Reply to Reviewer #2

We appreciate the comments left by the reviewer, which have helped to improve the paper and clarify the details of our model. Our point-by-point response to reviewer #2 is included below.

RC – Reviewer comments
AC – Author's comments

1. Overview

The manuscript "Development of a variational flux inversion system (INVICAT v1.0) within the TOMCAT chemical transport model" describes the construction of a new 4D-Var data assimilation system for inverse modeling of different atmospheric tracers. Two adjoint tests were performed to validate the newly ATOMCAT adjoint model. Finally the variational system INVICAT accurately reproduced a set of surface fluxes in a twin experiment framework using SF6 pseudo-observations.
Overall the manuscript is well written but several issues need to be addressed:

- **The authors must replace the standard 3D-Var equations with the ones valid for 4D-Var approach. Consequently the observation term inside the cost function should include a summation reflecting observations at different times and this change should be reflected also in the cost function gradient formula. Thus a consistent part of Section 3 must be rewritten clarifying also some confusion aspects regarding TOMCAT model and the atmospheric transport component.**
- **A third adjoint test should be performed and the results should be plotted. This is called alpha test (see Navon et al. [6]) and gives the readers a chance to graphically notice the adjoint model accuracy. The authors must compute the expression $E(\alpha) = [J(x+ \alpha\delta x)-J(x)]/\alpha\delta x\nabla J(x)$. For values of α that are small but not too close to the machine zero, one should expect to obtain a value of $E(\alpha)$ that is close to 1. A figure depicting the values of E with respect to α is desired.**

I recommend the publication of the present manuscript with major revision.

AC: Thank you for these comments. We have implemented both of these major corrections that you have suggested: we have rewritten the equations and the revised document includes results from the 'alpha test'. We have also made further changes according to the specific points detailed below.

2. Specific Comments

RC: The authors should revise the title of the manuscript since the data assimilation system INVICAT is making use of the forward and adjoint TOMCAT models and consequently INVICAT is not a part of TOMCAT. A more suitable title would be "Development of a variational flux inversion system (INVICAT v1.0) using the TOMCAT chemical transport model".

AC: You are right. We have changed the title of the paper to *"Development of a variational flux inversion system (INVICAT v1.0) using the TOMCAT chemical transport model".*

RC: 7118.25 The phrase starting with "In order to accurately model .." is too long and the authors should split it in two sentences.

AC: We have split this sentence in two.

RC: 7119.10 Remove "an" before "atmospheric species".

AC: Done.

RC: 7120.15 The authors should mention what type of 4D-Var approach did use to build the INVICAT variational system. Is it the incremental method proposed by Courtier et al. [2] or the standard 4D-Var introduced in meteorology by Le-Dimet and Talagrand [5].

AC: We have mentioned the fact that we are using the standard 4D-Var methodology within the text and included the appropriate reference.

RC: 7120.20 Replace "similar that" with "similar to that".

AC: Done.

RC: 7120.20 NWP data assimilation schemes employ variational approaches so the sentence makes no sense.

AC: We have clarified this point in the text; we did not intend to infer that NWP data assimilation schemes are not 'variational', merely that our inversion scheme is not 'true' 4D-Var, but is variational in nature.

RC: 7120.20 In the abstract the authors describe the construction of a new adjoint model. Why here it is referred as something already existing?

AC: We have clarified here that the adjoint model is also newly developed.

RC: 7121.10 "Earlier references" should be removed since it creates confusion.

AC: Done.

RC: 7121.15 The phrase starting with "The variational inverse modelling .." is too long and need to be revised. Perhaps splitting it is a good idea.

AC: This sentence has been split.

RC: 7122.10 As already mention above, the authors should cite the work of Courtier et al. [2] where low resolution linearized model replaces the high fidelity non-linear model during the optimization process. Also cite the work of Stefanescu et al. [10] and Amsallem et al. [1] where non-linear reduced models are used as constrains instead of the full non-linear models. Both approaches are capable to alleviate the computational burden of data assimilation experiments.

AC: We have now cited the work described above in this section of the text.

RC: 7123.5 Remove "an' before "atmospheric species".

AC: Done

RC: 7123.10 Split into two parts the phrase starting with "The state vector" one detailing on the control variables and the other one describing the temporal and spatial resolutions.

