The authors appreciate the reviewer's constructive and friendly comments. We have substantially revised the manuscript. New data and figures are present in the main text. A new supplementary document is included in the revision. We reply to the reviewer's comments point by point.

**Anonymous Referee #2**

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The study of Chang et al. developed the GSI 3Dvar capability to assimilate AOD, scattering/absorbing coefficients for MOSAIC scheme. A few DA tests (both simultaneously and separately experiments) were conducted for northwestern China and compared with surface observations at Kashi. The authors should have spent great efforts on the system development and presented very comprehensive results.

Based on my current understanding, some more work need to be done to facilitate the readers to understand, including some essential considerations of the DA core details and the clarifications of the texts. In this way, the system would be better under- stand/promoted and readers would be more convinced.

My general comments are as below:

1. Actually GOCART is understood for the better performance of dust simulation and the relevant optical properties had been well verified; while the MOSAIC scheme is thought to be more suitable for anthropogenic emission related simulation, but the optical simulation is rather complex.

Response: Agree. The GOCART dust emission scheme is popular for dust simulation. Here, we applied the GOCART dust scheme to simulate the dust and used the MOSAIC scheme to simulate anthropogenic aerosols.

In this study, the system is developed for MOSAIC but the verification is conducted for a site in desert. This required intensive investigation of the DUST related properties representation in the MOSAIC scheme, for example,

(a) the refractive index of OIN since it is mostly treated as DUST (while there should be distinctive differences between the two);

Response: Yes, the OIN is not equivalent to dust. WRF-Chem has a dust option (dust_opt=13) for simultaneous simulation of dust and anthropogenic aerosols with the GOCART dust scheme and the MOSAIC scheme, respectively. With this option, dust is added to OIN. Surely, this simplification is not perfect, but it did not hinder our verification of the DA system. In fact, even using the GOCART aerosol scheme, WRF-Chem computes aerosol optics with the Mie theory. Improving the dust representation in WRF-Chem needs further code development.

(b) the species partitioning (NO3 is not changed in option 2 which might not be reasonable and lead to unbalanced chemistry partitioning),

Response: NO3 is one of the control variables in the revision. We redid the DA experiments.

(c) the size distribution, (d) the number concentration, since the three factors determining the absorbing and scattering efficiency;

Response: We used multi-wavelength aerosol optical measurements to verify the DA system. The revised manuscript additionally shows the angstrom exponent result in Table 3.

(e) aerosol water content which are not considered but actually may change the optical properties. With very limited observational data to verify the above-mentioned information, the results in this study is really hard to interpret.

Response: Aerosol water content (AWC) is not a control variable in DA but is diagnosed in the GSI system according to the hygroscopic growth scheme, based on the analyzed aerosol dry mass concentrations. This treatment ensures the change in AWC is a physical constraint. Besides, AWC is low in the desert site and does not affect AOD a lot. In the revision, we dig the analyses by studying angstrom exponent, SSA, mass extinction coefficient. Hope the revised manuscript is convincible.

2. Some descriptions about DA core and observational data should be provided. For example, it seemed not only AOD, but also wavelength depended absorbing and scattering efficient were all assimilated, the corresponding observational operators and the errors should be given in more detail.

Response: Sorry for the confusion. The observational operators of scattering/absorption coefficient are implicitly involved in the operator of AOD in equation (3). In the revised section 2.2.3, we explicitly present the two observational operators in equation (4). We rewrote the statements about observation errors in the revised section 2.4:

"*The observation errors of PMx are handled in the conventional way (Schwartz et al., 2012; Chen et al., 2019), which contains the measurement error (e1) and the representative error (e2). The measurement error is the sum of a baseline error of 1.5 µg m–3 and 0.75% of the observed PMx concentration. The representative error is the measurement error multiplied by the half-squared ratio of the grid spacing to the scale distance. The scale distance denotes the site representation in GSI and has four default values of 2, 3, 4, and 10 km, corresponding to the urban, unknown, suburban, and rural sites. We used 3 km for the scale distance in this study. As we had a single site in Kashi, it is difficult to estimate the site representation error. Since the DA analysis was based on the child model domain with a horizontal resolution of 5 km, close to the site distance to the Kashi urban area, we assumed the aerosol optical measurement had good representativeness of the model grid covering the site. The observation error of CE318 AOD took the AERONET AOD uncertainty of 0.01 in cloud-free conditions (Holben et al., 1998). The AOD observational error was further divided by the total model layer thickness in GSI.*"

Comments by lines:

1. Line 65 and other places. Adjoint operator, is it referred as TL-AD? Please clarify

Response: We corrected the statements to "tangent linear".

