Dear Editor,

Please consider for publication in *Atmospheric Chemistry and Physics Discussions* the revised version of the manuscript

Ms. Ref. No.: acp-2011-473: "Ozone Data Assimilation with GEOS-Chem: a Comparison Between 3D-Var, 4D-Var, and Suboptimal Kalman Filter Approaches" by K. Singh et al.

The detailed response to reviews are below.

Thank you very much,

Kumaresh Singh

Reviewer 1

Comments to Author: This paper compares 3D-Var, 4D-Var, and suboptimal Kalman filter (KF) tropospheric ozone estimates based on ozone observations from the NASA Tropospheric Emission Spectrometer and the GEOS-Chem chemistry transport model. The produced analyses are evaluated on the similar lines of Geer et al., 2006. The novel aspects of this paper are:

- The first direct objective evaluation of global tropospheric ozone analyses obtained by the intercomparison of 3D-Var, 4D-Var and suboptimal KF assimilation solutions.
- The introduction of an original technique that propagates 3D-Var increments back to an equivalent initial condition, which provides a suitable comparison with 4DVar.

This is a generally good paper which contains results of interest for the general chemical assimilation community. It is quite close to being acceptable in ACP as it stands. However, I would like to see some changes related to the discussion in parts of the paper, which will improve the paper if it is implemented. These changes are listed below:

Major comments

p22264 / Eq. (21): The elements in the forecast error covariance matrix block (corresponding to each observation grid point) need to be squared.

Answer: Corrected.

p22268 / Fig. 4: More detail is needed in the discussion of the localized overcorrection in the mid west Australian region brought by the suboptimal KF system. Given the fact that 3D-Var and suboptimal KF processes are assumed to provide similar estimates, could you please explain in more detail why the overcorrection is visible in the KF solution and not in the 3D-Var one? There are issues related to the difference in error covariance characteristics specified for the two assimilation schemes that could lead to such discrepancies:

– The forecast error variances at observation time i+1, in the suboptimal KF case, are constructed by transporting variances at observation time i as passive tracers, which is not the case in the 3D-Var process.

– As used in this paper, the KF solves the same statistical problem as 3D-Var, but in observation space, then maps the solution to model space. This observation space approach of KF uses a forecast error covariance matrix that differs from the model-space 3D-Var background error covariance matrix. The KF scheme used in this study is more comparable to the PSAS approach (Physical-space as Statistical Analysis System), which solves the analysis in the observation space.

Answer: The above two comments correctly explain the differences in the amount of overcorrections in KF and 3D-Var cases, and have been added to the text as follows:
"*The overcorrection in the mid west Australian region which was not visible in the 3D-Var case for 5 days assimilation window, seems to be prominent in longer assimilation, while, for suboptimal KF, it has been accentuated. The fact that this phenomenon is seen only in 3D-Var and suboptimal KF could be attributed to the propagation of emissivity errors described earlier, combined with the localized correction property of these methods. Differences in the amount of overcorrection in the two methods could be attributed to differences in their implementations. For example, as described in section 2.3, the forecast error variances at observation time* $i + 1$ *in the suboptimal KF case, are constructed by transporting variances at observation time i as passive tracers, while in case of 3D-Var, they are generated from the current model state. Also, the KF implementation used in this study solves the underlying statistical problem generating analysis in observation space, then maps the solution back to the model space. While in 3D-Var, the same statistical problem is solved in the model space itself, leading to different forecast error covariance matrices*".

– Neglecting spatial correlation (diagonal background error covariance matrix) would lead to giving too large weight to the back-

ground in the 3D-Var analysis. In order to compensate for that effect, the assumed variance of the errors should be increased.

Answer: No change. Values of the forecast error covariance matrix entries and effects of spatial correlations for similar set up have been studied in detail in Singh et al. [2010a].

– The J_{min} (cost function at the minimum) approach provides a diagnostic of the consistency of an assimilation algorithm (Talagrand, 2003). The error covariances are considered to be properly specified in the case when:

$$E\left(\frac{2J_{min}}{N_{obs}}\right) \sim 1$$

Where E is the statistical average (expectation) and N_{obs} is the number of observations used in the analysis.

