Anonymous Referee #1
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We wish to thank the referee for his/her helpful comments. The full reviews are copied hereafter and our responses are inserted. The comments of the reviewer are in normal black and our answers in bold.

This paper describes a Bayesian inverse modeling system for reactive compounds, PYVAR-CHIMERE. It also provides an illustration of what this system can do with two one-day inversions of emissions over Europe. The paper is generally well written, and the topic of the paper is relevant for this journal. Although the results indicate that the system has potential, there are several major issues which should be dealt with before this work can be published in GMD.

1) poor quality of the figures and equations. On the screen, it is more or less acceptable (with the zoom in feature of my pdf viewer), but upon printing, many figures (especially Figs. 1-4 and Figs. 6a and 8a) and equations are impossible to read.
We apologize, the poor quality in particular of the equations was due to conversion from OpenOffice to pdf. The resolution of the equations and of the figures has been improved.

Figure 3 is really a Table and should be inserted as such.
We agree, Figure 3 is now the Table 1.

Some mathematical symbols (e.g. gradient on line 158, multiplication on line 167) are inappropriate.
These symbols have been corrected.

2) Section 3.2 is 'Development of the adjoint of CHIMERE'. But the adjoint of CHIMERE was developed a long time ago (publications by L. Menut, I. Pison). What are the specific developments realized for this study, besides the minor changes to CHIMERE mentioned in the text?
Indeed, the adjoint of CHIMERE was developed a long time ago. We first have changed the title section into "Development and parallelization of the adjoint and tangent-linear codes of CHIMERE".

We added sentences in Section 3.1: "PYVAR has been adapted to CHIMERE with an adjoint code without chemistry a first time by Broquet et al. [2011]. In order to couple PYVAR to the new state-of-the-art version of CHIMERE (see Section 3.2), to include chemistry, and to increase its modularity, flexibility and clarity, the new system described here has been developed. It includes elements of the inversion system (coded in Fortran90) of [Pison et al., 2007]."

Efforts have indeed been made for the parallelization of the code. This is now explained in the text in Section 3.2: "Then, it has been parallelized at LSCE and LISA. This work required a redesigning of the whole code, associated with a full testing scheme. Furthermore, the tangent-linear (TL) code has been developed and validated at LSCE. Changes have been implemented in the forward CHIMERE code embedded in PYVAR-CHIMERE to match requirements of the studies lead with PYVAR-CHIMERE. These changes have been implemented in both the adjoint and the TL codes. Compared to the CHIMERE 2013 version [Menut et al., 2013], the most important of these changes are:
•For the geometry, the possibility of polar domains and the use of the coordinates of the corners of the cells instead of only the centers
•For the transport, the non-uniform Van Leer transport scheme on the horizontal,

•For chemistry, various switches have been added to avoid going into the chemistry, deposition and wet deposition routines when no species requires them (e.g. no chemistry for methane at a regional scale).”

3) the results of inverse modeling studies are very dependent on the inversion setup, in particular the definition of the control vector and the construction of the covariance matrices R and especially B. What is the strategy in this regard? I understand that the main purpose of the paper is less to present specific inversion studies than to describe the general modeling framework. But the very simplistic choices made for the two one-day inversions suggest an absence of any strategy. The chemical lifetimes of the target species, the duration of the experiments, the initial and boundary conditions, and the assumed a priori uncertainties should all play a role in the inverse setup definition.

Indeed, we agree, the main purpose of a GMD paper is less to present specific inversion studies than to describe the general modeling framework. Nevertheless, we added information to explain our choices of illustrations in the introduction of Section 4: "We have chosen to present illustration of CO inversion over a 7-day window, the first week of March 2015. Considering the short lifetime of NO_x of a few hours [Valin et al., 2013; Liu et al., 2016], we have chosen to present illustration of NO_x inversion over a 1-day window, the 19th February 2015. These particular periods have been chosen as they present a representative number of super-observations during winter, and as the emissions are high during that period."

We also have added a new "Section 4.2.2. Covariance matrices B and R" to better describe the covariance matrices B and R.

4) the two illustrations of PYVAR-CHIMERE capabilities are unconvincing. Yes, the system finds a minimum to the cost function, and the a posteriori simulation matches the observations quite well; but no, the a posteriori emissions are not shown to be closer to reality. With its long lifetime, CO is largely determined by the initial and lateral boundary conditions, which are part of the control state vector being optimized. The paper does not provide information on the a priori uncertainties for these parameters. A discussion is needed, and possibly sensitivity simulations.

We now provide more information about the initial, lateral and top boundary conditions in the new "Section 4.1.2. CHIMERE set-up": "Different climatological values from the LMDZ-INCA global model [Szopa et al., 2008] or from a MACC reanalysis are used to prescribe concentrations at the lateral and top boundaries and the initial atmospheric composition in the domain."

