

Response to Referee #2

We greatly appreciate the Referee #2's insightful and constructive comments and suggestions, which are helpful and valuable for greatly improving our manuscript. We have addressed all of the comments carefully as detailed below in our point-by-point responses. Our responses start with "R:".

General comments:

The paper presents measurements, results and analyses of optical properties and size distributions of surface layer aerosols in Northwestern China. Mineral dust affects air quality and climate over very large areas and they can be observed very far from their sources. In observations far from the sources the aerosol is typically aged and mixed with other particles. Therefore, it is very valuable that measurements are conducted also near the sources. This manuscript presents measurements very close to or essentially at the source and is valuable as such. One of the weaknesses of the work is that particle size range of the optical measurements was limited to 2.5 μm . In dust storms there are often larger particles like the authors' own APS measurements show. But now the data are here and also they yield good information. The authors could use the full extent of the data to obtain also more information as I will suggest below.

I can recommend publishing the paper in ACP, but I did find something to be revised.

R: Thanks very much for your good suggestions and the acceptance of this work, we have addressed all of the comments carefully as detailed below.

Detailed comments:

There is no text on the calibration of any of the instruments. In dusty conditions such as the sites where the measurements were conducted, instruments get quickly dirty and

calibrations change. How did you deal with this? Write about calibrations, flow checks etc.

R: We have added the details of the calibration and flow checks for all the aerosol-related instruments in Section 2.2, and the accuracy for each instrument is also listed in Table 1.

In addition to calibrations, also data processing needs some revision.

The nephelometer suffers from a problem called truncation which leads to underestimation of scattering. The error is the larger the particles are. Read and cite Anderson and Ogren: Aerosol Sci. Tech., 29, 57–69, 1998 and Müller et al.: Aerosol Sci. Tech., 43, 581–586, 2009. and use their algorithms to correct the scattering. The corrected scattering coefficients will be larger than the ones presented now. And so will the corrected single scattering albedos also be.

R: We are sorry for the misleading. In this study, the datasets of the aerosol optical properties have already been corrected based on the nonideal detection developed by Anderson and Ogren (1998), and one of the sentence has been added in Section 2.2 as “For reducing and quantifying the uncertainties in aerosol optical properties measured by the nephelometers, the data reduction and uncertainty analysis for the scattering datasets due to nonideal detection are followed by Anderson and Ogren (1998).”

The nephelometer used in the campaign also measures backscatter coefficient. Why is there nothing about that in the whole manuscript? It would be a valuable addition to the paper. If the instrument was working I strongly recommend presenting and discussing also backscatter coefficients and backscatter fractions at 3 wavelengths, both in figures and tables.

R: Due to the backscatter coefficients shows the same trends with the total scattering coefficients but in a relatively small magnitude, we plotted a new figure suggested by the reviewer (Figure 4a). Then, we calculate the backscattering fractions at the wavelengths of 550 nm shown as Figure 4c in the revised manuscript. Additionally, the detailed information of backscatter coefficients and backscattering fractions of $PM_{1.0}$ and $PM_{2.5}$ at the wavelengths of 450, 550, 700 nm are listed in Table 2.

There was the APS. Why was that not utilized more? I have some suggestions, not requirements. First, calculating integrated volume concentrations for $PM_{2.5}$ would yield some quality control when compared with the TEOM. And if they correlate well, they would together yield an estimate of the dust particle density, at least in such cases when particles were dominated by supermicron particles. That would be valuable.

R: Following the reviewer's suggestion, we found that the integrated volume concentrations of $PM_{2.5}$ measured by APS and the mass concentration measured by TEOM are correlated well as Figure S2 shown. Then, we calculated the dust particle density under different atmospheric conditions during the dust field campaign, and the relative discussion were added in the result section in Page 23, Line 2–9, and Table 3.

Second: estimation of scattering coefficient would not be difficult either. If you don't have a Mie code, you can find them in the internet, calculate scattering efficiencies for the size channels of the APS and then calculate scattering coefficient of each size and finally integrate over the size range. An important question would be, for instance, how large a fraction of scattering did you not get measured because of the impactors in front of the nephelometers? Sure, the particles were not spherical and Mie theory not accurate but it would yield an estimate.

