Responses to the comments of reviewer 4

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 4:

The study presents light scattering measurements of Chinese loess dust. The authors have measured the scattering matrix elements of a single loess sample from the Chinese loess plateau once untreated (pristine loess) and once milled (milled loess) and performed some complementary measurements too. I find the topic very interesting and useful. However, I have some doubts about the paper being published in its current form.

Response:

Thank you very much for reviewing our manuscript and all these valuable comments and suggestions. We have tried our best to respond your comments point by point and modified related descriptions in the revised manuscript. And we hope that you will reconsider our manuscript.

First of all I have to question if the choice of the journal is adequate for the performed study. I might be wrong about this and if this is the case, then please just ignore this comment. However, this journal is called Atmospheric Measurement Techniques, and on its homepage it is stated that: “The main subject areas comprise the development, intercomparison, and validation of measurement instruments and techniques of data processing and information retrieval for gases, aerosols, and clouds.” This paper presents none of them. It shows some laboratory measurements with atmospheric relevance. It does not show a new measurement technique nor a newly developed instrument neither any instrument intercomparison. The only technical part of the paper is the one page section of 3.2 where the measurement apparatus is shortly introduced.

Response:

Thanks a lot for your comments.

We think our work can be classified into subject areas “techniques of information retrieval for aerosols”. Accurate retrievals of optical and physical properties of dust aerosols depend largely on the choice of suitable particle models of dust. So the model development for dust has always been worthy of attention, and we think non-spherical particle models for dust particles are still needed to be further verified or developed targeting for specific kinds of dust with different physical properties.

Chinese loess dust contributes a lot to Asian dust and is expected to affect radiative balance over both source and downwind regions. However, there is still no specific particle models for Chinese loess dust. Therefore, in our study, scattering matrices and essential physical properties of Chinese loess dust samples with different size distributions, which represent dust aerosols over source and downwind regions respectively to some extent, are investigated. All these measurement results are necessary constraints for the development of advanced particle models or the retrievals of best fitted shape distributions of widely used spheroid models (Dubovik et al., 2006). And we believe that these models will help to improve the retrieval accuracy of physical properties of Chinese loess dust aerosols over both source and downwind regions. Furthermore, the updated average scattering matrix for loess is also instructive to the model development of loess dust and useful for improving the retrieval accuracy of dust aerosol properties over other loess regions in the world.

My other main concern is: if the manuscript contains strong enough scientific material to be published in AMT. The scattering matrix element measurements of the two differently treated loess sample come from 6 single measurements, and the manuscript is based completely on this. It would considerably improve the manuscript if more measurements were included. To give you some ideas: include measurements and a comparison of different kind of loess samples collected either on the Chinese Loess Plateau at other places or get loess samples from outside of China. Another idea could be to include some other types of mineral dust and make a comparison. I know well, that it is not always possible to perform more measurements additionally. The manuscript could be improved with much thorougher discussion about comparing existing literature data with your dataset as well, or perform some numerical simulations based on the measured size distribution and shape (e.g. Mie theory and a theory for non-spherical particles) and discuss the results.

Response:

Thank you for the comments and suggestions.

In this study, we experimentally investigated scattering matrices as well as other basic physical properties of Chinese loess dust with two distinct size distributions, from a meaningful perspective of long range transport of dust particles. Furthermore, we explored reasons for the discrepancies in scattering matrices based on qualitative analyses of optical simulations in literatures, and updated the previous average scattering matrix for loess dust. All the discussions are focused on the loess dust.

Until now, experimental studies of scattering matrices are still very rare. Before our series of studies, only one Hungary loess sample was characterized at 441.6 and 632.8 nm wavelengths by Volten et al. (2001). In our previous study, fine loess particles sampled from two typical regions of Chinese Loess Plateau were investigated at 532 nm wavelength and compared with measurement results of Hungary loess (Liu et al., 2019). Comparisons showed that measured scattering matrices for different samples have good consistencies, thus an average scattering matrix for loess dust was built. The average scattering matrix for loess published in our previous study (Liu et al., 2019) is called as “previous average scattering matrix” in the current study, and we updated it using new coarse “pristine loess” sample. Therefore, in other words, the differences between average matrix before and after update are also the differences between “pristine loess” and the other three samples, and differences among these three samples can be referred to Liu et al. (2019). As can be seen in Figure 6 in the manuscript, compared to other three samples, phase function for “pristine loess” has larger forward scattering peaks and smaller values at side and back scattering directions. “Pristine loess” has larger $-F_{12}/F_{11}(\theta)$ values at near side scattering angles, has larger $F_{22}/F_{11}(\theta)$ values at almost all scattering angles, and has smaller values of both $F_{33}/F_{11}(\theta)$ and $F_{44}/F_{11}(\theta)$ at backscattering angles, when compared with the other three samples.

As for discrepancies in scattering matrices among different kinds of mineral dust, Volten et al. (2001) had already made such comparisons, these discrepancies are obvious, but it is still very hard to discriminate dust types using angular distributions of matrix elements. This is because physical properties such as size distribution, refractive index and micro structure are all different to some degree, and direct and rough comparison may be not so meaningful. In our another previous work (Liu et al., 2018), we compared scattering matrices for anthropogenic cement dust with that for natural mineral dust, it is also difficult to discriminate one certain dust type from others based on scattering matrices. We did not made such analysis in our manuscript, because the average scattering matrix for loess may also cover measurement results of other dust types, and this will confuse readers. And there is an essential underlying premise for the application of average scattering matrix for loess dust, that is the tracing of airborne dust using models like HYSPLIT Model ensures its source is loess regions.

