Detailed response to Reviewer's comments.

Review of the manuscript

Implementation of aerosol assimilation in Gridpoint Statistical Interpolation v. 3.2 and WRF-Chem v. 4.3.1 by M. Pagowski et al. GMDD 7, 2483-2500 (2014) General comments

In this manuscript, the authors present an extension of the GSI assimilation tool which allows for the assimilation of aerosol PM2.5 and PM10 concentrations from surface measurements and aerosol optical depth from satellite measurements.

Given the importance of having accurate aerosol representation in air-quality studies, this extension is certainly useful for forecast models. In addition to that, the proposed package also provides statistics and conversion tools which are helpful for model users.

The manuscript, however, should be improved before publication in GMD. In particular:

• some technical terms needs to be explained; • more accuracy is desirable in Section 3 and 3.1; •the figures need to be improved; • language can be improved (e.g., the use of articles).

Please find more detailed comments below.

Authors appreciate Reviewer's attention and comments.

Major remarks

P2484-L17: I understand that this is a technical paper, but I would try to rephrase this sentence in a more positive way (e.g., "Scientific aspects are also briefly discussed").

Agreed, corrected as suggested.

P2484-L21: it is not clear to me what you mean by "continuously recycled". Please clarify.

Agreed, corrected: ..." while chemical species were obtained from the previous forecast without referring to observations."

P2485-L2: please explain what 3-D and 4-D refers to. I guess that 3-D is for time, latitude and longitude, whereas 4-D also includes the vertical coordinate. How is such vertical coordinate defined?

3-D stands for spatial dimensions, 4-D - for spatial dimensions plus time. Vertical coordinate is defined as in the model, can be e.g. pressure, height, a derived coordinate such as e.g. sigma-p.

P2485-L2: provide a definition for "ensemble Kalman filters".

We provide references for variational and ensemble Kalman filters. We feel that it would be a distraction from the narrative to introduce more detailed descriptions of variational and Kalman filter methods.

P2486-L13: if possible, provide a reference for the BUFR format.

Reference on BUFR provided: Dragosavac, M.: BUFR User's Guide, ECMWF Technicalnote,availableonlineat:http://www.wmo.int/pages/prog/gcos/documents/gruanmanuals/ECMWF/bufr_user_guide.pdf, 2007.

P2487-L2: in case observation values are not available in some grid boxes, how are these treated in the assimilation procedure? Are they simply ignored or are they interpolated from the nearest valid gridboxes?

We modified description of Equation (1) to make it more transparent.

Observation operator "H" calculates model value of observation. In case of surface aerosol observation it is just bilinear interpolation to observation location. For AOD mixing ratios of aerosol species are interpolated to latitude/longitude and summed up in the vertical for all species and levels. Difference between this calculated value and the observation itself is used in minimizing the cost function in the equation. Observations are never ignored unless they fail QC criteria or are outside of the modeling domain.

P2487-L9-26: this paragraph needs to be extended with a more detailed explanation. Methods like "recursive filters" and "incremental approach" are not explained at all and just providing a reference is not sufficient. The terms in Eq. (1) are listed but the actual meaning of some of them (e.g., the observation operator H) is not explained. I would recommend to add a few sentences to make this paragraph easier to understand, especially for non-experts.

We modified the paragraph, note response to the previous query.

P2488-L9-10: please specify the values of the size bins used for dust and sea- salt.

Corrected as requested.

P2488-L18: the factors that account for the size cut-off at 2.5 micron requires an assumption on the size distribution of dust and sea-salt particles. Is that the case? If so, please provide more information on such assumption.

Yes, that is correct. It is assumed that dust and sea salt have lognormal distributions. Details are given in WRF-Chem guide available on-line.

P2488-L25: here aerosol size modes are mentioned (lognormal modes, I guess), while in the previous paragrah size bins for dust and sea-salt are mentioned. This is a bit confusing. Is the model using size modes or size bins?

We clarified that in the text. For calculating AOD for each aerosol species and within each size bin a lognormal distribution is assumed. Parameters of the distributions are given in Liu et al. 2011.

