[Reviewer 1]

Review comments on "Inferring iron oxides species content in atmospheric mineral dust from DSCOVR EPIC observations" by Go et al. submitted to ACP

We appreciate the referee's thoughtful reading, valuable suggestions and time that we hope helped us to improve the manuscript. Our point-to-point replies are presented below.

&&&GENERAL COMMENTS&&&

The iron oxides contents of goethite and hematite in mineral dust play a key role to quantify the dust light absorption, and then influence its radiative effect. Even though the different spectral behavior of refractive indices, the direct retrieval of goethite and hematite concentration from remote sensing measurements is difficult due to the limited information content. This paper, based on the existing EPIC MAIAC product of aerosol type and spectral imaginary refractive indices, proposed a method to infer columnar goethite and hematite mass/volume concentrations by fitting the EPIC MAIAC spectral (UV-Vis) imaginary refractive indices assuming the Maxwell-Garnett effective medium mixture of non-absorbing host and absorbing hematite and goethite. The results are evaluated with in situ measurements. Overall, this study is well-written and within the scope of ACP. I would recommend this paper to be published in ACP after some comments and concerns being addressed.

 Since it's a sequential approach relying on the product of EPIC MAIAC spectral (UV-Vis) imaginary refractive indices (k), the quantitative validation of spectral k product is critical while not included in this study. The SSA validation by Lyapustin et al. (2021, FRS) may imply the quality of k. However, I would suggest to perform the validation of EPIC MAIAC k product directly and to quantify the uncertainty of derived hematite and goethite concentrations due to input k uncertainty.

Response: We agree that validation of the spectral imaginary refractive index is important. However, any quantitative comparison of $k(\lambda)$ with AERONET, as mentioned in Lyapustin et al. (2021), is associated with high uncertainty. First, AERONET states a 30-50% accuracy for the imaginary refractive index at AOD440 nm > 0.4 (Dubovik et al., 2000), with uncertainties being higher for the coarse mode dust as well as for optically thin aerosols (lower AOD). Second, the main sensitivity of MAIAC for SAE (parameter b) comes from the 340-443 nm range, whereas AERONET provides spectral dependence of refractive index for the non-overlapping range of wavelengths 440-1020 nm. For these reasons, Lyapustin et al., (2021, Lines 156-161) provided a qualitative evaluation of spectral imaginary refractive index $k(\lambda)$.

Nevertheless, to address the Reviewer's question, we conducted a direct comparison of parameters k_0 and b with AERONET. The AERONET "parameter b" was derived from $k(\lambda)$ values at 440nm and 680nm. The results are shown in the figure below for the (R1) northern Africa, (R2) the Sahel, (R3) East Africa and the Middle East, (R4) central Asia, (R5) East Asia. In case of k_0 , 95.7% (R1), 100% (R2), 100% (R3), 43.8% (R4), 73.3% (R5) of the MAIAC EPIC k_0 -values are within the expected error (±0.003). Regarding b values, 32.9% (R1), 25% (R2), 0% (R3), 61% (R4), 63.3% (R5) of the MAIAC EPIC b-values are within the expected error (±0.5). A pixel-level assessment of the uncertainty for the derived hematite and goethite concentrations is currently under development for the next version of the MAIAC EPIC algorithm.



Fig. 1. Figure R1. Validation results of AOD at 443 nm, SSA at 443 nm, k at 680 nm, b values of MAIAC EPIC with AERONET data for the (first row) total, (R1; second row) northern Africa, (R2; third row) the Sahel, (R3; fourth row) East Africa and the Middle East, (R4; fifth row) central Asia, (R5; sixth row) East Asia. Spatial and temporal collocation criteria within 30 km and 3h are used.