Many studies had shown that Mie calculations of spheres cannot reproduce measured scattering matrices for mineral dust at all (Meng et al., 2010; Merikallio et al., 2015; Mishchenko et al., 2003). Therefore, we did not make direct comparisons between Mie calculations and measured scattering matrices, because no more information can be extracted except that non-spherical shape of loess. In contrast, we are more prefer to conduct optical modeling with non-spherical models. However, optical modeling of irregular dust particles with large sizes is still a very challenging subject, and only few researcher focus on it. We had tried to contact these experts for cooperation, but didn't get any response. We want to attract interest of modeling experts by presenting some meaningful experimental results, and we hope to establish cooperation with these experts in future, since only combinations of experiments and optical simulations can make our work more complete and useful. Even so, in the subsection “4.1 Experimentally Determined Scattering Matrices”, we tried to find the main factor that resulting in these distinctions in measured scattering matrices for two loess samples based on qualitatively analyses of numerical simulations, simulation results of non-spherical Gaussian particles and agglomerated debris particles were selected for analyses. And we found that Gaussian spheres with effective radii same as measured size distribution of loess samples can qualitatively explain these measured distinctions in scattering matrices.

We have modified related descriptions about comparisons between previous and updated average scattering matrix in 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. In other words, the differences between average matrix before and after update are also the differences between “pristine loess” and the other three samples, and differences among these three samples can be referred to Liu et al. (2019). As shown in Figure 6, compared to other three samples, phase function for “pristine loess” has larger forward scattering peaks and smaller values at side and back scattering directions. “Pristine loess” has larger $-F_{12}(\theta)/F_{11}(\theta)$ values at near side scattering angles, has larger $F_{22}(\theta)/F_{11}(\theta)$ values at almost all scattering angles, and has smaller values of both $F_{33}(\theta)/F_{11}(\theta)$ and $F_{44}(\theta)/F_{11}(\theta)$ at backscattering directions, when compared with the other three samples.”

You could also improve the paper by stating clearly what your main message is for the reader. You just present the scattering matrix elements but do not draw any further conclusions. How is Chinese loess scattering treated in radiative transfer models? Will there be a big difference if these models are updated with your results? How representative is your single loess sample?

Response:

Thanks for your valuable comments.

The main messages for readers in this study can be concluded as three aspects: (1) there are obvious discrepancies in measured scattering matrices for Chinese loess dust with different size distributions, and these discrepancies are different from that for other kinds of mineral dust with various size distributions. (2) Qualitative analyses of numerical simulation results in literatures showed that the large difference in size distributions (effective radii differ by more than 20 times) plays a major role in leading to these discrepancies in scattering matrices. And Gaussian spheres may be promising models for simulating scattering matrix for Chinese loess dust, but more detailed quantitative verifications using measured size distributions and refractive indices are still needed. (3) The previously published average scattering matrix for loess dust was updated using measurements of new coarse loess sample, which is meaningful for validating existing models and developing more advanced models suitable for optical simulations of loess dust, and finally helps to retrieve dust aerosol properties with higher accuracy over both source and downwind areas.

To our best knowledge, there is no study focus on the selection of optical model for Chinese loess in radiative transfer models. As for optical models for mineral dust, Dubovik et al. (2006) used simulated scattering matrices of spheroid models with different aspect ratios to reproduce measured results for different kinds of mineral dust published by Volten et al. (2001), and a best-fitted shape distribution of spheroids was recommend. 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 conducted based on this shape distribution. Tian et al. (2019) retrieved the total aspect ratio distributions of spheroids for all kinds of aerosols over Chinese Loess Plateau from depolarization ratios observed by lidars, however, aspect ratio distributions for loess dust still cannot be separated. Furthermore, optical simulations and radiation transfer calculations conducted by Li et al. (2019) showed that shape distributions of spheroids have obvious effects on scattering matrices and further affect radiance distribution and polarization properties of sky light. Therefore, we think the best fitted shape distributions of spheroids for loess dust with distinct sizes are still highly in demand, and the accuracy of retrieved dust aerosol properties will be further improved with the help of these best fitted models.

As mentioned in manuscript, original loess sample was collected from Luochuan Loess National Geological Park, the only national park for loess landform in China. So this sample represents Chinese loess to some extent, but it cannot represent all loess distributed in China. In our previous work (Liu et al., 2019), we investigated fine loess particles sampled from Luochuan and Yangling, the latter located at the southern edge of Chinese Loess Plateau. Even through measured scattering matrices have good consistencies, there are still obvious discrepancies in the angular distributions of matrix elements, and these discrepancies even larger than the differences between loess with different size distributions in this study. This means local variations of loess also have significant effect on scattering matrix.

The measurement results of all these loess samples were included in the average scattering matrix for loess, however, these samples may still cannot represent all loess in China, even all loess in Chinese Loess Plateau. Therefore, we will further update the average scattering matrix for loess dust in future using measurements of more samples collected from different regions of China and more samples with different sizes.

We have added necessary descriptions in revised manuscript:

"Fine loess dust sampled from Luochuan and Yangling, two regions of Chinese Loess Plateau, were investigated by Liu et al. (2019). Local variations of loess dust also have obvious effects on the measured scattering matrices. It should be noted that all these samples investigated may still cannot completely represent the loess in Chinese Loess Plateau and China, so one of the efforts in the future is to investigate more loess samples collected from more regions and with more size distributions, accordingly, the average scattering matrix for loess will be updated constantly."

You probably cannot implement all of my main suggestions to improve the manuscript, and it is not necessary either. I just wanted to show you some possible options how it could be done. The data and the work you do is valuable but I only can recommend the manuscript's publication if it is significantly improved.

Response:

Thank you very much for all these meaningful comments and suggestions. We have tried our best to response the comments and revise our manuscript. We also explained the reasons for some of your suggestions cannot be implemented right now, and listed them as our future works. And we hope that you can re-consider our manuscript.

Other Comments:

1. I suggest a careful English language editing of the manuscript.

Response:

Thanks for your suggestion. We have tried our best to correct language mistakes by repeatedly reviewing the manuscript, and we also have invited native speakers to edit the manuscript.

2. Page 4, Lines 99-111: Even after a longer search I could not find details about the SALD-2300 instrument and how it exactly measures the particle size distribution and refractive index. Please add details how it exactly works. What I think it does is measuring the light scattering at many angles and trying to reproduce the measurement with a guessed number size distribution and a refractive index using theoretically calculated scattering values. Does it use the Mie theory (which is valid for spherical particles only)? Or how can it calculate the scattering for particles with unknown shape? How does it influence your derived number size distribution and refractive index? What is the uncertainty of this measurement method for non-spherical particles? Please add a discussion on this. Are you sure that the refractive index difference between 1.65+0i for "pristine loess" and 1.70+0i for "milled loess" is real?