In the case when $E\left(\frac{2J_{min}}{N_{obs}}\right) > 1$ the covariances are underestimated, and vice versa. If the authors have already produced this diagnostic, it may be useful to report the results of it in the paper.

– The χ^2 (chi-square) diagnostic can also be used as a self-consistency check of the specified error covariances for the KF assimilating system (Khattatov et al., 2000). For each assimilation analysis the value of χ^2 can be computed as:

$$\chi^2 = \left(x^{obs} - Hx^f\right)^T \left(HBH^T + R\right)^{-1} \left(x^{obs} - Hx^f\right)$$

Assuming that observation error covariance matrix R is known, and the Gaussian assumptions are considered appropriate, the statistics of χ^2 can be used as verification tools for the forecast error covariance matrix P^f . This latter is considered to be properly specified in the case when :

$$E\left(\frac{\chi^2}{N_{obs}}\right) \sim 1$$

In the case when $E\left(\frac{\chi^2}{N_{obs}}\right) > 1$ the covariances are underestimated, and vice versa. If the authors have already produced this diagnostic, it may be useful to report the results of it in the paper.

Answer: We have not generated diagnostics mentioned in the above two comments but error covariance matrix for KF system used in this study is same as described in the well accepted article by Parrington et al. [2008].

p22270 / Fig. 7 : Could you please explain why, in the longer assimilation case, the localized overcorrection in the mid west Australian region seems to be prominent in the 3D-Var solution and accentuated in the KF one?

Answer: The increase in overcorrection with longer assimilation window could be accredited to two factors. The observation data over mid west Australian region seems to be spurious and use of such data over time causes an overestimated analysis which could not be smoothened out due to localized correction property of 3D-Var and KF and use of diagonal error covariance matrices. This leads to another issue where we construct our forecast variances using the propagated analysis every observation time step (with different implementations though) leading to inaccurate entries in the error covariance matrices.

Minor comments

p22270/ Sect. 6.3 : It should also be mentioned in Sect. 6.3 that, in a 4D-Var analysis, error covariance propagation plays at least some part in explaining the differences between 3D-Var and 4D-Var.

Answer: We have added the following text:

"*It is also worth reiterating that the background error covariance matrix in 4D-Var is static and flow independent, while in case of 3D-Var, it is constructed every observation time through model evolution of the analysis generated at previous observation time.*"

In more recent studies to make 4D-Var operationally affordable, researchers are using what is called incremental 4D-Var proposed by Courtier et al. [1994] where the minimization of the full nonlinear cost

function is approximated by a series of minimizations of quadratic cost functions with linear constraints. These are derived by assuming that the evolution of small perturbations to a given base trajectory can be approximated using a linear model. Usually this linear model is taken to be the tangent linear model (TLM) of the discrete nonlinear model [Lawless et al., 2005]. In such a case, the background error variances along with analysis variances implicitly become flow dependent and are propagated to each observation time through TLM. This is not the case in our study.

p22270/ Sect. 6.3 : It would also be extremely useful for chemical assimilation community to compare 4DVar with 3D-FGAT (First Guess at the Appropriate Time), a variant of 3D-Var in which the objective function is calculated by comparing observation values with the background at the relevant observation times. The 3D-FGAT scheme uses a more exact innovation vector than does standard 3D-Var, in which all observations over the assimilation window are compared to the same first-guess field. 3D-FGAT can be easily implemented by taking the adjoint model as the unit operator in Eq. (11) of the paper. The gradient of the cost function in 3D-FGAT should not use any explicit dynamics other than the trivial dynamics expressed by the identity operator. The increment to be added to the background state is, hence, constant over all the assimilation window instead of propagating it with a linear model. Therefore, it is possible to perform a direct comparison of the ways 3D-FGAT and 4D-Var use the observation information to constrain the model at the beginning of the assimilation interval.