2. Line 86 GIS ?

Response: Corrected to "GSI"

3. Line 100 Zang et al 2016, acutally a different DA system was used other than GSI in this study. Please check.

Response: We corrected the statements in the revised introduction:

*"Li et al. (2013) developed a 3D-Var scheme for assimilating the surface $PM_{2.5}$ and speciated aerosol chemical concentrations for the WRF-Chem MOSACI aerosols. Zang et al. (2016) applied this scheme to incorporate aircraft speciated aerosols in California. They proved that the assimilation of aircraft profile extended the DA benefit to aerosol forecast."*

4. Line 181 regarding of the low anthropogenic and biogenic emissions in the desert, why not use GOCART instead?

Response: The research purpose is to introduce the new GSI system to work with the MOSAIC aerosol scheme. We used the GOCART scheme to simulate dust. In the revised section 2.1:

*"The dust emission was simulated using the GOCART dust scheme (Ginoux et al., 2001), and the dust mass was included in the OIN concentration. We performed the MOSAIC aerosol simulations with four-size bins (0.039–0.156 µm, 0.156–0.625 µm, 0.625–2.500 µm, and 2.5–10.0 µm dry diameters) for the anthropogenic aerosols."*

5. Line 184-190. Actually the optical properties of NH4SO4, OC, dust, NaCl, H2O are treated as wavelength depended in the model, this information should be investigated and provided. As it seemed that multi-wavelength aerosol scattering and absorption coefficients are assimilated. The uncertainties of the assumption in the model and observational data should be provided.

Response: In the revision, we give the complex refractive index in table S1 in the supplementary document; Section 2.2.6 describes the refractive index; The revised section 2.4 describes the observational errors; the revised section 3.3 and section 4.2 states the uncertainties associated with dust morphology and aerosol radiative forcing. Hope the revisions make the manuscript more complete.

6. Line 228. Why NO3 is not considered? In this case, it may lead to unbalanced chemistry partitioning.

Response: Nitrate is a control variable in the revision.

7. Section 2.2.3 It seemed that scattering and absorbing coefficients are also observational assimilated. Please provide details.

Response: The revised section 2.2.3 provides the observation operators of scattering/absorption coefficients.

8. Line 101: are the Mi,z,k in the two terms the same, maybe possibly dry and wet mass concentration respectively? If not, please clarify.

Response: Mi,z,k denotes the aerosol composition.  It could be aerosol water content when calculating the internal mixing refractive index. In the revised section 2.2.4,

*"Note that the dry ($r_{dry,z,k}$) and wet ($r_{wet,z,k}$) particle radiuses are both present in Eq (21). Because aerosol water content is not a control variable, $r_{dry,z,k}$ is used in Eq (19) and appears in Eq (21). Aerosol water*

*content participates the computation of internal mixing refractive indexes, and thus $r_{wet,z,k}$ is also present in Eq (21).*"

9. Line 315: is rwet related with aerosol water content, considering the hygroscopicity? Any uncertainty by not considering aerosol water content. Please clarify.

Response: $r_{wet}$ is the wet particle radius when aerosol water content (AWC) is counted in the aerosol composition. At the end of the revised section 2.2.1:

"*The AWC was diagnosed according to the analyzed aerosol mass concentration and the background relative humidity in each DA outer loop. The hygroscopic growth was calculated using the WRF-Chem code coupled with the revised GSI.*"

10. Line 352, please clarify mizk as dry or wet mass?

Response: Mi,z,k denotes the aerosol compositions. It could be aerosol water content when calculating the internal mixing refractive index.