R: Thanks very much for your comments and suggestions. We use the Mie theory and the aerosol number size distribution measured by APS to estimate the scattering coefficient compared with that derived by the nephelometer. The real part of the refractive index was assumed to be 1.53, which was widely used for mineral dust in literatures (Müller et al., 2009; McConnell et al., 2010) the imaginary part of the refractive index was determined using Mie calculations. As shown in Figure 14, the Mie-calculated scattering coefficient and measured scattering coefficient are highly correlated. For instance, the imaginary part of the refractive index (0.0010) for natural dust during dust storm in Zhangye and the background weather condition in Dunhuang are similar to the result of SAMUM-1 in Saharan (Müller et al., 2009). Based on the Mie calculation in this study, the PM_{2.5} scattering fraction, which defined as the contribution of the light scattering of PM_{2.5} to the total scattering (the calculated scattering coefficient in the size range of 0.5–20 μm), is ~36.4 % during dust storm, while is in the range of ~37.9–85.1 % during floating dust episode. Detailed information of Mie-calculated and measured scattering coefficient is summarized in Table 3. Generally, most of the $\sigma_{sp, Mie}^{2.5}$ agree well with $\sigma_{sp, neph}^{2.5}$, which can reflect a good quality of the datasets of $\sigma_{sp}^{2.5}$ during this dust field campaign.

Third: the APS data could also be used for calculating some weighted mean diameter, e.g., volume-weighted mean diameter VMD of the size distribution and compare that with the \hat{A}_{sp} . That would be valuable since satellite-derived products use wavelength dependency for estimating size.

R: We use the APS data to calculate the volume-weighted mean diameter (VMD) under

the diameter of 2.5 μm and 1.0 μm . We found that the $\text{VMD}_{2.5}$ and $\dot{A}_{\text{sp}}^{2.5}$ are correlated well during the whole dust field campaign (Figure S3 in Supplement). However, there is no significant linear correlation between $\text{VMD}_{1.0}$ and $\dot{A}_{\text{sp}}^{1.0}$. The highly possible explanation is that the $\text{VMD}_{1.0}$ is calculated based on the aerosol size diameter ranging from ~ 0.5 to 1 μm measured by APS, while the variation of $\dot{A}_{\text{sp}}^{1.0}$ is affected by the aerosol diameter under 1 μm .

There was also an SP2 in the campaign, at least according to Fig 3. Why was it and its data not discussed at all? It would potentially yield also interesting and important results. Comparison with MAAP in different cases for instance. The MAAP measures light absorption which may also be due to absorbing mineral aerosols, not just BC.

R: We feel sorry for the misleading. Yes, we also measure the BC concentration and its size distribution by using the SP2 instrument shown as Figure 3. But the major innovation of this manuscript is the difference of the optical and physical properties of natural and anthropogenic dust. Therefore, the datasets measured by SP2 are used to analyze the mixing status of BC with the other aerosols during this dust field campaign in another manuscript (In preparation). A comparison of the BC mass concentration between SP2 and MAAP instruments is given in Figure S1 in the Supplement. The result indicates that the tendency of BC mass concentrations are much similar, but the values measured by MAAP was relatively larger than that measured by SP2. We note the relative large bias between MAAP and SP2 instruments may result from the size distribution of BC measured by using different sampler inlet impactors of 2.5 μm and 1 μm .

P6, L5 – What is Hexi corridor? Not well-known for non-Chinese.

R: The explanation of the geographical location of Hexi corridor is given in Section 2.1 based on the reviewer's suggestion as follow: "The Hexi Corridor is a ~1000km northwest-southeast-oriented chain of oases in northwestern China (mainly in the Gansu Province), surrounded by the Qilian Mountains (elevation: ~4000m), the Beishan Mountains (elevation: ~2500m), Heli Mountains (elevation: ~2000m) and the Wushao Mountains (elevation: ~3000m). The Hexi Corridor is considered to be a heavily polluted area because of the combination of local topography and the human activities occurring over northwestern China."