Response:

Thank you very much for the comments.

As you point out, SALD-2300 measures the angular distribution of scattered light intensity, then many combinations of values of number size distribution and refractive index are employed for Mie calculations to reproduce the measured light intensity distribution, the best fitted size distribution and refractive index of sample are obtained at last. SALD-2300 has 84 light detectors in all, including 78 forward detector elements, one side detector and five back detectors. Liu et al. (2003) revealed that Mie theory can be used to reproduce forward scattering intensities of nonspherical particles with moderate aspect ratios at scattering angles smaller than 20 °. Since over 70% of the detectors of SALD-2300 are set at angles smaller than 20 °, so we think the retrieved size distributions of nonspherical loess dust are of high accuracy. It is hard to further evaluate uncertainty of measured size distribution, because there is still no complex model for loess dust suitable for its optical simulation, which is the final goal of our work.

The smallest calculation steps of real and imaginary part of refractive index for SALD-2300 is 0.05 and 0.01, and these two values are chosen to retrieve refractive index for loess samples. All three repeat measurements obtained the same refractive indices for both “pristine loess” and “milled loess”. Kinoshita (2001) retrieved refractive indices for alumina dust with different sizes using the same method as SALD-2300, there were also small difference 0.05 in the retrieved real part, and he explained this phenomenon as the effect of nonspherical property of dust. So we think there is indeed difference in the retrieved refractive indices for the two loess dust samples with distinct size distributions. And this is also due to the nonspherical nature of loess particles. From the perspective of numerical simulation, the effect of refractive index on angular distribution of scattered light intensity of nonspherical complex particles are still unclear enough (Muinonen et al., 2007; Zubko et al., 2013). Furthermore, to our best knowledge, there is still no certain conclusion of refractive index of loess, so it is hard to evaluate the uncertainty of our retrieved results.

We have added more detailed descriptions about SALD-2300 in the revised manuscript:

“SALD-2300 has 84 scattering light detectors in all, including 78 forward detector elements, one side detector and five back detectors. The best fitted number size distribution and refractive index *m* can be obtained by reproducing measured angular distribution of light intensity based on Mie calculations. Liu et al. (2003) revealed that Mie theory can be used to reproduce forward scattering intensities of nonspherical particles with moderate aspect ratios at scattering angles smaller than 20 °. Since over 70% of the detectors of SALD-2300 are set at angles smaller than 20 °, the retrieved size distributions of nonspherical loess dust based on Mie theory are of relatively high accuracy. 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.”

3. Page 4, Lines 110-111: “larger particles have relatively larger real part of refractive index”: if I understood correctly your method of producing the milled loess sample, it contains exactly the same material (your chemical analysis verifies it) as the pristine

loess and therefore one would expect the two samples having the same refractive index. Are you sure, again, that your result is real and are not only a measurement artifact/uncertainty? Or do you think that the milling caused some strange structural changes in the loess sample which homogenized or inhomogenized how the chemical components are distributed within a single particle and/or between the particles?

Response:

Thanks a lot for the comments.

First of all, there is a clerical error in this sentence, it should be "larger particles have relatively smaller real part of refractive index". There is no doubt that refractive index of specific material is unique. And we don't think the milling process obviously modified the distribution of chemical components within a single and between the particles.

Retrieved refractive index of particles based on measured light intensity distribution is a kind of optically equivalent refractive index, it is close to inherent refractive index of the measured material but not necessarily the same. Kinoshita (2001) retrieved refractive indices for alumina dust samples with 1 μm and 5 μm diameter. The inherent refractive index of alumina is known as 1.76, while the retrieved real parts are 1.80 and 1.75 respectively, larger particles have smaller real part of refractive index and the difference is small but cannot be ignored. Kinoshita explained this phenomenon as the effect of nonspherical nature. Our study also found larger particles have slightly smaller real part of refractive index, so we think this difference is real and can be explained by the same reason.

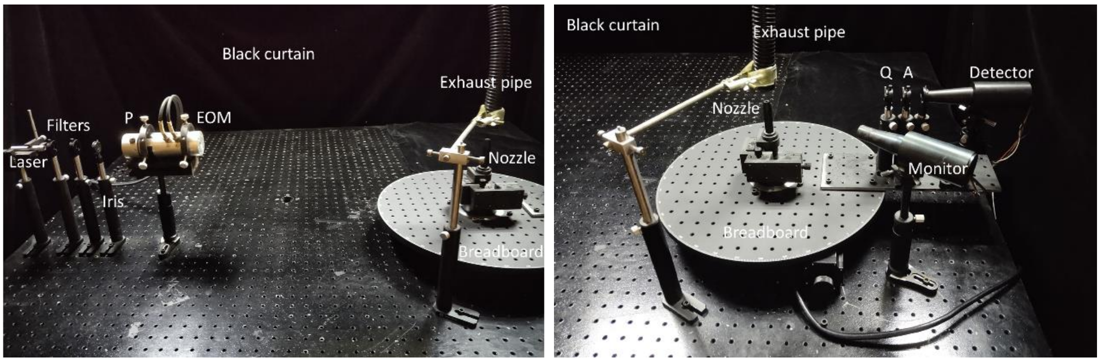
We have modified the mentioned clerical error and added necessary discussions in revised manuscript:

"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."

4. Page 6, Section 3.2: I assume that this is not the first paper which uses this experimental apparatus. Please add a reference to the paper where a more detailed description of your instrument is available. If there is no such paper, please add a more detailed description.

Response:

Thank you for pointing this out. During the revision of the manuscript, we improved the experiment apparatus by extending the maximum angle coverage from 160 °to 175 °, the photograph of improved apparatus are shown below, and re-measured scattering matrices for the two loess samples.