AC: Done

RC: 7123.15 If the authors refer to M as a matrix than the atmospheric transport model in the TOMCAT model is linear. Is it really the case?

AC: Yes, this is in fact true. We have added text to the paper to clarify this point.

RC: 7123.15 Since this is a 4D-Var data assimilation strategy the observation term should include a summation reflecting observations at different times.

AC: You are correct, and this has been added to all necessary equations.

RC: 7123.20 What is the nature of the observation operator H? Is it linear or non-linear? If the observation operator role is only to interpolate from the modelled atmospheric concentrations to the observation space than the state variables include also the atmospheric concentrations. I think the authors should clarify this in the text and describe the state and control variables.

AC: H is a linear interpolation operator from the 3D model field onto the observation space. The state vector does include the initial 3D atmospheric concentration field, and text has been included to make this clear.

RC: 7124.5 The observational term in equation (4) should be rewrite according to 4D-Var. Otherwise the variational method considered here is 3D-Var

AC: Done.

RC: 7124.15 The contribution of the observational term to the cost function gradient in equation (5) should be modified in order to reflect the 4D-Var algorithm.

AC: Done.

RC: 7124.20 Here the authors mention that the adjoint version of the TOMCAT model is equivalent to the transpose of the matrix M. If this is correct this means that the TOMCAT model is the same as the atmospheric transport model M which is linear. This is hard to believe since the chemical transport models deal with very high nonlinearities. The authors should clearly explain what is the difference between TOMCAT and atmospheric transport model M. Enumerate also the models variables. I also think that it is more difficult to compute the entire adjoint M* that only a part of it M^T. Thus the entire 7124.15 and 7124.20 should be rewritten.

AC: The transport section of the TOMCAT model is in fact linear, and so *M* and *M'* are equivalent, the matrix **M** does indeed represent the full transport section of the forward model, and the adjoint matrix **M*** represents the adjoint of *M* (or *M'*). We have included text emphasizing that the matrices are representing the transport section of TOMCAT, which includes no nonlinearities.

RC: 7125.25 The authors should clarify what are the models used for computing the gradient $\nabla J(x)$. If the authors employ the TOMCAT and the new proposed adjoint model than the equations (1,4,5) should be written accordingly using M and M* notations.

AC: Done.

RC: 7125.5 The authors should cite the pioneer work on LBFGS optimization approach such as Perry [8], Shanno [9], Nocedal [7].

AC: We have now cited these important references.

RC: 7125.20 The phrase starting with "Assuming that ..." should be rewritten since the tangent linear model is just a first order approximation of the first derivative of the operator M.

AC: We have rewritten this sentence to reflect this fact.

RC: 7126.20 Remove "which remains a possibility for the discrete adjoint".

AC: Done.

RC: 7127.5 It would be nice if the authors can comment (outside the manuscript) on automatic differentiation software performances for generating tangent linear and adjoint models for non-linear Newton based discrete models.

AC: Since we have no experience of automatic differentiation software and the transport in our model is linear, we feel that we cannot comment adequately on this subject. As the reviewer notes, this is not an issue for our paper.

RC: 7127.15 The authors should also refer to WRFDA variational data assimilation system. It makes use of an adjoint model and it is employed to generate atmospheric analysis (see Huang et al. [4]).

AC: Done.

RC: 7128.10 Another way to incorporate model errors is to introduce an additional term in the cost function as in the weak constraint 4D-Var. While for the present manuscript the authors investigates the performance of the variational system using an idealized case for the future ones real SF6 observations should be used. Consequently in addition to representativeness errors the authors should also consider measurements errors inside R. Do you have any idea how to quantify these measurements errors for SF6 observations?

AC: We are aware that assessment of measurement error is an important part of inverse modelling, and that we will have to quantify these errors for an inversion using real observations. Whilst we do not currently have plans to perform inversions with SF6 observations in the future, other species such as CH4, CO and CO2 will be included in the inverse model. We will initially include simple 'repeatability' accuracy for in-situ observations (around 3 parts per billion for CH4, for example), whilst satellite observations include individual error estimates for each observation, which may be used. An extra level of sophistication may be included in future through improved representation of model errors, such as in Ganesan et al., (2014) and others.