P7, L3- Define or explain floating dust.

R: The definition of floating dust has been added in Section 3.1 as "Floating dust is generally defined as a weather phenomenon in which fine mode dust particles suspended in the lower troposphere under calm or low-wind condition, with horizontal visibility less than 10 km."

P8, L3-4, The detection limits of the scattering coefficients were obviously taken from the Table 4 of Anderson et al., 1996 for 300 min averaging time. But in that table there is not the multiplication by 10. So, the detection limit of total scattering at 450 nm is 0.44 Mm^{-1} , not $0.44 \times 10 \text{ Mm}^{-1}$ like the authors claim on L3.

R: We have corrected this sentence as "the detection limits are 0.44 Mm^{-1} , 0.17 Mm^{-1} , and 0.26 Mm^{-1} ($1 \text{ Mm}^{-1} = 10^{-6} \text{ m}^{-1}$), respectively" in Page 8, Line 20–21.

P8, L7: MAAP wavelength: the MAAP manual claims it is 670 nm but Müller et al. Atmos. Meas. Tech., 4, 245–268, 2011 measured it to be 637 nm. You should reprocess

the data. First correct scattering for truncation, then use MAAP data for calculating SSA. But, instead of assuming the wavelength dependence of absorption, use the wavelength dependency (\AA_{sp}) of truncation-corrected scattering and interpolate the scattering to 637 nm and present SSA at 637 nm. This way you avoid assumptions. The point is that the wavelength dependency and Ångström exponent of absorption by absorbing mineral dust may significantly differ from 1.

R: We have adjusted the absorption estimated by MAAP to 637nm following the method of Müller et al. (2011). Then, we interpolate the scattering coefficients to 637 nm in order to calculate SSA at 637 nm. We also replotted all of the related Figures as well as Table 2 based on the corrected datasets in the revised manuscript.

In Figure 1, show

- Hexi corridor – not well-known for most readers of ACP

R: The same with the above explanation.

- show a kilometer scale also in the upper panel

R: We have added the kilometer scale in the upper panel of Figure 1.

- use and show sub-panel letters a – d. Also for the upper panel.

R: We have added the sub-panel letters a–d in Figure 1.

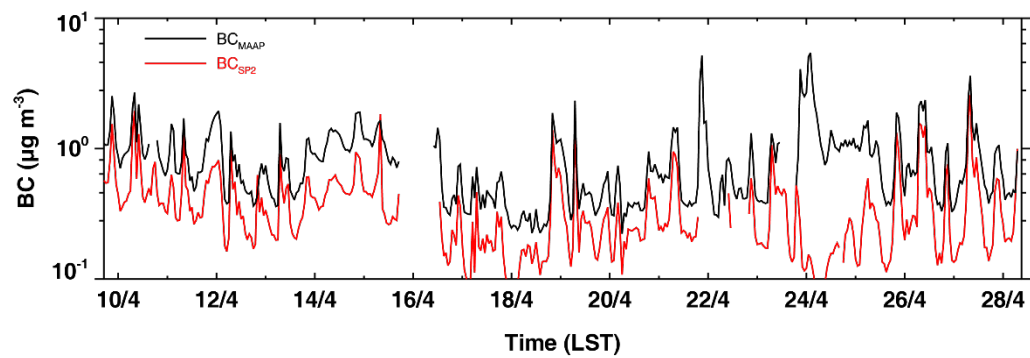


Figure S1. Temporal variations of BC mass concentration under the diameter of 1 μm and 2.5 μm measured by SP2 and MAAP in Zhangye from 9 to 28 April, respectively.