Newly measured scattering matrices were in good agreement with measurement results using the previous apparatus in the range of 5-160°. As we mentioned in the original manuscript: “For more details, it can be referred to Muñoz et al. (2010) and Liu et al. (2018).” We are sorry that this description may be not clear enough and confusing. Therefore, we have modified the confused descriptions and added more details of the improved apparatus in the revised manuscript:

“The main improvement is that angle coverage at backscattering angles are extended to 175°, while the maximum coverage of previous apparatus is 160° (Liu et al., 2018).”

“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*.”

“Furthermore, the improved apparatus is validated using water droplets. Measured all six non-zero scattering matrix elements for water droplets can be well fitted using Mie calculation results, indicating that the measurement accuracy of apparatus are satisfactory. For more details about the measurement principle and validation method of the apparatus, it can be referred to Liu et al. (2018).”

5. Page 6, Lines 59-62: Since your main results are the measured matrix elements, probably it would be worth explaining exactly from which polarization states which matrix elements were derived and how, and not only referencing a paper for it.

Response:

Thanks a lot for your suggestions. We have add a new table and more details about the relationship of combinations of optical elements and matrix elements in the revised manuscript:

“All the matrix elements of dust samples can be determined as functions of scattering angles with the help of various combinations of orientation angles of above optical elements as shown in Table 3, which is just the same as Muñoz et al. (2010).”

“**Table 3.** Combinations of orientation angles of optical axis of all the optical elements.”

Combination	γ_P	γ_{EOM}	γ_Q	γ_A	$DC(\theta)$	$S(\theta)$	$C(\theta)$
1	45 °	0 °	-	-	$F_{11}(\theta)$	$-F_{14}(\theta)$	$F_{13}(\theta)$
2	45 °	0 °	-	0 °	$F_{11}(\theta)+F_{21}(\theta)$	$-F_{14}(\theta)-F_{24}(\theta)$	$F_{13}(\theta)+F_{23}(\theta)$
3	45 °	0 °	-	45 °	$F_{11}(\theta)+F_{31}(\theta)$	$-F_{14}(\theta)-F_{34}(\theta)$	$F_{13}(\theta)+F_{33}(\theta)$
4	45 °	0 °	0 °	45 °	$F_{11}(\theta)+F_{41}(\theta)$	$-F_{14}(\theta)-F_{44}(\theta)$	$F_{13}(\theta)+F_{43}(\theta)$
5	90 °	-45 °	-	-	$F_{11}(\theta)$	$F_{14}(\theta)$	$-F_{12}(\theta)$
6	90 °	-45 °	-	0	$F_{11}(\theta)+F_{21}(\theta)$	$F_{14}(\theta)+F_{24}(\theta)$	$-F_{12}(\theta)-F_{22}(\theta)$
7	90 °	-45 °	-	45 °	$F_{11}(\theta)+F_{31}(\theta)$	$F_{14}(\theta)+F_{34}(\theta)$	$-F_{12}(\theta)-F_{32}(\theta)$
8	90 °	-45 °	0 °	45 °	$F_{11}(\theta)+F_{41}(\theta)$	$F_{14}(\theta)+F_{44}(\theta)$	$-F_{12}(\theta)-F_{42}(\theta)$

"Multiple groups of values of measurable quantities, that is the DC component $DC(\theta)$, first harmonics $S(\theta)$ and second harmonics $C(\theta)$ of voltage signal, are recorded at every scattering angle for each combination of optical elements. The first step of data processing is to average these recorded values and get their errors. The optical platform is surrounded by black curtains to avoid the effect of environmental stray light, and background signals need to be measured and subtracted. Fluctuations of dust aerosols can be eliminated by normalizing measurements of the "detector" using $DC(30°)$ measured by the "monitor". Scattering matrix elements can be extracted from preprocessed $DC(\theta)$, $S(\theta)$ and $C(\theta)$ according to Table 3. Subsequently, $F_{11}(\theta)$ is normalized to 1 at 10 ° scattering angle, and the remaining matrix elements $F_{ij}(\theta)$ are normalized to $F_{11}(\theta)$ at the same angle. At last, whether measurement results of scattering matrix satisfy Cloude coherency matrix test should be examined (Hovenier and Van Der Mee, 1996). Three iterations of measurements are performed for each particle sample, the final results are average of three groups of experiments, and the errors are also calculated which contain errors during every measurement and errors for repeat measurements."

6. Page 7, Line 193: "all six non-zero matrix elements are limited to narrow regions, respectively" I don't understand what you mean here. What narrow regions? Angle range? Y-value range? Or do you mean that your error bars are small? Please clarify!

Response:

Thank you very much for pointing this out. What we want to say in this sentence is that matrix elements for both "pristine loess" and "milled loess" present similar angular behaviors. More specifically, angular distributions of all six non-zero matrix elements, in other words Y-values in each sub plot, are limited to narrow regions, respectively. We have modified these confused descriptions in the revised manuscript:

"Matrix element ratios for "pristine loess" and "milled loess" present similar angular behaviors, more specifically, angular distributions of all six non-zero matrix element ratios are limited to narrow regions, respectively."

7. Page 7, Lines 199-201: Please comment on the angular behavior of F_22. Next to it: it looks like that the milled loess sample deviates more from unity than the pristine loess sample. Does this suggest that the milled loess has a more irregular shape than pristine loess?

Response:

Thanks a lot for your valuable comments. As we mentioned in manuscript, $F_{22}(\theta)/F_{11}(\theta)$ equals to constant 1 when particles are homogeneous spheres, otherwise, particles are nonspherical and irregular. However, this does not mean that different $F_{22}(\theta)/F_{11}(\theta)$ measurement results can directly indicate the discrepancy of particle irregularity. Because optical simulations of nonspherical Gaussian particles conducted by Liu et al. (2015) showed that $F_{22}(\theta)/F_{11}(\theta)$ values are not only sensitive to particle irregularity but also to particle size. We have added necessary discussions in the revised manuscript:

"Experimentally determined $F_{22}(\theta)/F_{11}(\theta)$ values of "milled loess" are larger than "pristine loess", especially at side and back scattering angles. It should be noted that discrepancies in measured $F_{22}(\theta)/F_{11}(\theta)$ cannot be directly used to indicate difference of particle irregularity, because optical calculations of Gaussian spheres showed that $F_{22}(\theta)/F_{11}(\theta)$ values are sensitive to not only particle irregularity but also to size distribution (Liu et al., 2015)."

