

Interactive comment on “Retrieval of Desert Dust and Carbonaceous Aerosol Emissions over Africa from POLDER/PARASOL Products Generated by GRASP Algorithm” by Cheng Chen et al.

Anonymous Referee #2

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The manuscript by Cheng et al. presents a top-down emission estimates of desert dust, black carbon and organic carbon aerosols over Africa and the Arabian Peninsula. The authors describe the inversion methodology, perform sensitivity tests and apply their tool to the fitting of one year of AOD and AAOD GRASP retrievals from PARASOL measurements. The authors show an improvement in the simulated AOD and AAOD when the outputs of two models are compared against satellite and ground-based observations.

General comments:

The manuscript presents interesting and new results that contribute to the research

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field. The sensitivity tests are appropriate, the results presented are very good and the implementation of the posterior emissions in a different model confirms the quality of the results. More importantly, this work shows that it is possible to compute top-down estimates of emissions with a relatively high spatial and temporal resolution. The manuscript shows a detailed and complete work, but the quality of the figures and the general presentation of the text overshadow the work.

Despite the fact that English is not my mother tongue, I have found a large number of grammatical errors. Please correct carefully the English language to accomplish with the journal standards and to improve the readability of the manuscript. I have pointed out some of these errors in the minor changes section (only for the first pages of the manuscript).

In general, the figures are low quality for publication. Labels are small and captions are incomplete as they do not explain well the elements in the figures. In most of the maps there is no latitude/longitude labels, etc. Please follow the guidelines from https://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html

The methodology section (3.1) is not clear. The notation is not consistent throughout the text and there are elements in the equations that are not explained. I would suggest to avoid details about the minimization procedure (they are already explained in Dubovik et al 2008) and to focus on the improvements and differences with the Dubovik et al 2008 work.

Basic information about the assimilation is missing in the manuscript. The definitions and values of the error covariance matrices and the regularization parameter are nowhere stated. There is a whole subsection about the weights of the observational error covariance matrix without knowing the error covariance matrix. How do the authors account for the model error in their assimilation? How valid is the diagonal assumption of the observational error covariance matrix, knowing that the assimilated AOD and AAOD are issued from the same algorithm and measurements? Do the GRASP

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algorithm reports uncertainty of AOD and AAOD? Are these uncertainties taken into account in this work? Which are the uncertainties of the emissions factors? Are they the same for the different aerosol types, locations and seasons?

Reporting uncertainties of the posterior emissions would be an additional major contribution of this work, but in practice it is not always possible to compute them confidently. Would be possible to provide these values?

Specific comments:

Title

Following the ACP guidelines, the title should not be capitalised in every word.

Short summary

I think that the qualifier “important” in “This study is an important contribution to” is out of place (and sounds like a subjective and personal appreciation that do not add information to the summary).

P1 L14-17

Sentence too long. I suggest to split the sentence after “(OC) emissions” and continue with “AOD and AAOD from ... has been assimilated .. ”.

P1 L18

These tests show..

P1 L19

Remove “For example”

P1 L21

Please rephrase “...an additional about 1.8 times differences...”

P1 L24-25

Remove “GEOS-Chem inventory of”. The inventory is not from GEOS-Chem, it is from Bond et al. and GFED.

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P1 L29

Please change "independent of and more"

P1 L31 – P2 L1

I am not sure if it is appropriate to list all the statistics (as acronyms) of the comparison in the abstract.

P2 L6-8

Despite it is cited in the next sentence, this sentence would improve with a reference.

P2 L14

".. the role that atmospheric.. "

P2 L15

Please clarify that it is about short-wave radiation. The measurements are not only due to aerosols, the interaction of light with molecules, surface and clouds is also important.

P2 L24

"aerosols"

P2 L24

To my knowledge, sensors provide measurements, algorithms can provide retrievals.

P2 L27-28

Reference needed. All the cited satellite retrievals of aerosols are based on visible, UV and near infrared measurements, so this sentence do not add new information. I would say that, given the current state of the instruments, algorithms and knowledge of the system, UV and polarimetric measurements are needed to better retrieve absorption properties of aerosols in the visible.

P2 L29

"high degree" sounds odd.

P2 L30

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Please change “answering question regarding” for something more specific.

P2 L30

“fates”?

P2 L31

I think that “incorporate” is not the best word

P3 L3

I would add aerosol processes... “of atmospheric and aerosol processes..”

P3 L4

Please change “are found to”

P3 L12

“i.e. fitting satellite observations and model estimates and by adjusting aerosol emissions”. This sentence is not clear and it is inaccurate. Inverse modelling is also applied in mathematics, geophysics, etc (there are a lot of examples in the Tarantola (2005) book). Please be more concise in the definition of inverse modelling.