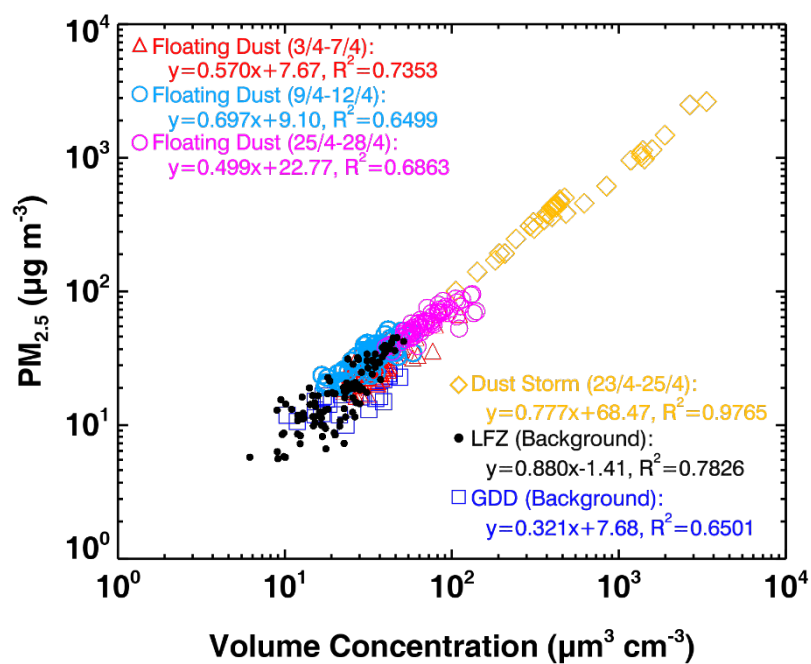


Figure S2. Scatter plot of mass concentration of PM_{2.5} versus the integrated volume concentration under the diameter of 2.5 μm during the dust field campaign. The color symbols represent different atmospheric conditions during the dust field campaign.