RC: 7129.10 What do you mean by it "performed well"? This particular text should be removed since it is not a rigorous statement.

AC: This sentence has been rephrased in order to better describe the performance of TOMCAT within the TransCom experiment.

"The TOMCAT model previously submitted the results of long-term SF6 simulations to the TransCom CH4 intercomparison project (Patra et al., 2011), where it captured the seasonal cycle of SF6 at three ground-based observation sites to a high level of statistical significance, and reproduced the interhemispheric gradient of SF6 to within 0.05 parts per trillion (ppt)."

RC: 7129.15-20 It is not straightforward what is the space resolution of the TransCom initial conditions. Did you need interpolation from fine to coarse resolutions?

AC: We linearly interpolated the initial conditions provided by TransCom from a 2.8 x 2.8 degree horizontal grid with 23 vertical levels to the coarser 5.6 x 5.6 degree, 31-level TOMCAT grid. We have added text to clarify this. However, since the model simulation begins in 1988, we feel that there has been adequate spin-up time so that the initial conditions do not significantly influence our results.

RC: 7129.25 The authors should define "ppt" first before using it.

AC: Done.

RC: 7129.20-25 Rephrase the sentence starting with "The model replicates ...".

AC: We have split this sentence in two and rephrased it. We have now used more quantitative measures of the model performance in this sentence than before.

RC: 7132.20 Why do you need a new version of TOMCAT model? It is enough only to store the forward variables required by the adjoint model (this is called checkpointing - see Griewank and Walther [3]). If your model is linear you don't need checkpointing.

AC: This was indeed unclear and we have attempted to clarify these details. Firstly, we have now explicitly used the term 'checkpointing' in Section 3.1, before Equation (9), to describe the process of saving model data for use with the adjoint model. Secondly, we have explained in the first paragraph of Section 5 that since the forward model is linear, we do not need to 'checkpoint' forward model concentrations in order to run the adjoint model. However, we do save variables that are used the forward model, such as convective flux rates, for use in the adjoint model as this is quicker than re-calculating them..

RC: 7133.5 Another way to test the accuracy of the adjoint model is to compute $E(\alpha) = [J(x+ \alpha\delta x)-J(x)]/\alpha\delta x\nabla J(x)$:
For values of α that are small but not too close to the machine zero, one should expect to obtain a value of $E(\alpha)$ that is close to 1. The adjoint model helps to compute the gradient justifying the usefulness of the test. This linearity test is derived from the alpha test described by Navon et al. [6]. It would be nice to introduce a figure describing the results of this test.

AC: We have carried out this new 'alpha test' for ATOMCAT. We have cited the work of Navon et al., and included a new figure showing the model performance for different values of α.

RC: 7134.5 I think that only one iteration is not enough to accurately test the adjoint subroutines and longer experiments are required.

AC: We have now extended number of iterations carried out during the equality test for the full model (rather than for individual subroutines), from one iteration (one model hour) to 480 iterations (20 model days). ATOMCAT still passes this test with an extremely high level of accuracy (equal to machine precision).

RC: 7136.25 The phrase starting with "A quasi-Newtonian ..." is too long and needs to be splitted.

AC: This sentence has been split in two.

RC: 7137.5 (7136.25) Define MLISO.

AC: We have now defined the line-search program MLIS0.

RC: 7140.10 In 7124.25 the authors mentioned that the adjoint version of the entire TOMCAT model was written. However this contradicts the first sentence of Summary section which states that only adjoint version of the transport section of the TOMCAT CTM was created.

AC: You are correct, this was a little unclear. So far, we have indeed only created the adjoint for the transport section of TOMCAT, and we have changed the text in 7124.25 in order to reflect this.

[1] A. L. Ganesan, M. Rigby, A. Zammit-Mangion, A. J. Manning, R. G. Prinn, P. J. Fraser, C. M. Harth, K.-R. Kim, P. B. Krummel, S. Li, J. Mühle, S. J. O'Doherty, S. Park, P. K. Salameh, L. P. Steele, and R. F. Weiss, Characterization of uncertainties in atmospheric trace gas

inversions using hierarchical Bayesian methods, Atmos. Chem. Phys., 14, 3855–3864, 2014.