8. Page 7, Lines 206-207: The sentence is very confusingly phrased, please rephrase it. I am not sure if I understood what you wanted to tell the reader but I don't see any significant difference between the 5° relative phase functions.

Response:

Thank you for pointing this out. There is a drawing error in subplot $F_{11}/F_{11}(10°)$, we are very sorry about that. In the original manuscript, the display range of $F_{11}/F_{11}(10°)$ was set as 0.005-10 in log scale, but relative phase function at 5° for "pristine loess" is about 15, this can be checked from the dataset previously published by us at https://doi.org/10.5281/zenodo.3361852. We have corrected this error by resetting the maximum display value as 20 in subplot $F_{11}/F_{11}(10°)$ in revised manuscript:

"On the other hand, the discrepancies in matrix elements for "pristine loess" and "milled loess" are still obvious. Compared to "milled loess", there is an enlargement of relative phase function at 5° scattering angle for "pristine loess"."

9. Page 7, Lines 206-208: From F11 it looks like that milled loess has a higher forward to backward scattering ratio than pristine loess. I would expect exactly the other way around because the pristine loess sample contains much larger particles and larger particles usually have a much higher forward scattering compared to the backward scattering value. Please comment on it.

Response:

Thanks a lot for you valuable comments. Similar to the response to Comment 8, we are sorry there is an error in subplot $F_{11}/F_{11}(10°)$. And we have corrected it in the revised manuscript. We agree that larger particles have much higher ratios of forward scattering to backward scattering. In our study, the forward (5°) to backward scattering (175°) ratio of “pristine loess” is about 3.60 times larger than that of “milled loess”.

10. Page 8, Lines 119-223: Is there no way to produce samples containing smaller particles than the original without changing their form? Just by sieving the sample (the size distribution of the pristine loess seems to me broad enough)? Would that not work? If it would, then measuring such samples could save you from speculating about, if the measured differences are due to the different size or shape. It would be also very nice to have more samples with different sizes and not only two. You show that the particle size differs much more than the shape between the two samples, and your speculations might be true as well. However, how can you be sure that every component of the scattering matrix is comparably sensitive to the changes in size and shape? Let’s assume, that one matrix component is 1000 times more sensitive to the changes in the particle shape than to the changes in the size? Please provide some proof that such a case is not to be expected, and then your argumentation becomes valid.

Response:

Thank you very much for your meaningful comments.

During our sample preparation stage, we did try to use 20 μm and 10 μm sieves to obtain loess samples with different size distributions, but we ended in failure. Only very few particles were obtained using 20 μm sieve, which is far from meeting the requirement of light scattering matrix measurements, and there were almost no particles can be obtained using 10 μm sieve. We also did not find other available methods that can be used to prepare enough particles for experiments based on the limited original loess samples, so we use ball milling method. We think ball milling can modified particle shapes to some extent, but we also have a question that whether the particles with different sizes in original loess sample can be described by the same morphology, since, to our best knowledge, there is still no effective method to adequately describe real morphologies of irregular dust particles using several parameters.

For loess particles with effective radii smaller than “pristine loess” but larger than “milled loess”, it can be summarized from optical simulations of Gaussian spheres that both size and irregularity have roughly similar effects on matrix element ratios F_{33}/F_{11}, $F_{34}/F_{11}$and F_{44}/F_{11}, so it is almost impossible to tell which of the two factors plays a major role (Liu et al., 2015). In such cases, qualitative analysis is far from enough, only quantitative analysis in cooperation with optical model experts can separate the effects of size and irregularity. So we did not investigate samples with effective radii between “pristine loess” and “milled loess” in this study, and we want to investigate such samples in combination with quantitative optical simulations by cooperating with optical modeler, only in this way can our research more meaningful.

As far as we know, it is hard to evaluate the sensitivity of each scattering matrix element to the changes of size and irregularity, because the effects of these two factors are usually both complex and even coupled. Another reason is that it is very hard to use morphological models to adequately describe real dust particles, so the determination of variation ranges of model morphological parameters is hard, and then it is hard to assess the effects of size and irregularity using the same relative change standard. Liu et al. (2015) calculated scattering matrices for Gaussian spheres whose size parameter ranges from 1 to 1000 and standard deviation of radial distance (irregularity) ranges from 0 to 0.2. The effective size parameters of “pristine loess” and “milled loess” are about 580 and 30 respectively. As this parameter increases from 30 to 580, there are significant variations in matrix elements, the variations of F_{11}, -F_{12}/F_{11}, F_{33}/F_{11} and F_{44}/F_{11} are more obvious than the effect of irregularity increasing from 0.05 to 0.2, the sensitivity differences of F_{22}/F_{11} and F_{34}/F_{11} on size and irregularity are definitely less than 1000 times. It should be noted that the comparisons are not rigorous enough, because the irregularity range 0.05-0.2 may not exactly applicable to our loess samples. On the other hand, commercial laser particle size analyzers such as SALD-2300 employ Mie theory to retrieve size distribution of irregular dust particles based on light intensity distribution (matrix element F_{11}), this is because F_{11} is more sensitive to size than particle irregularity, especially in forward scattering angles, otherwise these instruments cannot be used to measure particle size at all.

11. Page 8, Lines 224-240: It would considerably strengthen the manuscript if numerical calculations based on your measured size distribution and particle shape were added and not only the existing literature was analyzed. If that is not possible, you should show how the size and shape of your samples compare to the size and shape of the particle in the referenced papers. Irregular dust does not necessary mean comparable size distribution and/or particle shape.

Response:

Thanks a lot for the valuable comments.