Information on the prior uncertainties was given in the former Figure 3, now replaced by Table 1. We also added sensitivity tests in Section 4.1.3: “With its lifetime of about two months, CO could be strongly driven by the initial and lateral boundary conditions prescribed in the CTM. In fact, as seen in Figure 4b, initial and boundary conditions provide a relatively flat background and the patterns which appear clearly over the background are linked to surface emissions (Figure 4a). To characterize the uncertainties in the concentration fields due to the initial and lateral boundary conditions, we performed a sensitivity test by using either climatological values from LMDZ-INCA or a MACC reanalysis: the results were not significantly different, with relative differences in concentrations of less than 15% over continental land (Figure 5c).”

We also added text in the new "Section 4.2.2. Covariance matrices B and R": "Based on the sensitivity test in Figure 5, the errors assigned to the CO lateral and top boundary conditions and to their initial conditions are set at 15%. As these relative errors are significantly lower than those for the emissions and as variations in the CO surface concentrations are mainly

driven by emissions (Figure 4), we assume a small relative influence of the correction of initial and boundary conditions on our results. "

Note that, although the a priori simulation overestimates CO over Central Europe (south of Poland), the inversion increases the emissions there by about a factor of 2! Over Germany, the emissions are almost doubled in the Southern part, but are unchanged elsewhere. How can this be justified?
Figure 6 has been updated. The emissions are increased over Central and Eastern Europe, except in the south of Poland.

Even for NOx, in spite of their shorter lifetime, the initial and boundary conditions play probably a very important role.
We also checked the impact of initial and boundary conditions on NO_2 tropospheric columns. Due to its short lifetime, the impact is even smaller than for CO. We chose not to show this sensitivity test in the paper.

The discussion of the results for NOx (lines 373-376) is impossible to understand. It says that the optimization of NOx fluxes has only a small impact on the model biases. This is not true. Comparison of Fig. 7c and 7d show that the optimization works very well!
We do not agree, as Fig 7c and Fig 7d were not comparable (they did not have the same legend). Nevertheless, we agree, it could have been confusing. We now present the impact of the optimization of the NO_x fluxes differently, in Figure 7.

The authors claim that PYVAR optimizes only the NO2 fluxes, not those of NO. I don't believe this, it doesn't make sense. Please check this. In any case, clarifications and possibly a sensitivity analysis are in order.
As both the reviewers have been disturbed by the illustration with only NO_2 fluxes, we now present inversion for NO_x emissions.

Other comments:
- throughout the text, replace "NO2 emissions" by "NOx emissions" (if indeed, as should be the case, NOx emissions are optimized, not just NO2)
See comments above. "NO_2 emissions" have indeed been replaced by "NO_x" emissions throughout the text.

- lines 107-110: please refer also to GEOS-Chem adjoint papers (Henze,Kopacz, Caoetc.)
A reference to Henze et al. 2007 has been added. A reference to the adjoint-based four-dimensional variational (4D-Var) assimilation system, WRF-CO2 4D-Var, has also been added.

- l. 149 what is meant by "the control of emissions"?
We have rephrased: "By definition, the observation errors combine errors in both the data and the observation operator, in particular measurement errors and errors in the conversion of satellite measurement into concentration data, errors from the CTM, representativity errors due to the comparison between point measurements and gridded models or due to the representation of the fluxes as gridded maps at a given spatial resolution, and aggregation errors associated with the optimization of emissions at a given spatial and/or temporal resolution (as specified in the control vector) that is different from (usually coarser than) that of the CTM [Wang et al., 2017]."

-l. 168-169 What are the thresholds for the ratio between final and initial gradient norm, and for the number of iterations?

This information was already given in Section 4. It is now in the introduction of Section 4: "For practical purposes, we recommend to reduce the norm of the gradient of *J* by 90%. We no longer give information about the number of iterations as it depends on each configuration system. These information are now given in Section 4.2.3 for CO: " Ten iterations are needed to reduce the norm of the gradient of *J* by 90% with the minimization algorithm M1QN3" and in Section 4.2.4 for NO_x: "Six iterations are needed to reduce the norm of the gradient of *J* by 85% with the minimization algorithm M1QN3".

-Figure 1. In the orange box on the right, the order of operators should go backwards, shouldn't they?
Indeed, the adjoint go backwards but these operations are made simultaneously in CHIMERE.

- l. 195 leads (instead of lead)
It has been changed.

-Figure 3. Explain the meaning of the "correction type" and of the three numbers in column "B variance coefficients".
The correction type describes the way the emissions are corrected by the inversion. We have added the following descriptions in "Section 3.3. Definition of the control vector": "Several types of corrections can be applied, they are defined in the code as "add", "mult" or "scale". Both the corrections "add" and "mult" are applied to gridded control variables. For correction type "add" the control variables are increments added to the corresponding components of the model inputs. For correction type "mult", the control variables are scaling factors multiplying the corresponding components of the model inputs. The difference between the two options "add" and "mult" plays a role when inverting fluxes which can switch from positive to negative values (like CO_2 natural fluxes). For type "scale", the corrections consist in applying scaling factors to activity maps and/or masks for regions (which is similar to the control of budgets for different regions, types of activities, and/or processes in inversions where the control vector is not gridded [Wang et al., 2018]) and adding the obtained values to the corresponding components of the model inputs."

