

## Reviewer comments

Emissions of aerosols species remain very uncertain and contribute to the large uncertainty in the impact of aerosols on radiative forcing and climate. This study makes a significant contribution in reducing the uncertainty in the emissions of several aerosol species (i.e. black carbon, organic matter, sea salt and desert dust). The author's use the Bayesian inversion method to assimilate MODIS total AOD and fine mode AOD (over the oceans only) to constrain emissions in several regions globally. The posteriori results are then compared to independent AERONET data. The author's use scientifically sound methods and these are generally well presented. I therefore, recommend this manuscript for publication after a few minor changes are made.

## General comments

I agree with the comments made by the first reviewer, hence, I will try not to repeat things already mentioned here.

The introduction includes a fairly thorough account of previous work in the area of aerosol emissions estimation using data assimilation, however, the purpose of this study is only mentioned briefly in the final paragraph. I would suggest that the authors expand the discussion of what their study contributes, for instance, is it that there have been no previous top-down estimates of SS, POM and SO<sub>2</sub> (previous studies on BC and DD are mentioned)? Also, that this is the first multiple aerosol species inversion.

The writing is sometimes unclear and could be made more succinct.

## Specific comments

p3079, 115: for clarity "the analysis vector" or "state vector" (and hereafter)

p3079, 116-17: this sentence is confusing and should be rewritten

p3079, 123: "e.g." Rodgers et al 2000 (there are many texts about this)

p3080, 118: for clarity "simulated ( $Hx^b$ ) and observed ( $y$ ) values"

p3080, 123-24: the variational approach becomes advantageous only when the  $H$  cannot be defined (either it is too large or it's terms are not known explicitly).

p3083, 12: should outline what these considerations were and add some statement about how these emission inventories i.e. from 1 decade ago are different from recent ones

p3083, 15-11: this should go in the introduction (see general comments).

p3085, 116: the authors say the data were "thinned" do they in fact mean that they were averaged to the lower model resolution. Please clarify.

p3086, 13-4: perhaps the covariance between errors in e.g. OC and BC emissions are difficult to determine, however, one could imagine that such a covariance could be large since they are both emitted by e.g. biomass burning. What is the potential impact of ignoring possibly large covariances such as these?

p3087, 116-17: why are there two error numbers listed directly one after the other?

p3090, 115: approximately how many AERONET data points went into the monthly mean at each site?

p3099, 113: “assess the impact of the assimilation on the errors” here it is not clear which errors are meant. Diagonal elements of  $A$  are the posterior uncertainties of the state variables (as is mentioned 112) so what is the error being referred? It appears as the terms “error” and “uncertainty” are being used here interchangeably, which makes this whole paragraph confusing.

p3100, 113-17: these three sentences repeat information and should be made clearer and more succinct

p3096, 122: Could the authors offer an explanation as to why the MODIS and AERONET data so different at Mauno Loa and for other stations with large differences in general?

Fig 8: MODIS data are missing at Solar Village.

### **Technical comments**

p3080, 120: “the sensitivities of the observation operator ( $H$ ) and the relative weights of the  $R$  and  $B$  matrix” (remove “to”)

p3080, 122: need to use consistent terminology either “state” or “analysis” vector

p3081, 128: “caused by”

p3084, 110: “provided” not “delivered”

p3084, 111: “chose” not “choose”

p3084, 112: “onboard the Terra satellite”

p3085, 115: “south of 40°S” not “over”

p3093, 120: “associated with”

p3102, 121: “corresponding to” or “correspondent with”

p3102, 121: “than at present”

p3103, 16: “large uncertainties in the aerosol impact on climate”

p3103, 123: “one year’s worth”

Fig 8 - 10: figures are difficult to read – the axis labels, titles and legends are too small. The legends could perhaps be removed as it is the same in every sub-plot and simply given in the captions.