

[Reviewer 3]

This work presents the application and discuss the results of a new algorithm that applies to MAIAC EPIC data to retrieve the mass concentration of hematite and goethite content in the global atmosphere over land. The aim of the study is of relevance for dust investigations, from the earth's radiative budget analyses to the biogeochemical studies. The retrieval scheme and the Maxwell Garnett approximation applied seem reasonable. Hypothesis and steps of the retrieval are clearly presented. Data from laboratory and field observations are used to compare and validate the results. Regional and seasonal variability of hematite and goethite mass concentration retrievals are analyzed. The paper is well written and properly organized.

I am in favour of the publication of the paper after the authors have addressed some points below.

- The validation of k from MAIAC, which is the starting point of the analysis, is actually missing. Can you compare the k values against lab and field observations as you did for the retrieved hematite and goethite mass concentrations?

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.

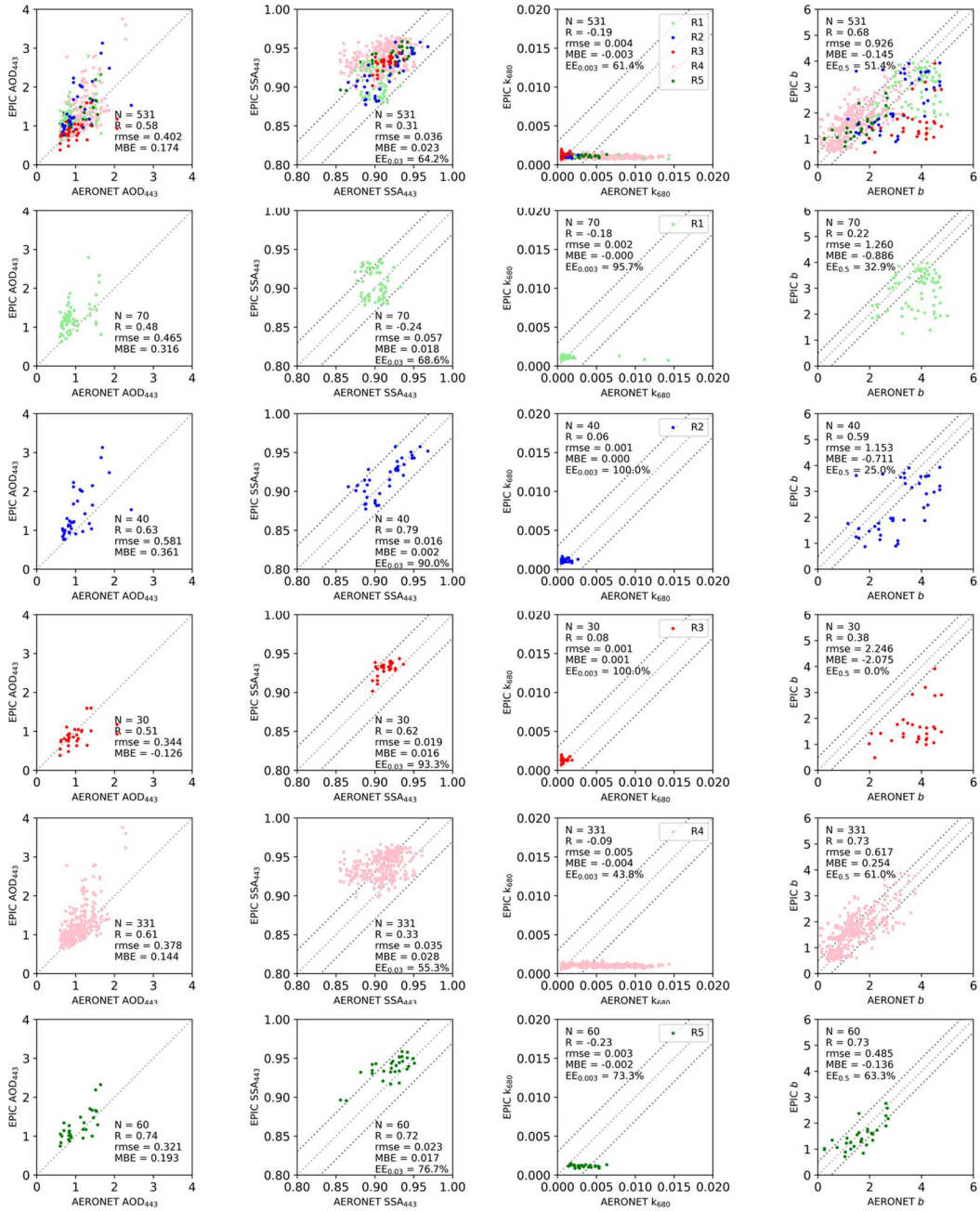


Fig. 1. Figure R1. Validation results of AOD at 443 nm, SSA at 443 nm, k at 680 nm, b values of MAIAC EPIC with AERONET data for the (first row) total, (R1; second row) northern Africa, (R2; third row) the Sahel, (R3; fourth row) East Africa and the Middle East, (R4; fifth row) central Asia, (R5; sixth row) East Asia. Spatial and temporal collocation criteria within 20 km and 3h are used.

- To my understanding the algorithm applies only on cases with AOD larger than 0.6, when MAIAC EPIC provides outputs of k, AOD and b. This is one point to discuss when analyzing the regional/seasonal concentrations of hematite and goethite, perhaps. What is the impact of this assumption on the retrieval and its exploitation/application? Is it a real "global" climatology that is obtained?