The morphology of irregular dust particle is difficult to be adequately described by several parameters, even the most advanced models cannot always correspond to real particles directly. Until now, researches on optical simulations of irregular particles are still very rare. Simulations of agglomerated debris model and rough Gaussian model only cover very small size range of dust particles and calculations of larger particles are very time-consuming (Zubko et al., 2007, 2013). Therefore, the influence of morphological parameters such as size and irregularity can only be roughly summarized and extended to large particles. However, optical simulations of Gaussian sphere model cover most of the particle size distributions of our loess samples, so they are used for direct qualitative analyses in our study (Liu et al., 2015).

Furthermore, quantitative analyses can only be performed using measured particle size distributions, morphological parameters are very hard to be taken into consideration. The most advanced optical modeling method for dust particles can only employ a few irregular shape models with specific morphological parameters (which cannot correspond to real dust particles directly), and the measured particle size distributions are employed. Then, calculation results were used to reproduce the measured scattering matrix, the best fitted number fractions of irregular shape models mentioned above (shape distributions) can be retrieved finally, and these shape distributions represent particle irregularity to some extent. Optical modeling of irregular dust is still an urgent and challenging problem. In future, we hope to combine experimental measurements and optical simulations of models much closer to real morphology of dust by cooperating with optical modeling experts to make our investigations more meaningful.

12. Page 9, Section 4.2: During calculating the synthetic scattering matrices you follow the works of Dabrowska et al., 2015 and Escobar-Cerezo et al., 2018. They used the very same measurement technique, had only different kind of samples (Lunar and Martian dust). You clearly follow their work, by extrapolating the measurements to the angles you could not measure as well. The extrapolation in the forward direction is based on the Mie theory and is performed for a narrow angle range of 0-3◦ or 0-5◦ (in your case). This is for me a justified assumption. However, in the backward region, the extrapolation is based on a polynomic fit and not on any kind of scattering theory. In this case, I can believe that it works well for the very narrow 177-180◦ angle range in the works of Dabrowska et al., 2015 and Escobar-Cerezo et al., 2018. But you applied it for a much broader angle range of 160-180◦, and here I really need some solid proof of this method being justified. The later calculated back-scattering depolarization ratio values cannot be accepted either before your extrapolation is not verified.

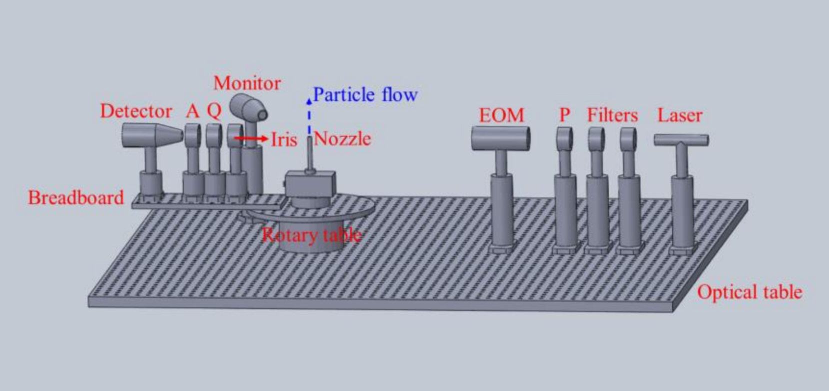
Response:

Thank you very much for the constructive comments.

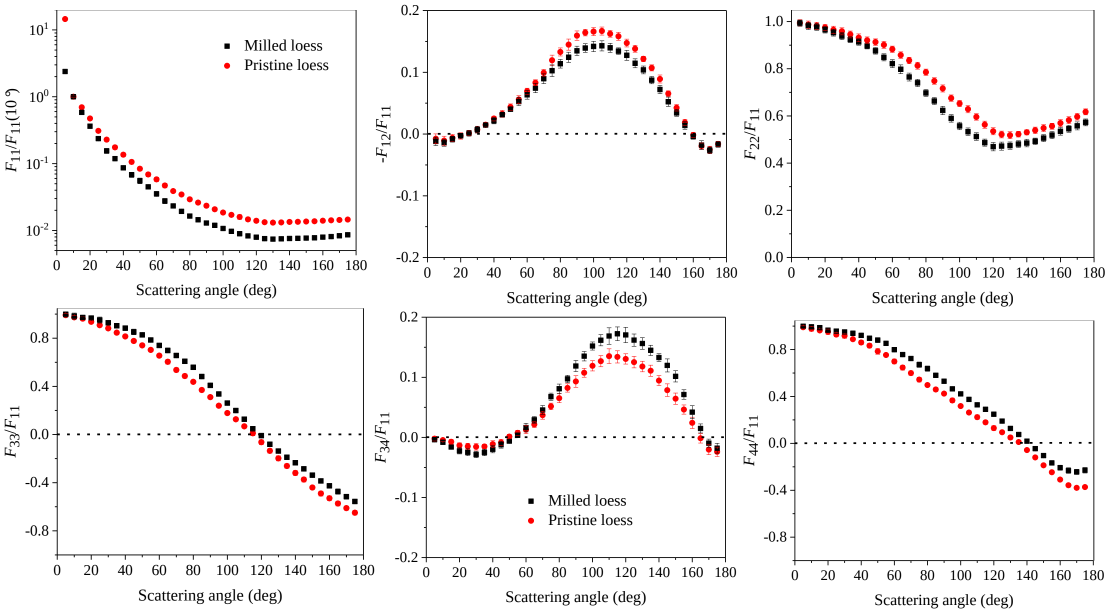
We re-measured scattering matrices for both “pristine loess” and “milled loess” using an improved matrix measurement apparatus covering scattering angles from 5 °to 175 °. Newly measured scattering matrices were in good agreement with measurement results using the previous apparatus in the range of 5-160 °. We also re-constructed synthetic scattering matrices for these two loess samples based on the measurements over 5-175 °, and we think the extrapolated results at 180 °angle are much more rigorous than before. Based on extrapolated values of F_{22}/F_{11} at 180 °, we re-calculated backscatter depolarization ratios for these two loess samples. At last, we re-updated average scattering matrix for loess dust.