11. Line 367. Any uncertainty by considering constant radius?

Response: It is hard to estimate the uncertainty of this constant radius in this study. Appling this constant radius is to simplify the mathematical derivation of the tangent linear operator for AOD. This simplification was applied by Saide et al. (2013). We hope to remove this assumption in the future and could discuss the relevant uncertainty.

Saide, P. E., Carmichael, G. R., Liu, Z., Schwartz, C. S., Lin, H. C., da Silva, A. M., and Hyer, E.: Aerosol optical depth assimilation for a size-resolved sectional model: impacts of observationally constrained, multi-wavelength and fine mode retrievals on regional scale analyses and forecasts, Atmos. Chem. Phys., 13, 10425-10444, doi:10.5194/acp-13-10425-2013, 2013.

12. Line 703-706. Please dig more on this issue.

Response: We add a paragraph in the revised section 3.3:

"*The irregular morphology had a significant influence on the dust simulation. Okada et al. (2001) found that the aspect ratio (the ratio of the longest dimension to its orthogonal width) of the mineral dust particles (0.1-6 µm) in China arid regions exhibited a median of 1.4. Dubovik et al. (2006) suggested the aspect ratio of ~1.5 and higher in desert dust plumes. Kok et al. (2017) found that the dust' sphericity assumption underestimated dust extinction efficiency by ~20–60% for the dust particle larger than 1µm. Tian et al. (2020) found that using a dust ellipsoid model could increase the concentration of coarse dust particle (5-10 µm) by ~5% in eastern china and ~10% in the Taklimakan area because of the decrease in gravitational settling, comparing with the simulations with dust sphericity model. Nevertheless, the aspect ratio of the spheroid dust is uncertain. Even after applying the spheroidal approximation, Soorbas et al. (2015) found that the model underestimated 550 nm aerosol scattering and backscattering values by 49% and 11%, respectively, because of the uncertainties in particle axial ratio, complex refractive index, and the particle size distribution. To date, the assumption of spherical particles has been widespread in* *models (including WRF-Chem) for computational efficiency. Impact of dust morphology to DA deserves a further investigation.*"

13. Line 765. Please investigate the uncertainties of the modeled and observed absorption coefficients.
Response: We check the differences in DA analysis as using the different imaginary part of dust refractive index and background error of BC. Please refer to the revised section 3.5.

14. Figure2. Why the domain averaged standard deviation (c) is significantly larger than that of column averages (d, e)?
Response: The vertical profiles in figure 2(c, d, e) are based on different grids. As shown in Figure 5c, Kashi and the desert point we picked up for figure 2(e) are not on the track of dust storm. Thus, the dust variations at the two points (figure 2d, e) are smaller than the average of binning standard deviation (figure 2c).

15. Figure 3. Why background error standard deviation of the OIN is two magnitudes larger than the other species? Indicating dominating contribution of dust? In this case, is it meaningful to investigate other species changes?
Response: We accidently lowered anthropogenic aerosols in Kashi. The revised simulations correct the emissions and show that the OIN is still the predominant composition, accounting for 62% of $PM_{2.5}$ and 82% of $PM_{10}$ in April. The qualitative conclusion is the same.

16. Table 1. Please explain how the errors are determined?
Response: In the revised section 2.4:

"*The measurement error is the sum of a baseline error of 1.5 µg m*$^{-3}$ *and 0.75% of the observed PMx concentration. The representative error is the measurement error multiplied by the half-squared ratio of the grid spacing to the scale distance. The scale distance denotes the site representation in GSI and has four default values of 2, 3, 4, and 10 km, corresponding to the urban, unknown, suburban, and rural sites. We used 3 km for the scale distance in this study. As we had a single site in Kashi, it is difficult to estimate the site representation error. Since the DA analysis was based on the child model domain with a horizontal resolution of 5 km, close to the site distance to the Kashi urban area, we assumed the aerosol optical measurement had good representativeness of the model grid covering the site. The observation error of CE318 AOD took the AERONET AOD uncertainty of 0.01 in cloud-free conditions (Holben et al., 1998). The AOD observational error was further divided by the total model layer thickness in GSI.*"