Answer: In this study, we have selected the most widely used data assimilation methods. Comparison with other potentially very useful algorithms such as 3D-FGAT will be done in future work.

Typos etc

p22249/ l1 : to explore the role of of uncertainties (OF).
p22251/ l12 : the characteristics 3-D-Var and 4-D-Var (OF 3-D-Var and 4-D-Var).
p22251/ l16 : Geer et al., 2006 PROVIDE (not PROVIDES) an intercomparison of STRATOSPHERIC ozone (not TROPOSPHERIC).
p22252/ l2 : The assessment of analyses generated through different

assimilation systems IS (not ARE).
p22256/ l16-17 : Remove l16 and replace AND in l17 by WHERE.
p22259/ l6: The corresponding TES observation operator (3).
p22274/ l17: The mathematical formula of the 3-D-Var equivalent initial condition $x_0^{e(3)}$ needs to be corrected.
p22276/ l24: the IONS datasets were (not WHERE).

Answer: All the suggested corrections made.

Reviewer 2

Comments to Author: Singh et al. present tropospheric ozone analyses obtained using TES observations, the GEOS-Chem model and three assimilation schemes based on, respectively, 3D-Var,KF and 4D-Var. The evaluation of the analyses is carried out by means of comparisons against independent ozonesonde observations. At this stage, I found the description of the experiments not clear enough. As a result, the review of the paper was difficult. Whatever the accuracy of my understanding of the paper, I suggest that you clarify it or improve the experimental setup. This will certainly need an additional iteration in the review. However such a study is very valuable and I encourage the authors to not give up.

Major comments

The setup of the different systems and the use of the analyses do not seem to be done properly. As I found the text not very clear, I will first present my understanding of your work and my point of view on it.

- TES data are assimilated from the 1st of August initialized by a GEOS-Chem free model run. Two comparisons are performed considering TES data for 5 days and 2 weeks. Four assimilation runs are performed: one 3D-Var, one KF and two 4D-Var. The first two are conducted with an assimilation frequency of 4 hours while the 4D-Var experiments are performed with an assimilation window of 5 days and 2 weeks, respectively.

  Answer: There are four assimilation runs performed: 3D-Var and suboptimal KF both with 2 weeks assimilation window and 4D-Var with 5 days and 2 weeks assimilation windows. Each of these runs started with the same forecast generated by free GEOS-Chem model run. The observations were read every 4 hours during each 3D-Var, KF and 4D-Var assimilations. For further clarification, following paragraph has been added to the text:
  "*To assess the quality of analysis generated by the above mentioned assimilation techniques, we provide in section 6.2, various plots including comparison of analyses against ozonesonde observations and global ozone distribution for 5 days and 2 weeks assimilation*

windows. It is important to note that, since 3D-Var and suboptimal KF are sequential in nature, we did not have to run 5 days assimilation for these two methods separately. Rather, we used the saved analyses generated during the 2 weeks assimilation."

– On P22266/L17-18, it is mentioned, for sequential methods, that "as we move forward in time, the analysis field agrees better with the true state of the atmosphere". This is because the initial conditions used on 1st of August are too far away from the data. Before comparing the different schemes, you should run several cycles of assimilation such that the differences between the forecast and the observations reach a certain constant value. The analyses during this period, which is called the spinup period, should not be used in the comparison. Typically, this period is between one and two weeks, depending on the data coverage. Note that this period also exists for 4D-Var and you should also perform several 4D-Var cycles to ensure that the spinup period is not taken into account in the comparison. This could explain the difference between the two 4D-Var analyses using a 5-day and 2-week assimilation window, respectively.

  Answer: That is correct. However, in order to ensure that all data assimilation algorithms use the same settings, we use same initial conditions for all of them. If we consider spinup and would like to perform assimilation for August 1st onwards, we will have to start from sometime in July to perform the spinups, where spinup periods for 3D-Var, KF and 4D-Var vary. Our study actually provides a rough estimate of spin up periods as well. Also, it shows the efficiency of one method over the other in terms of speed of convergence, CPU time, and memory usage.