Response: Thank you for this comment, it is indeed not sufficiently explained in the text. Yes, currently the algorithm only applies the retrieval for pixels of AOD>0.6. And, for the global climatology (Section 3.3), pixels of AOD>1.0 were used to compute the average due to the low sensitivity when pixels of AOD<1.0. This could create some differences due to the omission of the fine mode dust, which contains hematite and goethite in the fine mode (clay) according to some publications (e.g., Journet et al., 2014; Menut et al., 2020).

Below is the figure from Journet et al. (2014) that illustrates the (b) hematite in the clay (diameters $D_p=0-2 \mu\text{m}$) fraction, (c) goethite in the clay fraction, and (d) goethite in the silt (diameters $D_p=2-50 \mu\text{m}$) fraction.

The hematite content in the clay fraction is usually below 1.5% but reaches 5% in some regions, including the longitudinal band from Montana to Texas in the US, a latitudinal band across southern Russia, and arid regions of northern Africa, while soils in southern Brazil/northern Argentina have a high hematite content exceeding 5%. Goethite occurs in both the clay- and silt-sized fractions. The amount of goethite in the clay fraction is generally higher than the amount of hematite, and more variable (from 0 to 15%). Goethite is generally more abundant in humid tropical environments while hematite becomes more abundant in the seasonally dry tropics.

I added the following sentences at Line 622-628.

“The seasonal average EPIC data is based on the AOD larger than 1.0, and this may cause the omission of fine mode dust such as clay fraction of hematite (Journet et al., 2014; Menut et al., 2020). Journet et al. (2014) mentioned that the hematite content in the clay fraction is usually below 1.5% but reaches 5% in some regions, including the longitudinal band from Montana to Texas in the US, a latitudinal band across southern Russia, and arid regions of northern Africa, while soils in southern Brazil/northern Argentina have a high hematite content exceeding 5%. Error or uncertainty associated with the omission of the fine mode dust is beyond the scope of this study, and will be provided in the next version of the MAIAC EPIC algorithm.”

