

***Interactive comment on “A three-dimensional variational data assimilation system for multiple aerosol species with WRF/Chem and an application to PM<sub>2.5</sub> prediction” by Z. Liet al.***

**Anonymous Referee #2**

Received and published: 28 August 2012

**1 General summary**

**This paper presents the design of a 3D-Var assimilation system for a complex aerosol model (WRF/Chem). The underlying aerosol scheme is**

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**called MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) and allows for the treatment of several aerosol species distributed over four size bins. The building blocks of the variational assimilation system are described in detail, in particular the choice of the analysis variables and the construction of the background error covariance matrix for those variables. This task can be quite daunting for an aerosol model, where there are many variables that can be chosen to be analyzed but often there are no corresponding observations to constrain such variables. In this paper, the authors chose to use the total mass concentrations of five main species: Elemental Carbon, Organic Carbon, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and other organic aerosols (OTR). Increments in these total mass concentrations are then re-distributed into the number and mass**

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concentrations in the individual bins. The repartition assumes weights that are proportional to the inverse of the root-mean-square of the mass concentration background error for each species and each bin. Forecast experiments over the month of May-June 2010 for the Los Angeles area show that the system performs as expected when observations of  $PM_{2.5}$  are assimilated with an increased correlation and reduced RMSE with respect to the assimilated observations in the analysis. Comparisons with independent observations of individual species concentrations, reveal that model biases in compositions cannot be eliminated by the assimilation of  $PM_{2.5}$  observations, even if the analyzed variables are the individual mass concentrations of the five species. Overall the forecast of  $PM_{2.5}$  over 24h is improved in the assimilation run, in particular for

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$SO_4^{2-}$ , but not equally for all species. This highlights the fact that this type of assimilation systems are limited by the availability of appropriate observations to constrain the analysis, and rely heavily on the model, by construction.

The research topic is interesting and the study is well constructed, although very focussed. For example the background error statistics have been constructed exactly for the period and the area under study, and not all results may be general. The paper itself is well-written. The discussion of results could be conducted in greater depth but overall I would recommend publication with some revisions (see below).

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**p. 13515 I.19 The abstract refers to a “downscaling” simulation but the meaning of this is not clear.**

**I. 24 “have” rather than “impose”**

**p.13517 I.9 not only aerosol fields**

**p.13519 I.1 I am not sure that 3DVAR is the most computationally efficient. On parallel architectures Ensemble methods can be shown to be more efficient. Please qualify this statement.**

**I.15 the limiting factor is also the availability of constraining observations**

**p.13522 I.9 OTR is mentioned here but then it disappears from the examples and the error statistics. Any particular reason?**

**p.13530 I.3 The NMC statistics were estimated only**

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**for one month, and exactly the same month for which the DA experiments were run. It would be interesting to see if statistics generated for another (longer) period yield similar DA results. In any case, it should be mentioned.**

**section 6.2 Have you thought at assimilating the single species concentrations and verify with the other observations? It would be an interesting “single observation” experiment**

**p.13534 I.20 verification of the analysis against assimilated observations should be called a “sanity check”**

**p. 13534 Summarize results from control and assimilation run in a table (or use bigger labels - some plots are not very legible)**

**p.13535 I.8 remove “and EC” - it must be a typo because it’s repeated twice.**

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**I.12 replace “inherit” with “inherent”**

**p.13536 I.1-5 elaborate sentence: what is usually observed in meteorological data assimilation, what is different here, possible explanations and solutions**

**I.17 since 3DVAR is a statistical optimization, it should not be expected that all points are improved in RMSE. However, the fact that the RMSEs are higher in coastal location only might be indicative of a bias.**

**I.25 Replace  $\text{NO}_3^-$  with  $\text{SO}_4^{2-}$ . This must be a typo because from the plots it looks like the variable mostly improved in the forecast was  $\text{SO}_4^{2-}$  (unless the plots are mis-labeled).**

**p.13538 I.8 Why were those results not presented? It could be a good argument for the careful construction of the background error statistics**

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**which is a theme of the paper.**

**Figures 6-8-9 need to have bigger labels. Contours in figure 8 are not visible.**

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13515, 2012.

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