P2489-L5: the parameters pdk is not defined and the value of ktop is not given.

ktop stands for index value at the top of the model, that would vary on an application, ρ_d has been defined.

P2489-L19: the default value 0.5 for the parameter α is somewhat arbitrary. What does it actually represent and how should it be chosen?

The default value of 0.5 was obtained by tunning, i.e. to obtain better verification statistics for forecasts. To the best of our knowledge there is no objective method to determine representativeness error. In essence, value of α determines magnitude of the total observation error.

P2489-L23-24: the reason for this thresholds is not clear. Why are such cut-off values applied?

Such cut-offs are applied to sort out unrealistically high values of measurements which are not quality-controlled. User can modify these values if required, possibly depending on model grid resolution.

P2490-L4: what is meant by thinning? Is this a regridding to a coarser resolution? Please clarify.

"Thinning" is a technical term used for this kind of procedure e.g.

Ochotta, T., Gebhardt, C., Saupe, D. and Wergen, W. (2005), Adaptive *thinning* of atmospheric observations in data assimilation with vector quantization and filtering methods. Q.J.R. Meteorol. Soc., 131: 3427–3437. doi: 10.1256/qj.05.94

We added a synonym in the brackets in the manuscript.

P2490-L13: why can variance and correlation length only vary zonally and vertically? Is there no dependence on time and/or longitude?

Variance and correlation lengthscales vary zonally and vertically in GSI by design.

Justification is given by latitudinal dependence of atmospheric thermodynamics and dynamics. For chemistry, such assumption may not be fully justified as these parameters depend also on emission sources. For that reason an alternative approach is offered – "ratio approach", discussed in the section on background error.

Time dependence would be possible if the statistics were derived separately for different simulation times. Then, for the assimilation at a specific time matching statistics would be used. In practice, such approach is not common in applications of 3D-Var data assimilation.

P2492-L9: as an alternative, the diffv operator from the CDO package is also very useful (<u>https://code.zmaw.de/projects/cdo</u>).

Thanks for pointing to this package, we include a note in the text.

Figure 1: on the vertical axis, I would provide the actual pressure or altitude coordinate.

 $-log(p/p_s)$ display is common in papers on data assimilation and unless strongly objected we would prefer to retain it. Also, $-log(p/p_s)$ maintains certain affinity with height coordinate that results from the solution to hydrostatic equation for isothermal atmosphere. It is difficult to provide absolute values for pressure or height on the ordinate since these values depend on surface pressure, which is itself dependent on topography. In the caption we provide approximate pressure values corresponding to $-log(p/p_s)$ in the figure.

Figure 2 and Figure 3: units are missin

Figures have been corrected.

Minor remarks

P2484-L3: please provide in brackets the country of the National Centers for Environmental Predictions (USA?). Corrected as requested.

P2484-L4: "the implementation" (article missing) P2484-L8: "make" \rightarrow "to be made"

Corrected as requested.

P2484-L20: "meteorological assimilation was only applied to meteorology"; this sounds like a repetition, I would delete "meteorological".

Corrected as requested.

P2486-L3: the URL seems to have changed to airnowapi.org. Please check and update.

Corrected as requested.

P2486-L8: "and rural" \rightarrow "and on rural".

On request from the other reviewer we removed this sentence and the detailed description of the data.

P2486-L19: "both aerosol" \rightarrow "both PM aerosol".

Corrected as requested.

P2488-L4: "The forward models" (article missing).

Corrected as requested.

P2488-L8: I guess P25 is a typo for PM2.5.

P₂₅ is the name of unspecified aerosol which is a component of PM_{2.5}

P2488-L8: "(BC1, BC2)" \rightarrow "(BC1 and BC2, respectively)". P2488-L9: "(OC1, OC2)" \rightarrow "(OC1 and OC2, respectively)".

Corrected as requested.

P2489-L15: "A representativeness error" (article missing).

Corrected as requested.

P2489-L25: "distance" \rightarrow "difference" or "deviation".

Corrected as requested.

P2490-L4: "the volume" (missing article).

Corrected as requested.

P2490-L9: "section on background error" \rightarrow "the next section".

Corrected as requested.