Accordingly, we have modified the related descriptions of apparatus, experimental results, synthetic scattering matrices and average scattering matrix in the revised manuscript, and we also re-drawn Figures 3-6, as shown below. In addition, we re-uploaded measured results to a new dataset, which is available at https://github.com/liujia93/Scattering-matrix-for-loess-dust.

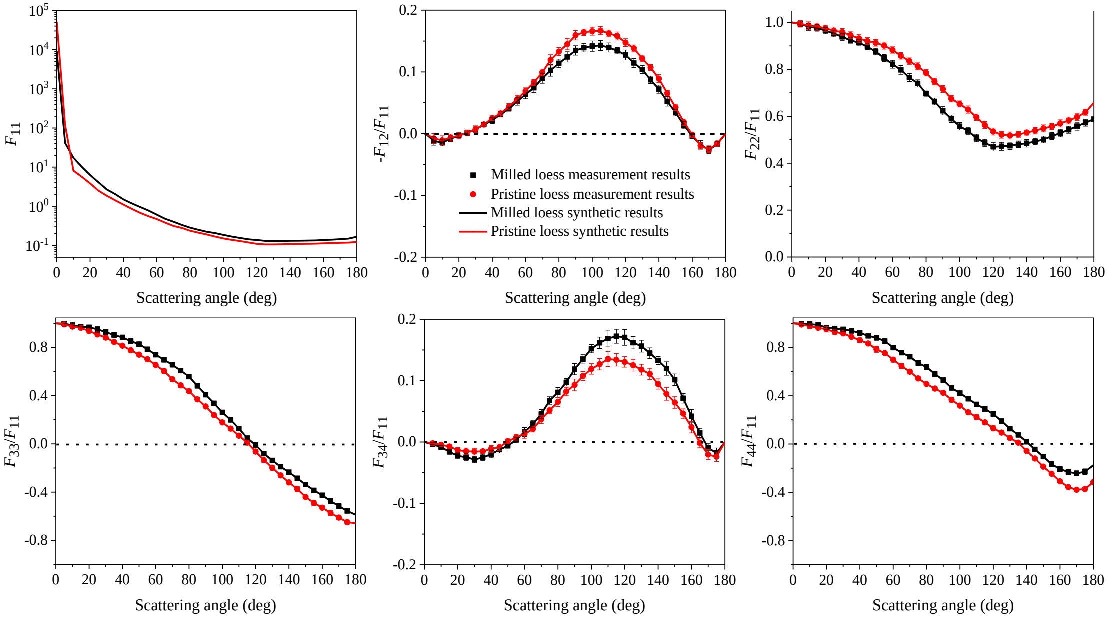
We have made necessary modifications in the revised manuscript:



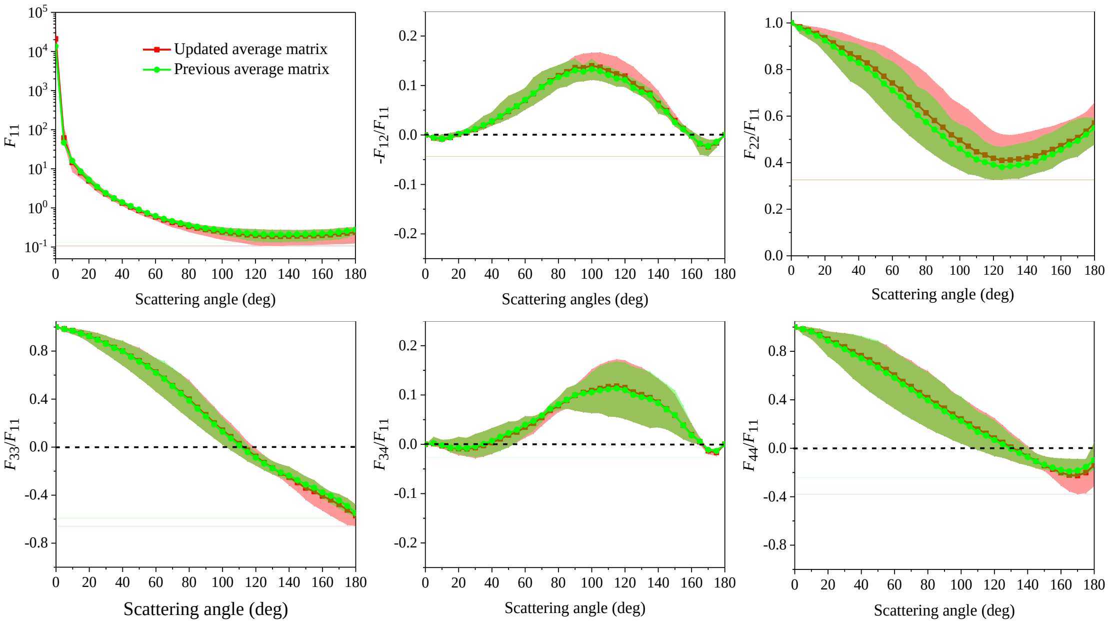
“Figure 3. Layout diagram of the experimental apparatus after backscattering angle expended.”



“**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.** Previous average scattering matrix (green lines and solid circles) (Liu et al., 2019) and updated average scattering matrix (red lines and solid squares) for loess dust. Reddish and green shadows stand for the areas covered by results for different loess samples with or without “pristine loess” included, respectively.”

Technical Comments: I did not do any language/technical correction because the manuscript needs a bigger revision.

Response:

Thank you again for all the valuable comments. We have tried our best to response these comments and modified related descriptions in the revised manuscript. We also have tried our best to correct language mistakes in the manuscript. We hope that you can re-consider our manuscript.

References

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B. N., Mishchenko, M., Yang, P., Eck, T. F., Volten, H., Muñoz, O., Veihelmann, B., van der Zande, W. J., Leon, J. F., Sorokin, M., and Slutsker, I.: Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, Journal of Geophysical Research: Atmospheres, 111(D11), doi:10.1029/2005JD006619, 2006.