– The comparison of the analyses should be done at their optimal time. For 3D-Var and KF, this time corresponds to the analysis time. 4D-Var analyses, at the beginning of the assimilation window, behave more likely as the observations (noisier analyses). On the other hand, 4D-Var analyses become more likely a pure model at the end of the assimilation window (smoother analyses). Thus, the optimal time of 4D-Var analyses is at the middle of the

assimilation window, not the end of the assimilation window as shown in Figs. 3, 4, 6 and 7.

Answer: Yes, the optimal time of 4D-Var analyses is at the middle of the assimilation window. However, if we consider this time for comparison, 3D-Var and KF routines will not have used all the observation data as they are sequential. Hence, we made the choice of comparing the analyses at the end of assimilation windows for all the methods.

– It is also not clear why two assimilation windows are used in the case of 4DVar and why the two assimilation windows are so long. Such long windows might be justified if the aim of using 4D-Var is to constrain unobserved region or unobserved species. As only one analyses could be used within a cycle (see my previous comment), your setup provides only one analyses every 5 days or every 2 weeks. If the final goal of this study is to use GEOSChem to provide tropospheric O3 analyses, this frequency seems too long with respect to the transport time scale in the troposphere. Maybe using a one-day assimilation window would be a better choice (insofar as this is a natural choice that corresponds to a diurnal cycle).

Answer: There are various reasons for the choice of assimilation window lengths:

(1) 1-day assimilation window length is typical for regional models such as CMAQ, however, for global models like GEOS-Chem, window length of several days is suitable to capture all the physical and chemical dynamics.

(2) The Aura satellite covers the whole globe in two weeks. Therefore, the two weeks assimilation window was chosen to use maximum observation data. Window length of 5 days was chosen to see how the generated analysis varies by changing assimilation window lengths.

(3) Our global model includes stratospheric-troposhperic ozone exchanges through LINOZ chemistry routine which injects ozone from stratosphere into the troposphere. Since ozone

lifetimes in stratosphere are longer than several days, it was important to perform longer assimilations.

– It is stated in the paper that comparisons between the analyses follow the method presented in Geer et al. However, Geer et al. make the comparison for a much longer time period, using a bigger set of independent data with an evaluation of the analyses in different latitude and altitude regions. I found the domain of the comparison used in this paper to small to make any definitive conclusion (ozonesondes about Northern America). The authors might consider a longer period of assimilation (outside the spinup) and make comparison with ozonesondes in different regions. Using ozonesonde data from another network, such as NDACC, might increase the number of independent ozone profiles.

Answer: Agree and we have noted this point in our article. In fact, Figures 3,4,6, and 7 are provided to show that the generated analyses are not ambiguous on a global scale. Although, we do not claim anywhere that our generated analyses are the best estimate of the global ozone distribution since we lacked the data for global comparison. Our comparison is exact similar to the well accepted article by Parrington et al. [2008] which is specifically for North America.

The description of the model used in this study is not very clear, probably because the information is split in different places in the paper. I would recommend that you merge the information in Sec. 3 and the first of Sec. 6.1 in Sec. 5. Moreover, Sec. 5.1 does not aim at describing the different schemes. It is rather devoted to a discussion on the setup of the assimilation window. Perhaps this section could be renamed accordingly. Most important: I still do not know whether the chemical scheme of GEOS-Chem used in this study is based on linoz (see P22261/L9), SMVGEAR or KPP (see Sec 6.1). This is also important to understand Sec. 6.3.1 (which I failed; see my specific comments).