- 2. In my view, EPIC MAIAC dust detection is another key information used to select pixels and perform the hematite and goethite inversion. The authors claimed that some dust + mixed/smoke aerosol cases, which are classified into dust, may affect the retrievals for some specific regions. In this sense, I would encourage to include the maps of AE in the analysis which may provide some insights for dust detection.
 - <u>Response:</u> Thank you for the comments. Because EPIC is a single-view instrument with insufficient information content, MAIAC does not retrieve the Ångström Exponent (AE) over land. The dust retrievals are performed for (AOD, k₀, b) assuming real refractive index, spheroidal model and fixed size distribution.
- 3. How do you evaluate the derived columnar goethite and hematite mass concentration with *in-situ* near-surface measurements? How do you assume the vertical distribution? Please clarify in the text.
 - <u>Response:</u> We assume the vertical distribution with a 2km box shaped profile in MAIAC lookup tables. For instance, the boundary-layer aerosol is assumed uniformly distributed in 0-2km layer for 1km effective height. In case of 4km, the 90% of aerosol is assumed at 3-5km, with 10% below. We added the following sentence (line 146): "MAIAC retrievals are reported for the effective aerosol heights of 1 km and 4 km with 2km box-shaped profile, representing the typical boundary layer and free troposphere transported aerosol.".
 Since the retrieval results are columnar goethite and hematite mass concentration, we did not compare the goethite and hematite mass concentrations directly with the in-situ near-

not compare the goethite and hematite mass concentrations directly with the in-situ nearsurface measurements. Instead, iron oxides mass fraction to the column-integrated total mass concentration was compared with in-situ measurements as in Fig 8. Di Biagio et al. (2019). Di Biagio et al. (2019) analyzed aerosols generated from natural soil samples.

- 4. The quality/clarity of the figures (e.g., Figures 1, 5, A1, S1) can be improved.
 - <u>Response:</u> Corrected. Regarding Figure 5, A1, the resolution of the RGB image cannot be improved.

&&&SPECIFIC COMMENTS&&&

- Figure 1: the quality/clarity of the figure should be improved. <u>Response:</u> Corrected. Regarding Figure 5, A1, the resolution of the RGB cannot be improved.
- Line 107: a recent study by Wang et al. (2021, 10.1016/j.atmosenv.2020.117959) developed an algorithm to derive aerosol components from effective density and spectral refractive indices.

Response: Thank you for the information. The reference is added (line 110).

• Section 2.1: You mentioned the validation results of MAIAC EPIC SSA. Have you evaluated the MAIAC EPIC imaginary part of refractive index (k), since the fitting of MAIAC EPIC k is then used to derive hematite and goethite?

Response: This question was addressed above (see General comments, #1).

• Line 180: the link does not work, please check.

<u>Response:</u> Yes, we are aware of that, but this is the exact link that Scanza et al. (2015) provided.

• Line 215: why don't you fit real part together with imaginary part?

<u>Response:</u> MAIAC EPIC assumes the real part of the refractive index in the (AOD, k_0 , b) retrievals, it does not retrieve n_{rtr} . However, as mentioned in line 214-216, the imaginary refractive index of mixture (k_{mix}) is a function of both real and imaginary refractive index of the inclusions 1, 2, and host. Therefore, realistic values of the real refractive indices of inclusions 1, 2, and the host are still required along with the imaginary part. This assumption will be included as part of our uncertainty analysis in the future.

• Line 227: what do you mean flexible retrievals?

<u>Response</u>: Flexible retrieval means the retrieval algorithm of {AOD, k_0 , b}. To improve clarity of presentation, we deleted the word "flexible" and added additional reference Lyapustin et al. (2021).

• Equations 11-13: So, you use the coarse mode volume concentration to approximate the total, right? Then the C_{v, hema} = C_{vc} x f_{hema} is named as the hematite total volume concentration. Is the coarse mode hematite volume concentration more precisely?

<u>Response:</u> That's true. We changed the "total" to the "coarse mode".

• Line 283: Could you explain why the algorithm does not retrieve dust aerosol over South America and southern Africa?

<u>Response:</u> With EPIC lacking bands beyond 780nm, MAIAC cannot differentiate between the smoke and dust despite it detects absorbing aerosols. As smoke is much more ubiquitous aerosol type, MAIAC makes dust retrievals only over pre-defined dust regions. At present, South America and southern Africa are not designated as dust regions in MAIAC EPIC.

