

***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. Li et al.***

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**Thanks for the comments. Here we address these comments. Note that the original comments are in italics.**

***Assuming that errors are uncorrelated within***

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***several bins of a single species is also controversial. Since aerosol species from different bins are likely to come from the same emission sources their errors will be correlated. Also, correlation scales for a single species from different bins will be different. As an illustration consider PM<sub>1.0</sub> and PM<sub>2.5</sub>. These two “bins” have different paths to formation which affects scales. Also, because of different physical properties these two “bins” e.g. have different deposition rates which will affect both vertical and horizontal scales.***

**We agree that the correlations between different size bins may be significant. As this comment suggested, we have discussed this issue in the conclusion and discussion section: “This is an ad hoc assumption made simply to render the problem computationally manageable. The**

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correlations between different bins can be significant, because one species often arises from same sources and transfers across the bins. The consequences of this assumption should be quantified, and relaxation of this assumption should be pursued.”

*On p. 13538 l. 8 authors claim that 3D-VAR using total PM<sub>2.5</sub> as a state variable degraded the results. If the experiment showed that it is so it would be worthwhile to document how much benefit is achieved using the assimilation method with aerosol species as state variables in 3D-VAR.*

When PM<sub>2.5</sub> is used as a state variable, the data assimilation scheme then follows the two-step methodology as described in section 4. The first step is to compute the PM<sub>2.5</sub> increment, and the second step to distribute the PM<sub>2.5</sub> increment across different species and size bins. The

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increment for each species and bin has the same sign as the one associated with the PM<sub>2.5</sub> increment. In the experiment presented, the EC and OC concentrations show a positive bias, but the NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> concentrations a negative bias. Since the background state shows a negative bias in PM<sub>2.5</sub>, a 3DVAR scheme generates a positive increment in PM<sub>2.5</sub> and thus inevitably deteriorates the positive bias in the EC and OC concentrations.

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