Answer:

a. We would like to keep Section 3 separate as an introduction to

GEOS-Chem and its adjoint since that is our main model on which this study is based. However, we have merged the first part of Section 6.1 into Section 5.

b. Title of Section 5.1 changed from "Data assimilation schemes" to "Assimilation window lengths".

c. The following text is now present in Section 5 which seems to eradicate the confusion mentioned in the above comment:

"*The 3D-Var and suboptimal KF frameworks use Sparse Matrix Vectorized GEAR (SMVGEAR) solver for chemistry. However, to construct the adjoint of chemistry required by the 4D-Var, we implemented the Kinetic PreProcessor (KPP) solver [Damian et al., 2002] into GEOS-Chem which not only provides a suite of high performance chemical solvers to choose from but also generates automatically the continuous and discrete adjoint codes [Daescu, 2000, 2003; Sandu et al., 2003a,b]. A detailed discussion on interfacing KPP with GEOS-Chem and comparison with native SMVGEAR solver for accuracy and computational performance is presented in Eller et al. [2009]. Thus, 4D-Var has KPP, and 3D-Var and suboptimal KF have SMVGEAR as their underlying chemistry solvers.*

The GEOS-Chem model over which the three assimilation methods are built upon, has been modified further to use the linearized ozone (linoz) scheme [McLinden et al., 2000] for a better estimate of ozone exchanges at troposphere-stratosphere boundary. This scheme is available in GEOS-Chem v8 and higher (see `http://geos-chem.org` *).*"

I found the discussion in Sec. 6.3.1 not very clear. I would suggest that the authors follow the method elaborated by Fisher and Lary (1995) to derive the potential influence of one observed species on the other non-observed species included in the model.

Answer: We thank the reviewer for this observation. The plots in the paper represent *scaled sensitivities*, and this was not explained in the first draft; moreover, wrong units were assigned to these scaled sensitivities, and this has contributed to the lack of clarity. These

plots are similar to the metric of Fisher and Lary (1995). We modified the text for clarity and the section now has the following text:

" *TES ozone profiles provide information that can potentially constrain ozone precursor initial conditions as well. (This can be achieved by extending the vector of control variables to include the initial conditions of additional species, and continuing the optimization. Large gradient components corresponding to initial* CO*,* NO_X*, and* PAN *indicate that changes in these initial conditions lead to a considerable decrease in model-observation mismatch). Additional information can be obtained from influence functions, which are ratios of scaled sensitivities (Fisher and Lary, 1995).*".

At this stage, I have not yet evaluated the Sec 6.3.2.

Answer: Nothing to change.

I found numerous typos and misleading sentences. I strongly recommend that the authors take some extra time to reread carefully their paper before the next submission.

Answer: Should be better after the corrections.

Minor comments

P22250/L15-18: The first papers that discussed chemical variational assimilation are: Fisher and Lary (1995); Elbern et al. (1997); Khattatov et al. (1999); Errera and Fonteyn (2001). They could be mentioned.

Answer: Added.

P22250/P19: replace "Khattatov et al. (2000)" by "Khattatov et al. (1999)" as the first one only discussed suboptimal KF.

Answer: Changed.

P22250/4: Lahoz and Errera (2010) provide a review of chemical data assimilation and worth being cited.

Answer: Added.

P22257/L3-4 and next : The sentence says that P_f is discussed in sec 5.2 while it is discussed in the next . Please, clarify.

Answer: No change. The structure of the P_f is described in Section 5.2 "Specification of background error variances" along with background error variance used in 3D-Var. The next describes analysis error covariance matrix P_i^a.

P22257/L21: Remove 'gas phase' because I suppose your model also includes photodissociation and heterogeneous reactions.

Answer: 'gas phase chemistry' changed to 'tropospheric chemistry'.

P22258, title of sec 4: This section discusses mostly the observation operator. This should be mentioned in the title of the section.

Answer: removed 'observations' from the title. The title 'Tropospheric Emission Spectrometer (TES)' is now suitable to the text under this section as it introduces what TES is and how the observation operator and profile retrievals are related.

P22258/L18-19: I could deduce the connection between the thermal contrast and the high latitudes but I would like the sentence be more explicit.

Answer: The statement has been changed to "*Only TES profiles between* $60°S$- $60°N$ *are considered because lower poleward thermal contrast pose difficulty in measurements leading to higher inaccuracies in the retrieved profiles.*"

P22259/L6: what is 'The corresponding TES observation operator 3'? Is '3' referring to a version number? Please, clarify.

Answer: '(3)' refers to equation number. Missing parentheses have been added.

