Gaussian spheres showed that the difference in sizes can be used to roughly explain these discrepancies in scattering matrices for two loess samples (Liu et al., 2015). Furthermore, analyses of optical simulation results showed that both refractive index and micro structure do affect scattering matrix to some degree, but these two factors cannot be used to explain all the

discrepancies in scattering matrix elements (Liu et al., 2015; Muinonen et al., 2007). Based on limited available literatures focusing on optical simulations, we think that these discrepancies in scattering matrices are mainly caused by differences in size distributions, while differences in factors such as refractive index and micro structure have relatively small and recessive contributions. We have modified the related descriptions in the revised manuscript:

"In summary, different factors have different or similar effects on a certain matrix elements. The discrepancies in scattering matrices for "milled loess" and "pristine loess" can be mainly interpreted from the perspective of difference of effective radii, while differences in other factors such as refractive index and micro structure have relatively small contributions, and Gaussian spheres may be promising models for simulating scattering matrix for loess dust."

"Qualitative analyses of optical simulations of various morphological model showed that the large difference in size distributions (effective radii differ by more than 20 times) caused by milling process plays a major role in leading to discrepancies in scattering matrices for these two samples, while differences in factors such as refractive index and micro structure have relatively small and recessive contributions."

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