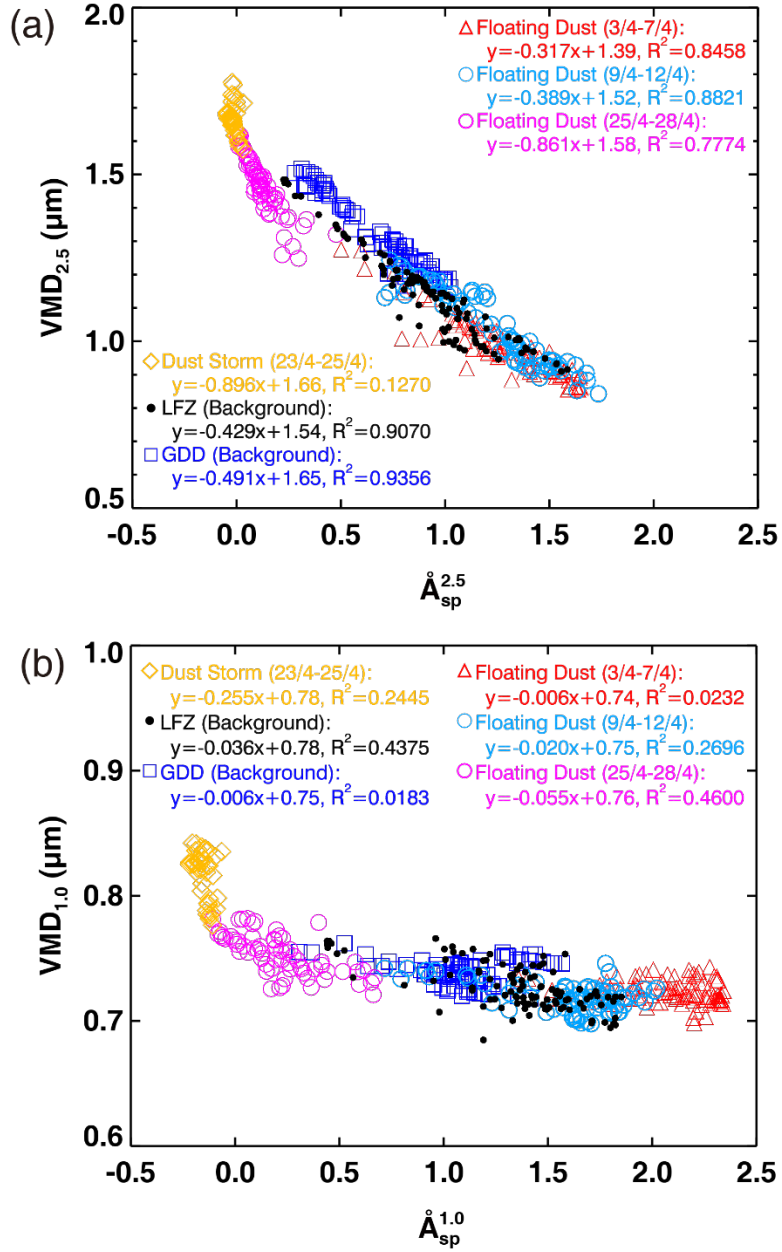


Figure S3. Scatter plot of the volume-weighted mean diameter (VMD) versus scattering Ångström exponent at 450–700 nm for (a) PM_{2.5} and (b) PM_{1.0}. The color symbols represent different atmospheric conditions during the dust field campaign.

Table 3. Statistical summary of optical and microphysical properties of aerosols in different atmospheric conditions during the dust field campaign.

	Floating Dust (3/4–7/4)	Floating Dust (9/4–12/4)	Floating Dust (25/4–28/4)	Dust Storm (23/4–25/4)	LZF (Background)	GDD (Background)
Dust particle density (g cm ⁻³)	1.32 ± 0.28	1.04 ± 0.17	1.24 ± 0.21	1.16 ± 0.51	1.33 ± 0.44	1.64 ± 0.50
Refractive index	1.53–0.0117 <i>i</i>	1.53–0.0862 <i>i</i>	1.53–0.0020 <i>i</i>	1.53–0.0010 <i>i</i>	1.53–0.0442 <i>i</i>	1.53–0.0010 <i>i</i>
$\sigma_{\text{sp, neph}}^{2.5}$ (Mm ⁻¹)	102 ± 37	86 ± 29	127 ± 31	1087 ± 991	49 ± 24	31 ± 9
$\sigma_{\text{sp, Mie}}^{2.5}$ (Mm ⁻¹)	107 ± 40	72 ± 30	132 ± 32	1163 ± 992	41 ± 25	31 ± 9
$\sigma_{\text{sp, Mie}}^{20}$ (Mm ⁻¹)	153 ± 51	102 ± 31	334 ± 75	2983 ± 2712	66 ± 32	71 ± 27
PM _{2.5} scattering fraction (%)	49.3	85.1	37.9	63.6	74.8	43.7

References

- Anderson, T. L. and Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 Integrating Nephelometer, *Aerosol Sci. Tech.*, 29, 57–69, doi:10.1080/02786829808965551, 1998.
- McConnell, C. L., Formenti, P., Highwood, E. J., and Harrison, M. A. J.: Using aircraft measurements to determine the refractive index of Saharan dust during the DODO Experiments, *Atmos. Chem. Phys.*, 10, 3081–3098, doi:10.5194/acp-10-3081-2010, 2010.
- Müller, T., Henzing, J. S., de Leeuw, G., Wiedensohler, A., Alastuey, A., Angelov, H., Bizjak, M., Collaud Coen, M., Engström, J. E., Gruening, C., Hillamo, R., Hoffer, A., Imre, K., Ivanow, P., Jennings, G., Sun, J. Y., Kalivitis, N., Karlsson, H., Komppula, M., Laj, P., Li, S. M., Lunder, C., Marinoni, A., Martins dos Santos, S., Moerman, M., Nowak, A., Ogren, J. A., Petzold, A., Pichon, J. M., Rodriguez, S., Sharma, S., Sheridan, P. J., Teinilä, K., Tuch, T., Viana, M., Virkkula, A., Weingartner, E., Wilhelm, R., and Wang, Y. Q.: Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops, *Atmos. Meas. Tech.*, 4, 245–268, doi:10.5194/amt-4-245-2011, 2011.
- Müller, T., Schladitz, A., Massling, A., Kaaden, N., Kandler, K., and Wiedensohler, A.: Spectral absorption coefficients and imaginary parts of refractive indices of Saharan dust during SAMUM-1, *Tellus B*, 61, 79–95, doi:10.1111/j.1600-0889.2008.00399.x, 2009.