P22259/L21: 'The points ...': do you mean the model grid points or the observation location? To avoid any misleading, use observation 'locations' instead of 'points' in the paper.

Answer: The sentence has been modified to "*The model grid points which do not lie on the observation locations in observation space remain unaffected by the assimilation*". Also observation 'grid points' changed to 'locations' throughout the text.

P22260/L16: Section 3 mentions GEOS-Chem v7.2.3 while Section 5 mentions v7-04-10. This is confusing and must be clarified.

Answer: Corrected to v7-04-10 throughout the text.

P22261/L9: In this study, does GEOS-Chem use a full chemical scheme or does it use the linoz ozone parameterization?

Answer: As described in a previous comment, GEOS-Chem uses full tropospheric chemistry (SMVGEAR in case of 3D-Var and sub-optimal KF, and KPP in case of 4D-Var), in addition to LINOZ for stratospheric-tropospheric flux exchange.

P22261/L26-28: I dont understand this sentence, especially with the fact that the B matrix is supposed to be diagonal.

Answer: The sentence "It is also worth mentioning that these new initial conditions are used to construct a new background error covariance matrix (17) every observation window" means that the analysis generated at observation time i which is propagated through the forward model acts as initial condition for observation time $i + 1$ and is also used to construct the background error covariance matrix required for cost function calculations. Even though B is diagonal, the diagonal entries are different for different observation times.

P22262/L9-11: On which base are the two assimilation windows for 4D-Var chosen?

Answer: Answered in a previous comment.

P22262/L15-17: Why is the cost function in 4D-Var updated every 4 hours and not at every model time step? Is the model time step 4 hours, too?

Answer: The cost function is updated every 4 hours for 4D-Var while a new analysis is generated every 4 hours in case of 3D-Var and suboptimal KF. This is explained in detail in Section 5.1. With the reorganized text, the first paragraph of this section (5.1) should be able to answer the above comment:

"*The TES data in all our data assimilation experiments were read once every four simulation hours; the observation operator called at model*

time t (hours) reads in all the measurements collected within the interval $t-2$ (hours) to $t+2$ (hours). This collective reading increases computational efficiency since reading through observation data files is an expensive process. However, this assumes that the model state does not vary significantly in a four hour time interval which is true in our case as we are using global GEOS-Chem model with $4^\circ \times 5^\circ$ resolution."

As requested in the comment, the model time step at $4^\circ \times 5^\circ$ resolution consists of 60 minutes for chemistry/emission/deposition and 30 minutes for rest of the science processes.

P22262/L17-20: This sentence is poorly written and partly wrong. It is stated in the first part of the sentence that: 'Contrary to ... at the initial time and'. It is not exactly true to say that 4D-Var analyses are produced only at the initial time. It would be more appropriate to write that 4D-Var produces new initial conditions and that these initial conditions can be used by the model to get the analyses at any time within the assimilation window. In the second part of the sentence, it is written: 'and accounts ... in the assimilation window'. In fact, this is the case for the three methods. What makes the difference between sequential and variational methods is more likely the fact that sequential methods suppose that the atmospheric state is static during the assimilation window, while in variational methods, a model is used to make the time interpolation of the model initial condition at the observation time.

Answer: The whole sentence 'Contrary to ... and accounts ...' changed to '*Unlike 3D-Var and suboptimal KF, where analysis states are generated sequentially every observation window, 4D-Var produces new initial conditions that could be used by the model to generate analysis at any time during the assimilation window.*'

P22262/title of sec 5.2: By 'Specification of error variances', I understand the error variance of the background and observation. This section only discusses the background errors. Accordingly, the title must be clarified.

Answer: 'background' added to the title to make it 'Specification of background error variances'.

P22262-22263/1 of sec 5.2: Do you mean that the background variances

are set identical for each model level and set as 50% of the mean ozone field at that level taken from the initial guess? If so, this can be shortened.

Answer: Correct. However, we wanted to show the similarities between background error variances of variational and KF case in our study. It also provides a clear structure of the matrices to readers who would like to implement something similar for their research.