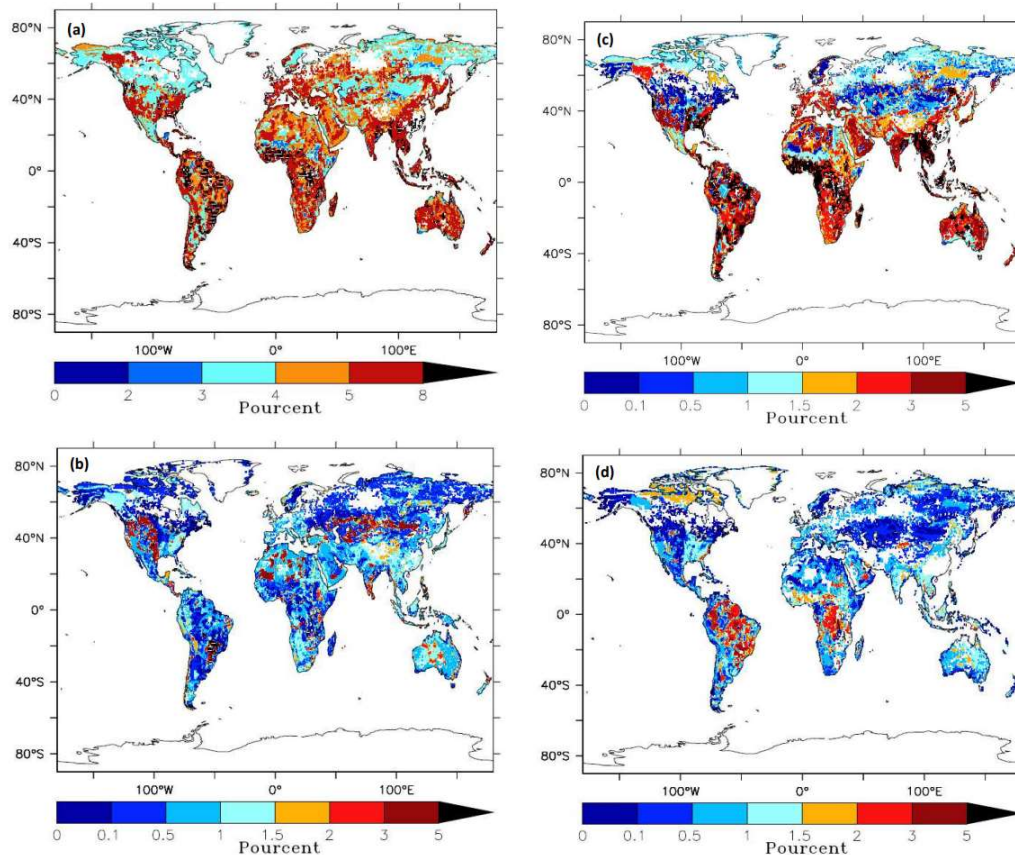


Fig. 7. Iron and iron oxides in soils: (a) iron in the clay fraction (CASE 1), (b) hematite in the clay fraction (CASE 1), (c) goethite in the clay fraction (CASE 1) and (d) goethite in the silt fraction.

Fig. 2. Figure R2. Iron and iron oxides in soils: (a) iron in the clay fraction (CASE 1), (b) hematite in the clay fraction (CASE 1), (c) goethite in the clay fraction (CASE 1) and (d) goethite in the silt fraction (Journet et al., 2014).

- This retrieval applies to the AOD for the coarse fraction, therefore hematite and goethite are referring to the coarse dust AOD only. Is this assumption biasing the retrieval in such a way? Considerations on the size-dependent composition of dust and distribution of Hematite and Goethite as a function of size should be added.

Response: Thank you for the valuable comments. The size distribution we assumed in the retrievals will always be an issue unless we have more information contents (e.g., multi-angle measurements, polarization). There is always a small fraction of the fine mode, but it is irrelevant for the dust storms at high AOD. The dust size distribution model we are using is from Oleg Dubovik's 2003 JAS paper (based on AERONET data at Solar Village). Based on AOD validation (in MAIAC MODIS and EPIC), it works well. The model has a bi-lognormal size distribution with $Rv_f=0.12$, $Sigv_f=0.5$, $Rv_c=1.9$, $Sigv_c=0.6$ for the fine and coarse modes. The volumetric concentration for the coarse mode increases with AOD, and is constant for the fine mode: $Cv_f=0.02$, and $Cv_c=0.02+0.9*AOD$. Even if we admit more fine mode in the retrievals,

the absorption in the Blue-UV range, and its spectral dependence, is still defined by the dominant coarse mode at the range of AOD>0.6 we are working with. In summary, we do not believe we have a sensitivity to the fine mode with EPIC in case of dust, at least within the size distribution model we are using presently.

