Dear Editor,
Below please find our responses to Reviewers' comments.
Best regards,
Mariusz Pagowski

Detailed response to Reviewer I 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 Technical note, available online at: 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 ρdk 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 (https://code.zmaw.de/projects/cdo).

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" → "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” → “and on rural”.

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

P2486-L19: “both aerosol” → “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.

P_{25} is the name of unspecified aerosol which is a component of $PM_{2.5}$

P2488-L8: “(BC1, BC2)“ → “(BC1 and BC2, respectively)”.
P2488-L9: “(OC1, OC2)“ → “(OC1 and OC2, respectively)”.

Corrected as requested.

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

Corrected as requested.

P2489-L25: “distance” → “difference” or “deviation”.

Corrected as requested.

P2490-L4: “the volume” (missing article).

Corrected as requested.

P2490-L9: “section on background error” → “the next section”.

Corrected as requested.

Detailed response to Reviewer II comments.

Review of Pagowski et al.
The paper concerns an important aspect of data assimilation, namely development of techniques to assimilate aerosol data. As such it is timely and of interest to the general data assimilation community. The paper is technical, rather than scientific, and its inclusion in GMD/GMDD is warranted. The paper should be suitable for publication in GMD once the authors address the general and specific comments below.

General comments:
Introduce acronyms when first used, both in the abstract and the manuscript, e.g., PM, MODIS. The English needs improving, including avoiding repetition of text.
The paper would benefit from inclusion of references to back statements made. See specific comments.

Authors appreciate Reviewer's attention and comments.

Specific comments:

P. 2484:
L. 2: Introduce NCEP acronym - it is well known in the data assimilation community.

NCEP acronym introduced.

L. 6: Perhaps better to say: "...we present illustrative results...".

Corrected as requested.

P. 2485:
L. 1-2: Many of the references cited concern tropospheric chemical data assimilation (e.g. the Elbern work). There has also been work on stratospheric chemical data assimilation, with a focus on ozone (e,g,, Geer et al., 2006). The authors should also refer to chemical data assimilation within the MACC, MACC-II projects, e.g., Massart et al. (2014).

References on stratospheric assimilation provided.

Regarding the variational and ensemble data assimilation methods introduced, general references could be provided here. Examples include: Bouttier and Courtier, 1999; Talagrand, 2010; Kalnay, 2010.

References on assimilation methods provided.

L. 10: I suggest the authors provide references for air quality forecast systems. One example is Rouil et al. (2009) – PREV'AIR in France; the work of Elbern provides

another example.

We include Rouil et al. (2009) in the text and references.

L. 16+: Indicate here what you will discuss in the paper.

More detailed description provided.

P. 2486:
L. 1+: Is all this detailed information necessary?

Agreed, some information was superfluous and has been removed.

L. 20: I suggest replacing "inaccuracy" with "error". Please specify whether this error is random or otherwise.

Agreed, corrected.

L. 29: Is this error the random error?

Remer et al. (2005) do not discuss distribution of observation errors. They derive error estimates using least-squares which are the best linear unbiased estimator of any linear combination of independent observations. We cannot infer whether observation errors have a normal distribution though it seems to be a justifiable assumption based on the figures in the paper. This paper is the most authoritative reference on MODIS errors (over 1100 citations).

"In most cases, MODIS and AERONET exhibit very similar annual cycles, often with very similar magnitudes of optical thickness. Two- thirds of the differences in optical thickness over land are less than 0.10. There is some indication that MODIS retrievals over land may be systematically biased high, but in most cases the difference is still well within the estimated uncertainty of +/-0.05 +/- 0.15."

"Globally 62%, 66%, and 70% of all retrievals over ocean at 0.55, 0.66, and 0.87 μm, respectively, are falling within the narrowly defined expected uncertainty. Only the 0.55-μm channel is falling outside of the error bars more often than the prelaunch expectations of 66%, albeit slightly. The average at 0.55 μm is 0.18 for the ocean global database at defined AERONET stations, the same as for land. Because the land and ocean databases include many of the same stations, this is not surprising. The percent error between MODIS ocean retrievals and AERONET observations at 0.55 μm is only 1%, showing the same absence of bias exhibited in Fig. 9."

"An extensive validation effort that collocated over 8000 MODIS retrievals with AERONET measurements of optical thickness show that globally, the MODIS products are accurate to within prelaunch expectations, namely, +/-0.05 +/- 0.15 over land and +/-0.03 +/- 0.05 over ocean. In particular, the retrieval of aerosol over oceans consistently shows remarkably good agreement with virtually no offset or bias through the range of optical thickness where most observations occur. Regional analysis, however, shows specific issues for certain locations."

P. 2487:
L. 15: Indicate that H is a non-linear operator. Is it linearized in your system?

H can be non-linear, it is linearized within the inner loop as in the incremental approach of Courtier et al. (1994). We extended description of Eq. (1) to clarify.

L. 16: R typically includes the representativeness error.

Agreed, corrected.

P. 2488:
L. 17: sea salt.

Agreed, corrected.

P. 2490:
L. 24: Define the "increment".

Increment defined.

P. 2492:
L. 4: The subscript should be "2.5".

The edited manuscript incorrectly subscripted PM25/PM10. PM25/PM10 are actual names to be entered in the table, i.e. no dot required in PM25.

L. 5: Please elaborate on what you mean by "regressions", and discuss their purpose here.

We removed reference to regressions as not essential. We believe it would be a distraction from the text to discuss this aspect of data assimilation in the manuscript.

L. 26: Quantify this "improvement". Significant in what sense? Statistical?

Figures clearly show large improvement in correlation, it is statistically significant. We rephrased the sentence.

Figures:
I suggest make the figures bigger, and provide details of the colour scale range in the figure captions.

Agreed, figures were enlarged and corrected to include aerosol name and units.

References:

Bouttier, F., and Courtier, P. (1999). Data Assimilation Concepts and Methods. ECMWF training notes. Available online at: http://www.ecmwf.int

Geer, A. J., Lahoz, W. A., Bekki, S., Bormann, N., Errera, Q., Eskes, H. J., et al. (2006). The ASSET intercomparison of ozone analyses: method and first results. Atmos. Chem. Phys. 6, 5445–5474. doi: 10.5194/acp-6- 5445-2006

Kalnay, E. (2010). "Ensemble Kalman filter: current status and potential," in Data Assimilation: Making Sense of Observations, eds W. A. Lahoz, B. Khattatov, and R.

Ménard (Berlin: Springer), 69–92.

Massart, S., Agusti-Panareda, A., Aben, I., Butz, A., Chevallier, F., Crevosier, C., et al. (2014). Assimilation of stratospheric methane products in the MACC-II system: from SCIAMACHY to TANSO and IASI. Atmos. Chem. Phys.. 14, accepted.

Rouïl, L., Honoré, C., Vautard, R., Beekmann, M., Bessagnet, B., Malherbe, L., et al. (2009). PREV'AIR: an operational forecasting and mapping system for air quality in Europe. Bull. Am. Meteorol. Soc. 90, 73–83. doi: 10.1175/2008BAMS2390.1

Talagrand, O. (2010). "Variational assimilation," in Data Assimilation: Making Sense of Observations, eds W. A. Lahoz, B. Khattatov, and R. Ménard (Berlin: Springer), 40–67.

Above references are included.