P22264/L7: I suppose that the vertical correlations are also neglected. If so, replace 'horizontal correlations' by 'spatial correlations'.

Answer: Changed as suggested.

P22266/L11: Do you mean 'We first consider a comparison between the analyses for the case where the 4-D-Var assimilation is based on an assimilation window of five days.'?

Answer: With the clarification provided in a previous comment, this should be fine.

P22265/L15-16. Can you specify if the 4D-Var system requires a huge CPU time or memory (or both)?

Answer: 4D-Var requires both higher CPU time and disk memory. Table 1 already showcases higher CPU time for 4D-Var. The following text has been added in Section 6.1 to provide memory estimates:

"*In our study, a full adjoint run for one simulation day requires about 12 GB of hard drive storage. This consumption could be reduced by almost 50% if rather than saving intermediate concentrations of chemical integration, they are recalculated by calling forward chemistry in the adjoint run, which would eventually lead to higher computational time.*"

P22265/L16-19: Model and adjoint runs require an substantial CPU time. Is the storage of the model history an issue? If not, the sentence could be terminated by 'forward and adjoint model runs'.

Answer: Refer to previous comment.

P22267/L22: as far as I understand the sentence, I would replace 'ozone distribution' by 'ozone variability'.

Answer: Changed as suggested.

P22267/L26/27: '... at the end of the assimilation window'. See my general comments.

Answer: Part of sentence changed from '5 days run' to '5 days assimilation window'.

P22268/L26: Do you mean 'We next consider comparing the analyses produced by the 4-D-Var assimilation method with an assimilation window of two weeks.'?

Answer: With the clarification provided in a previous comment, this should be fine.

P22271/L17-20: Why 'in principle'? If your 4D-Var system based on KPP, the backward integration of the model (using the adjoint of the KPP chemical solver) will provide information on the non-observed species if they are chemically coupled to the observed species. Is it not the case in your 4D-Var formulation?

Answer: Correct. Removed "In principle".

P22272/L5-10: what you call the 'adjoint sensitivities' seems to be the gradient of the cost function for a given species. Is that right? If so, use this terminology instead.

Answer: 'Adjoint sensitivity' is indeed the gradient of the cost function. However, this terminology is widely accepted in the scientific community [Sandu et al., 2003a; Daescu, 2003; Li and Petzold, 2004; Castaings et al., 2007; Hakami et al., 2007; Henze et al., 2007].

P22272/L5-10: In the case where KPP is the chemical solver, it is surprising that the system does not also reduce the gradient for these unobserved species. Moreover, the best way of measuring the influence of observed species on non-observed species is to use the influence function introduced by Fisher and Lary (1995).

Answer: As described in the article and answers to previous comments, KPP is the chemical solver integrated with GEOS-Chem to carry out 4D-Var assimilations. See the other response pertinent to the influence functions for an answer to the second point.

Typos etc

P22251/L7: typo: ‘implementations’, ‘performances’ with ‘s’.
P22251/L8: Replace ‘,’ by ‘.’ in ‘interest, A’.
P22251/L16: Replace ‘tropospheric’ by ‘stratospheric’ as Geer et al. is mainly concerned by the stratosphere.
P22252/L16-18: replace ‘by minimizing’ by ‘that minimize’. Add also at the end of the sentence ‘... and taking into account the error covariances of the observations and the forecast.’
P22253/L15: replace ‘state space’ by ‘model state vector’.
P22255/L15: I guess $\mathbf{H}$ must be replaced by $\mathcal{H}$.
P22265/L4: replace by ‘..., we set the KPP parameters RTOL=...’.
P22265/L28: add a ‘,’ between ‘.html’ and ‘Thompson’.
P22266/L7-10: A verb is missing in the sentence ‘Forecast scoring ...’.
P22267/L16: replace ‘(Nassar et al., 2008)’ by ‘Nassar et al. (2008)’.
P22269/L9: ‘4-D-Var still provides’ and not ‘provided’
P22269/L11: ‘it performs well’ instead of ‘performed’
P22269/L11: remove ‘as well’.
P22270/L3-6: the word ‘case’ is used too often in this sentence.
P22270/L7-10: The sentence is too long, which makes it unclear. Please rewrite.
P22272/L2: replace ‘Sandu et al. (2005a)’ by ‘(Sandu et al., 2005a)’
P22272/L26: ‘comparison’ is repeated twice.