P3 L13

Please avoid these kind of statements. The data is not inverted. Is the CTM (and the emission model/inventory) what is usually called “inverted”.

P3 L14

Please specify which kind of distributions: spatial? temporal? size distributions?

P3 L17

The emissions are not from MODIS AOD.

P3 L17-18

Please remove “works such as”

P3 L3-26

This long sentence could be written better.

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P3 L26-27

Is not POLDER the name of the instrument? (and PARASOL the satellite/mission)?

P3 L30

"decreases sharply.." This is true with respect to the wavelength. Wavelength is not the only way to account for the spectrum.

P3 L30-31

Please change "most strongly". "ubiquitously" sounds odd in this context.

P3 L31-32

Please decide : "shortwave", "short-wave" or "short wave". It could be possible to remove "shortwave" from here, as it is followed by "visible".

P3 L34

I do not understand the point of this sentence. MODIS and MISR also provide AOD over bright surfaces. May be it is better to show the accuracy of GRASP retrievals over the desert.

P4 L3-6

Should not this sentence go in the model description section?

P4 Section 2.1:

Please check the grammar of this section.

P4 L15-16

The variability has drawn the research? Please rephrase.

P4 L16-20

Sentence seems too long. I would prefer: Figure 1 shows the number of ..., the 28 AERONET sites,.. etc.

P4 L23

"Northern Africa Sahara and Arabian Peninsula desert region"?

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P5 L7

Please remove or explain the word "forward".

P5 L7

Please correct "with 47 layers vertical resolution"

P5 L10

"consist of" sounds odd.

P5 L11

Please correct the grammar.

P6 L2-6

Remove parenthesis in the citations of lines 2 and 3; check the grammar of the paragraph ("turbulent mixing of particles to the surface"? and all the line 5)

P6 L7

Check "for all"

P6 L8

".. width for ..".

Where are these parameters specified? Please add reference to the table if needed.

P6 L10 and L20

Hygroscopic, not hydroscopic

P6 L14

Is it really necessary to write "aerosol particle"? Only with "aerosol" (or particle) should be enough.

P6 L22, figure 2a

Why do the authors show Q_{ext}/r and not Q_{ext} ?

P7 L3

This is a personal blog. Please consider uploading the data as supplementary material.

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P7 L10-13

The authors should clarify what “to model fully adequate” means.

Table 1.

Would be possible to add the parameters of the rest of the aerosols simulated? (SS and SU)

P8 L5

I suggest to change “is an efficient tool” to “can be used as a tool”

P8 L20-21

Please rephrase and avoid qualifiers as “highly versatile and accurate” without referring to appropriate validations.

P9 L7-8

This sentence is not clear enough and it should be removed.

P10 L4

Please specify which version of AERONET products. (2, 3?)

P10 - P13 Section 3.1

Please read the related general comment. This section is full of mistakes and imprecisions, so I will list only some of them. The authors should note the $C_{(\cdot)}^{-1}$ are the inverse of the error covariance matrices and not the error covariance matrices. In this section there are several inconsistencies between the equations, and also in the text (for example, the matrix C of equation 5 is not the same as the one of equation 3. γ_r of eq. 3 is γ in eq. 4, Δf is not explained, etc). It is not clear what is K^{obs} (matrix of Jacobians? with respect to which variables? what are the “characteristics”?). The symbol J_p is important but it is not explained. The mass is written as M in the text and m in the equations. The definition of adjoint of L12-13 (P11) is vague and incomplete. An appropriate definition of adjoint operator can be found in the equation 2 of Talagrand and Courtier (1987).

P12 L14

Here the authors state that the minimization is performed with the L-BFGS-B algorithm, but equation 3 described the minimization by using a simpler steepest descend algorithm. Do the authors use the algorithm of equation 3 or do they use L-BFGS-B with the gradient as equation 9?. Please clarify.

P14 L5

What is a “well-known qualitative tendency”?

P14 L10

The authors should clarify in which scenario (A, B, C, D, E) were these tests performed.

P14 L18-19

This statement is in general not true. For example, taken the f function equal the identity in equation 3, the cost function is a quadratic function of S in R^n , and the problem is well-constrained (as J is a convex -and smooth- function, the problem has an unique solution), and the solution depends on the prior information.

P15 L7-9

Please add an introductory paragraph before L9.

Figure 7

Should not be divided by surface area?

P17 L8-9, L16

Not using a priori knowledge of aerosol emissions implies much more than the B settings. It is not equivalent.