Dubovik, O., Herman, M., Holdak, A., Lapyonok, T., Tanré, D., Tanré, J. L., Ducos, F., Sinyuk, A., and Lopatin, A.: Statistically optimized inversion algorithm for enhanced retrieval of aerosol properties from spectral multi-angle polarimetric satellite observations, Atmospheric Measurement Techniques, 4, 3255-3267, doi:10.5194/amt-4-975-2011, 2011.

Espinosa, W. R., Martins, J. V., Remer, L. A., Dubovik, O., Lapyonok, T., Fuertes, D., Puthukkudy, A., Orozco, D., Ziemba, L., Thornhill, K. L., and Levy, R.: Retrievals of aerosol size distribution, spherical fraction and complex refractive index from airborne in situ angular light scattering and absorption measurements, Journal of Geophysical Research: Atmospheres, 124 , 7997-8024, https://doi.org/10.1029/2018JD030009, 2019.

Kinoshita, T.: The method to determine the optimum refractive index parameter in the laser diffraction and scattering method, Advanced Powder Technology, 12.4, 589-602, https://doi.org/10.1163/15685520152756697, 2001.

Li, L., Li, Z., Dubovik, O., Zhang, X., Li, Z., Ma, J., and Wendisch, M.: Effects of the shape distribution of aerosol particles on their volumetric scattering properties and the radiative transfer through the atmosphere that includes polarization, Applied optics, 58(6), 1475-1484, https://doi.org/10.1364/AO.58.001475, 2019.

Liu, L., Mishchenko, M. I., Hovenier, J. W., Volten, H., and Muñoz, O.: Scattering matrix of quartz aerosols: comparison and synthesis of laboratory and Lorenz–Mie results, Journal of Quantitative Spectroscopy and Radiative Transfer, 79, 911-920, https://doi.org/10.1016/S0022-4073(02)00328-X, 2003.

Liu, J., Yang, P., and Muinonen, K.: Dust-aerosol optical modeling with Gaussian spheres: Combined invariant-imbedding T-matrix and geometric-optics approach, Journal of Quantitative Spectroscopy and Radiative Transfer, 161, 136-144, http://doi.org/10.1016/j.jqsrt.2015.04.003, 2015.

Liu, J., Zhang, Y., and Zhang, Q.: Laboratory measurements of light scattering matrices for resuspended small loess dust particles at 532 nm wavelength, Journal of Quantitative Spectroscopy and Radiative Transfer, 229, 71-79, https://doi.org/10.1016/j.jqsrt.2019.03.010, 2019.

Liu, J., Zhang, Y., Zhang, Q., and Wang, J.: Scattering Matrix for Typical Urban Anthropogenic Origin Cement Dust and Discrimination of Representative Atmospheric Particulates, Journal of Geophysical Research: Atmospheres, 123(6), 3159-3174, https://doi.org/10.1002/2018JD028288, 2018.

Meng, Z., Yang, P., Kattawar, G., Bi, L., Liou, K., and Laszlo, I.: Single-scattering properties of tri-axial ellipsoidal mineral dust aerosols: A database for application to radiative transfer calculations, Journal of Aerosol Science, 41, 501-512, doi:10.1016/j.jaerosci.2010.02.008, 2010.

Mishchenko, M. I., Geogdzhayev, I. V., Liu, L., Ogren, J. A., Lacis, A. A., Rossow, W. B., Hovenier, J. W., Volten, H., and Muñoz, O.: Aerosol retrievals from AVHRR radiances: effects of particle nonsphericity and absorption and an updated long-term global climatology of aerosol properties, Journal of Quantitative Spectroscopy and Radiative Transfer, 79, 953-972, doi:10.1016/S0022-4073(02)00331-X, 2003.

Merikallio, S., Muñoz, O., Sundström, A. M., Virtanen, T. H., Horttanainen, M., Leeuw, G. d., and Nousiainen, T.: Optical modeling of volcanic ash particles using ellipsoids, Journal of Geophysical Research: Atmospheres, 120(9), 4102-4116, https://doi.org/10.1002/2014JD022792, 2015.

Muinonen, K., Zubko, E., Tyynelä, J., Shkuratov, Y. G., and Videen, G.: Light scattering by Gaussian random particles with discrete-dipole approximation, Journal of Quantitative Spectroscopy and Radiative Transfer, 106(1-3), 360-377, doi:10.1016/j.jqsrt.2007.01.049, 2007.

Tian, P., Cao, X., Zhang, L., Wang, H., Shi, J., Huang, Z., Zhou, T., and Liu, H.: Observation and simulation study of atmospheric aerosol nonsphericity over the Loess Plateau in northwest China, Atmospheric Environment, 117, 212-219, http://dx.doi.org/10.1016/j.atmosenv.2015.07.020, 2019.

Titos, G., Ealo, M., Román, R., Cazorla, A., Sola, Y., Dubovik, O., Alastuey, A., and Pandolfi, M.: Retrieval of aerosol properties from ceilometer and photometer measurements: long-term evaluation with in situ data and statistical analysis at Montsec (southern Pyrenees), Atmospheric Measurement Techniques, 12(6): 3255-3267, https://doi.org/10.5194/amt-12-3255-2019, 2019.

Volten, H., Munoz, O., Rol, E., Haan, J. d., Vassen, W., Hovenier, J., Muinonen, K., and Nousiainen, T.: Scattering matrices of mineral aerosol particles at 441.6 nm and 632.8 nm, Journal of Geophysical Research: Atmospheres, 106(D15), 17375-17401, https://doi.org/10.1029/2001JD900068, 2001.

Zubko, E., Muinonen, K., Muñoz, O., Nousiainen, T., Shkuratov, Y., Sun, W., and Videen, G.: Light scattering by feldspar particles: comparison of model agglomerate debris particles with laboratory samples, Journal of Quantitative Spectroscopy and Radiative Transfer, 131, 175-187, https://doi.org/10.1016/j.jqsrt.2013.01.017, 2013.

Zubko, E., Muinonen, K., Shkuratov, Y., Videen, G., and Nousiainen, T.: Scattering of light by roughened Gaussian random particles, Journal of Quantitative Spectroscopy and Radiative Transfer, 106(1-3), 604-615, doi:10.1016/j.jqsrt.2007.01.050, 2007.