Answer: All the suggested corrections made.

References

Castaings, W., Dartus, D., Le Dimet, F.-X., and Saulnier, G.-M.: Sensitivity analysis and parameter estimation for the distributed modeling of infiltration excess overland flow, J. Hydrol. Earth Syst. Sci. Discuss., 4, 363405, 2007.

Courtier, P., Thepaut, J.-N. and Hollingsworth, A.: A strategy for operational implementation of 4D-Var, using an incremental approach. Q. J. R. Meteorol. Soc., 120, 13671387, 1994.

Daescu, D., Carmichael, G.R., and Sandu, A.: Adjoint Implementation of Rosenbrock Methods Applied to Variational Data Assimilation Problems, J. Comp. Phys, 165, 496-510, 2000.

Daescu, D., Sandu, A., and Carmichael, G.R.: Direct and Adjoint Sensitivity Analysis of Chemical Kinetic Systems with KPP: II - Validation and Numerical Experiments, Atmos. Environ., 37, 5097-5114, 2003.

Damian, V., Sandu, A., Damian, M., Potra, F., and Carmichael,G.R.: The Kinetic PreProcessor KPP - A Software Environment for Solving Chemical Kinetics, Comp. and Chem. Eng., 26, 11, 1567-1579, 2002.

Eller, P., Singh, K., Sandu, A., Bowman, K. W., Henze, D. K. and Lee, M.: Implementation and evaluation of an array of chemical solvers in a global chemical transport model, Geophysical Model Development, Vol. 2, p. 1–7, 2009.

Hakami A., Henze D.K., Seinfeld J.H., et al.: The adjoint of CMAQ. Environmental Science and Technology, 41(22):7807–7817, 2007.

Henze, D. K., Hakami, A. and Seinfeld, J. H.: Development of the adjoint of GEOS-Chem, Atmos. Chem. Phys., 7, 2413-2433, 2007.

Lawless., A. S., Gratton, S., and Nichols, N. K.: An investigation of incremental 4D-Var using non-tangent linear models, Q. J. R. Meteorol. Soc., 131, pp. 459476, 2005.

Li, S., Petzol, L.: Adjoint sensitivity analysis for time-dependent partial differential equations with adaptive mesh refinement, J. Comp. Phys., 198, 310325, 2004.

McLinden, C. A., Olsen, S. C., Hannegan, B., Wild, O., Prather, M. J., and Sundet, J.: Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, J. Geophys. Res., 105(D11), 14,65314,665, doi:10.1029/2000JD900124, 2000.

Parrington, M., Jones, D. B. A., Bowman, K. W., Horowitz, L. W., Thompson, A. M., Tarasick, D. W. and Witte, J. C.: Estimating the summertime tropospheric ozone distribution over North America through assimilation of observations from the Tropospheric Emission Spectrometer, Journal of Geophysical Research, Vol 113, D18307, doi:10.1029/2007JD009341, 2008.

Sandu, A., Daescu, D., and Carmichael, G.R.: Direct and Adjoint Sensitivity Analysis of Chemical Kinetic Systems with KPP: I - Theory and Software Tools, Atmos. Environ., 37(36):5083-5096, 2003.

Daescu D.N., Sandu A., Carmichael G.R.: Direct and adjoint sensitivity analysis of chemical kinetic systems with KPP: II - Numerical validation and applications, Atmos. Environ., 37(36):5097–5114, 2003.

Singh, K., Jardak, M., Sandu, A., Bowman, K. W., Jones, D. B. A., Lee, M.: Construction of non-diagonal background error covariance matrices in global chemical data assimilation, Geophysical Model Development, 4, 299-316, doi:10.5194/gmd-4-299-2011, 2011.