- Figure 3 can probably show goethite dataset for real and imaginary refractive indices as well, similar for Table 1, the corresponding information for goethite is missing

Response: Thank you for the comments. Unlike hematite, information on the complex refractive index of goethite is very scarce. We only found two reports of complex refractive indices of goethite having been published by Bedidi and Cervelle (1993) and Glotch and Roman (2009) for 0.45-0.75 and 8–50 μm wavelength ranges, respectively. Glotch and Roman (2009) wavelength ranges 8-50 μm is out of interest. The refractive index of Bedidi and Cervelle (1993) that we use is described in Figure 1.

- Line 284: what do you mean with « significant » events? in AOD or other?

Response: Yes, this is significant dust AOD event. Changed to “significant” → “significant AOD”.

- Line 291: Di Biagio et al. (2019) analyzed aerosols generated from natural soil samples and not soil properties, check elsewhere in the paper (in example sect 3.2 and following text) and clarify this aspect which is of relevance for the validation of the retrieved atmospheric aerosol hem and goet concentration from the present study.

Response: Thank you for the comments. I added “Di Biagio et al. (2019) analyzed aerosols generated from natural soil samples” (line 472). The following lines were also corrected accordingly: 127, 219, 294, 473, 475, 512, 527, 562, 567, 587, 717.

- What is the impact of fixing the real refractive index on the retrieval?

➔ Response: The figure below shows the refractive index of mixture (based on the Maxwell-Garnett internal mixing rule) with respect to the iron oxide fraction – in a way it shows the sensitivity indirectly. The real part of the mixture is expected to vary quite a bit depending on the iron oxide fraction. The current MAIAC algorithm does not retrieve real part of the refractive index due to the lack of information contents of the single view satellite, and the related sensitivity study regarding the real part of the refractive index is the beyond scope of this study. We will evaluate the impact of this assumption on uncertainty in a future study.

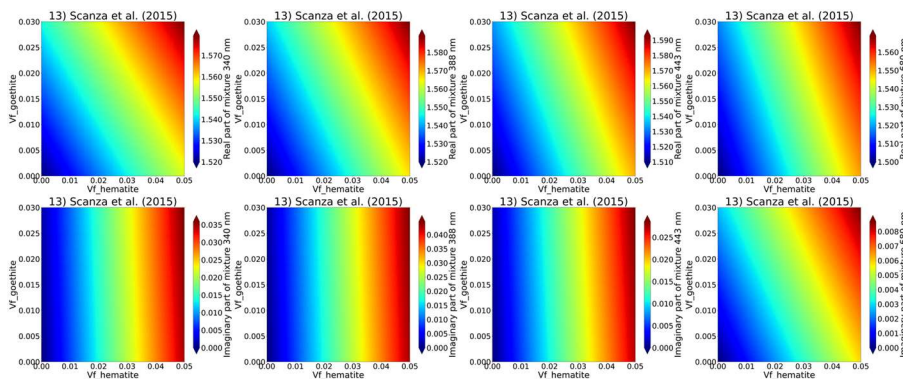


Fig. 3. Figure R3. Theoretical retrieval test. The real part (top row) and imaginary part (bottom row) of refractive index of mixture at 340, 388, 443, 680 nm as a function of the hematite volume fraction (x axis) and goethite volume fraction (y axis) of MAIAC EPIC parameter.

- I would discuss possible perspective applications over ocean, an aspect which could be of great interest for biogeochemical studies

[Response:](#) Thank you for the suggestion. We added the following sentence in the conclusion section:

[Line 727:](#) “In near future, the algorithm will be expanded over the global ocean to support ocean biogeochemical studies (e.g., Tagliabue et al., 2017).”

- Please, clearly state in the abstract the conditions to which this retrieval applies (land, AOD>0.6, ..)

[Response:](#) Corrected.

[References]

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