The three numbers in column "B variance coefficients" are standard deviation coefficient. We have added information: "The variances are specified by the user through standard deviation coefficient (Table 1), which can be a fixed value ("fx") or a percentage ("pc") to define the diagonal standard deviation matrix $\sum$."

- Figure 4. The legends mentions text in blue and in grey. I don't see that on the figure. Is this figure useful?
The sentence mentioning text in blue and in grey has been removed.

- l. 298 What is the resolution of ECMWF data? Are those data interpolated to the model grid?
The spatial resolution of the ECMWF data is 0.25°x0.25°. They have been interpolated to the model grid.

-l. 300 Derognat et al. 2003 does not present a chemical mechanism, but refers to earlier papers.
The reference to Derognat has been removed. We now refer to Lattuati, 1997 and to the lastest CHIMERE documentation. The text is now:"The chemical scheme used in PYVAR-CHIMERE is MELCHIOR-2, with more than 100 reactions [Lattuati, 1997; CHIMERE 2017], including 24 for inorganic chemistry".

-What are the a priori lateral boundary conditions?

The prior lateral and top boundary conditions are climatological values from the LMDZ-INCA global model [Szopa et al., 2008]. It was indicated in Section 4.1.1. We now describe them in "Section 4.1.2 CHIMERE set-up" and we also made a sensitivity test with a MACC reanalysis in "Section 4.1.3. Sensitivity to emissions and to initial and boundary conditions".

- on l. 303 and legend of Fig. 8, the information required to run the inversion are said to be listed in Table 1. This is not correct.
We agree, this is now true (Figure 3 is now called Table 1).

- l. 304 Are only anthropogenic emissions optimized? Or the total of all emissions?
Only the anthropogenic emissions are optimized here. This is now written in the introduction of Section 4: "The potential of the PYVAR-CHIMERE system to invert emissions of reactive species is illustrated with the inversion of CO and NO_x anthropogenic emissions in Europe respectively based on MOPITT CO data and OMI NO_2 data".

This is also now written in the description of the control vectors in Section 4.2.1.

-l. 306-307 The 3D initial conditions at the model resolution are said to represent 8585components of the control vector. But what about the vertical dependence?
The vertical dependence is indeed taken into account. The number of components in the control vector has been corrected.

-l. 311-314 What are the non-anthropogenic emissions used in the model?
The biogenic emissions, that we assume negligible in winter, are not used in our illustration. In addition, we should have described that PYVAR-CHIMERE only infers anthropogenic emissions at this stage. This is now added in Section3.2 : "It should also be noted that PYVAR-CHIMERE only infer anthropogenic emissions at this stage. The optimization of biogenic emissions, which are linearly interpolated at the sub-hourly scale in CHIMERE, is currently under development."

-Please provide a webpage and reference for EMEP emissions. Aren't there any publication or webpage for the TNO emissions?
We now provide two references for the EMEP emissions (a publication and a webpage). We also provide a reference for the TNO-GHGco used in this paper (submitted in December 2019 and published in February 2020). The text is now: "The prior anthropogenic emissions for CO and NO_x emissions come from the TNO-GCHco-v1inventory [Super et al., 2020], the last update of the TNO-MACCII inventory [Kuenen et al., 2014]. The prior anthropogenic emissions for VOCs come from the EMEP inventory [Vestreng et al., 2005; EMEP/CEIP website]."

-l. 328 Why the median?
The median is chosen here to take proper account of the AKs (we can not take the mean of the AKs).

-l. 341 With errors of 100% on the emissions, how can negative a posteriori emissions be avoided? How is this dealt with?
We now answered to this remark in "Section 4.2.2. Covariance matrices B and R": "With such a set-up, in theory, we could obtained negative posterior emissions since the inversion system does not impose a constraint of positivity in the results. Nevertheless, even 100% of uncertainty lead to a prior distribution mostly (>80%) on the positive side. The assimilation of data showing an increase above the background (at the edges of the domain; not shown)

further drive the inversion towards positive emissions for both CO and NO_x inversions. In practice, our inversion does not lead to negative posterior emissions (Figure 7b). Spatial and temporal correlations in B would further limit the probability to get negative emissions locally by smoothing the posterior emissions at a spatial scale at which the "aggregated" prior uncertainty is smaller than 100%. However, a positivity constraint should be implemented in future versions of the system."

-Section 4.2 The Figure 5 shows both underestimations and overestimations by the apriori simulation. This is not well reflected in the discussion.
Indeed, this is now reflected in the discussion in Section 4.2.3.

-Figure 7c and 7d should show absolute differences.
We no longer present these figures.

-A better color scale should be possible for Fig. 7a and 7c
Indeed, it has been done in Figure 8 and in Figure 10.