P17 23

The false source generation is prevented only over ocean; but it is still allowed to generate a source in the wrong place and time. Please clarify.

P17 25

Why do not the authors show this ratio for the retrievals A and B? , how is the uncer-

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tainty of this ratio computed?

P18 , Section 3.2.3

Beyond the synthetic tests, how do the authors account for the (known) sub-daily emission variability of DD, BC and OC emissions?

P20 , Section 3.2.4

Which are the emissions used for the “retrieval E”? I would guess that they are the same as “retrieval C”, but it is not clear. Are DU and OC included in this test? Is there any difference in retrieved DD and OC by changing the BC refractive index?

Figure 10:

Are these gridpoint values? Are they the accumulated emissions (16 days) of day-by-day emissions? What is the grey area (20% of what?)?, the lines? What is “Y”, “X”, “R”, etc.?

P21 L8-22

These conclusions about the idealised test are presented as a fact (and largely extrapolated to other contexts) without taking into account the nature of the synthetic measurements, and the limitations of the whole data assimilation system. I would suggest to present them as the authors’ choice regarding the parameter configuration of the assimilation procedure.

P21 L11

Why are needed 6 and not, for example, 5 wavelengths? Did the authors try with less wavelengths?

P21 L13

Please check the grammar

P21 L18

More stable and accurate than what? Where do the authors show the “stability” of the retrieval in the text? In which sense it is stable?

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P21 L19

Please check the first sentence. It should not be inverted (retrievals sensitive to refractive index) ?

P21 L25

Why are not SS and SU included? Errors in the emission of these aerosols will impact the quality of the posterior emissions. How is this taken into account?

P21 L22 or L24

I would recommend to indicate that this configuration/parameters of the assimilation procedure will be used in Sections 4 and 5.

P21 L28

Even though a fixed number of iterations is a very practical stopping criterion, do the authors compute any diagnostic on the optimality of the cost function after 40 iterations?

P21 Section 4

Please indicate which BC Case (refractive index) is used in the results of Sections 4.1, 4.2, 4.3.1 and 4.3.3.

P24 L13-14

Why could the coarse resolution of the model lead to the spectral differences of AAOD presented here?

P26 Section 4.3.1

How much of the DD reported in Figure 13 is produced in the Sahara? The retrieved emission seasonal cycle seems flat (but maybe it is only the scale of the plot). In comparison with other studies, would the authors think that the seasonal cycle is well captured?

Figure 13.

The units are Tg/month or Tg?

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P27 L11

This value is similar to the 3.4 factor of Kaiser et al 2012. I suggest to add and comment this reference in line 22.

The authors have indicated that the difference between Case 1 and Case 2 retrievals is close to 1.8 for BC. This value have been computed in the sensitivity tests; but in this section the factor should be close to $8/3$ (2.7). How do the authors explain this difference? Should not be better to include in the abstract and conclusion this value instead of the 1.8?

P27 L16-20

Please note that these values are not necessarily for the year 2008.

P27 L27

Can the authors report the uncertainty of the retrieved emissions?

P30 L5

Please indicate which BC case is used in this section.

Figure 18

The colour code of each square in the panels represent the number of pairs (observation,model) that fit in the square. The size of the squares are different in all the panels (in fact, some of them are not squares, despite the same limits of the x and y axes), so they are not comparable. The worst case is in the last column. The authors should write in the caption the size of the AAOD bins (and AOD for Figure 17). I strongly recommend to improve this figure.

P35 L22

This sentence should be written before (around line 10). Is the model sampled according to MODIS availability?

P36 L10-11

I could understand that these kind of measurements are more sensitive to the absorp-

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tion properties, thus the retrieval of them is better constrained, but this first sentence of the paragraph implies that these are the only measurements sensitive to the absorption, which is a strong statement. Could you provide a reference on this?

P36 and P37

From panels a, b and c of Figure 21, it seems that the model/satellite comparison is not collocated. This could introduce errors in the analysis of the results, and it should be mentioned. Also, I would suggest to plot with transparent colour the missing data, and not with blue (which is equivalent to zero AAOD)

P38 L17

Please rephrase this sentence to improve readability (It is not easy to figure out what a "800% lower" means).

References:

Talagrand, O. and Courtier, P. Variational assimilation of meteorological observations with the adjoint vorticity equation. I: Theory. Q. J. R. Meteorol. Soc. 113,1311–1328, 1987.

Kaiser, J. W., et al. Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, Biogeosciences, 9, 527–554, 2012.

Tarantola, A. Inverse problem theory and methods for model parameter estimation. siam, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-35>, 2018.

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