• Figure 5: There are more cloud-free pixels in the RGB images than that in the AOD, SSA, Hema and Goet plots. Could you explain how these pixels are selected?

<u>Response:</u> For reliable retrievals, we only select pixels with AOD>0.6 (Ln. 230). At lower AOD, the retrieval error grows rapidly.

• Line 449: How do you convert hematite and goethite volume / mass concentrations to the ironoxide mass fraction? sum of them? It seems missed in the methodology.

<u>Response</u>: We calculated $C_{M,hema}$, $C_{M,goet}$, $C_{M,host}$ values through equation (14). Then the ironoxide mass fraction can be calculated with $(C_{M,hema} + C_{M,goet}) * 100 / (C_{M,hema} + C_{M,goet} + C_{M,host})$. I added the following sentences at line 464.

"Iron-oxide mass fraction is calculated as (C_{M,hema} + C_{M,goet}) × 100 / (C_{M,hema} + C_{M,goet} + C_{M,host})."

• Line 469: with a size range of 2-6 days? Please check.

<u>Response</u>: The sentence is reworded to "a time span typical of 2 to 6 days of transport" at line 475.

• Line 486: The colocation method should include also in the main text. What's the spatial resolution of the product? Why the spatial window is different over Australia (+/-3 degree)?

<u>Response:</u> The spatial resolution of the MAIAC EPIC product is 10 km (~0.1 degree) (Ln. 145). To build the monthly variation plot for hematite and goethite (Figure 8), we used the spatial window (\pm 1°) (Ln. 492). Over Australia, the window was expanded to \pm 3° in order to accumulate a sufficient number of retrievals. We added the following sentence to describe this (Ln. 477-478):

"Each site represents the area of \pm 1° of MAIAC EPIC data except for Australia where the box size was expanded to \pm 3° to accumulate enough retrievals."

• Line 586: Why do you use the threshold AOD>1.0? Is it for dust detection?

<u>Response:</u> Satellite-data pixels with $AOD_{443} > 1.0$ were used in the average, as some pixels of goethite retrieval with $AOD_{443} < 1.0$ display "noise" or "blob" patterns (for example, see third and fourth rows, Fig. A1). This empirical threshold typically guarantees lack of noise. To explain this, we added the following sentence (Ln. 595):

"As in Fig. 8, pixels of AOD > 1.0 were used to compute the average, as some pixels of goethite retrieval with AOD < 1.0 display "noise" or "blob" patterns."

- Line 589: Since it may contain some mixed / smoke aerosol cases, I would suggest to include maps of AE in Fig S2-S5 that may provide some insights for aerosol types.
 <u>Response:</u> Please, see our response to question 2 of General Comments.
- Line 634: real refractive indices -> real part of refractive indices

Response: Corrected.

• Line 708: the reliable product of spectral imaginary part of refractive indices is a precondition. <u>Response:</u> Please, see our response to question 1 of General Comments.

[References]

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmos. Chem. Phys., 19, 15503–15531, https://doi.org/10.5194/acp-19-15503-2019, 2019.

Dubovik, O., Smirnov, A., Holben, B. N., King, M. D., Kaufman, Y. J., Eck, T. F., & Slutsker, I. (2000). Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements. *Journal of Geophysical Research: Atmospheres*, *105*(D8), 9791-9806.

Lyapustin, A., Go, S., Korkin, S., Wang, Y., Torres, O., Jethva, H., & Marshak, A. (2021). Retrievals of Aerosol Optical Depth and Spectral Absorption from DSCOVR EPIC. *Frontiers in Remote Sensing*, *2*, 7.

Scanza, R. A., Mahowald, N., Ghan, S., Zender, C. S., Kok, J. F., Liu, X., ... & Albani, S. (2015). Modeling dust as component minerals in the Community Atmosphere Model: development of framework and impact on radiative forcing. *Atmospheric Chemistry and Physics*, *15*(1), 537-561.