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Interactive comment on "Estimation of fossil-fuel CO_2 emissions using satellite measurements of "proxy" species" *by* Igor B. Konovalov et al.

Igor B. Konovalov et al.

konov@appl.sci-nnov.ru

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We thank the Referee for the positive evaluation of our manuscript and for the thoughtful comments and remarks. All of the referee's comments have been carefully addressed in the revised manuscript. Below we describe our point-to-point responses to the referee's comments.

Referee's comment: P2, L14-15: This statement needs a bit more explanation, the 2014 emissions will be the starting point for what?

We meant that the 2014 emissions would be the starting point for global and national emission reduction plans. However, this sentence is not quite necessary in the given context and is removed from the revised version of our manuscript.

Referee's comment: P3, L1: Technically speaking, the emissions (or emission parameters, depending on which is being optimized) are not in the transport model but are coupled to it.

We agree with this remark. The corresponding sentence is corrected accordingly.

Referee's comment: Section 2.2: *The authors do not mention what the boundary conditions were for other species used in the MELCHIOR scheme that react NO_2 and CO. Concentrations of NO_2 are strongly affected by atmospheric chemistry (lifetime of $\sim$1 day) so how important is the correct representation of chemistry on the NO2 simulations and thus in the emissions from the inversion? Similarly for CO, although owing to its longer lifetime this is perhaps not so important. How are the uncertainties in the lifetimes propagated into the emissions found from the inversions?*

It was mentioned in the reviewed manuscript (Sect. 2.2) that initial and boundary conditions for gases and aerosols were specified using monthly climatological data from LMDz-INCA global model. In the revised manuscript (Sect. 2.2), we addressed this point in more detail. In particular, we indicated several concrete species, for which the boundary conditions were specified using the LMDz-INCA data and noted that a full list of such species is provided in the CHIMERE documentation available on the web site www.lmd.polytechnique.fr/chimere. We also noted that influxes of other species, most of which are very reactive and short-lived (such as, e.g., OH and HO_2) into a model domain, are not specified in CHIMERE.

It is true that the correct representation of chemistry in the NO_2 simulations is an important prerequisite for inferring reliable NO_x emission estimates. The effect of uncertainties in simulations of chemical processes on our results was briefly discussed in Sect. 3.4 (P. 17, L. 13-17) of the reviewed manuscript. In particular, we argued that such model errors are likely to differ in different grid cells, and thus they are expected to contribute to the emission estimate uncertainties evaluated using the subsampling technique. In the revised manuscript, the discussion of this important point is improved and extended. In particular, we added that as the CO and NO_2 evolution is governed by essentially different chemical processes, uncertainties due to a "chemical" part of model errors are likely to be manifested in differences between the CO_2 emission estimates based on the NO_2 and CO measurements. But we also noted that it is nonetheless not quite infeasible that some model errors associated with the representation of chemical interactions can result in similar (positive or negative) biases across the CO or NO_x emission estimates inferred from the different data subsets, and so we cannot completely ensure that the confidence intervals for our CO and (especially) NO_x emission estimates actually account for all possible model errors. A similar caveat is provided in Sect. 5 (Summary and conclusions) of the revised manuscript.

In fact, the effects of "chemical" part of model errors on inversion results are very difficult to evaluate as the characteristics of such errors are mostly not known and probable errors in specific reactions or in the boundary conditions of certain species may constitute practically an infinite number of error combinations which would propagate into the emissions found from the inversions in different ways. So we believe that more accurate evaluation of the effects of possible errors in the model representation of chemical processes on NO_x and CO emission estimates that can be derived from satellite measurements requires further research going beyond the scope of this study. One of the practical ways to address this issue could involve multi-model inversions that might be performed in the framework of a dedicated project. The results of our study can provide an impetus for such a project.

Referee's comment: P9, L1-2: Why were cement emissions of CO_2 ignored? What is the impact of this if the system would be used in a region where cement emissions are more important?

Emissions from cement production are not considered in our study mostly because cement production, unlike FF burning, is not associated with significant emissions of either NO_x or CO, and so satellite measurements of the corresponding proxy species cannot provide strong constraints on cement emission of CO_2. A corresponding explanation is added in the revised manuscript. Even if cement CO_2 emissions would be more important in the case considered, our estimates of FF emissions would not be directly affected (again because cement production do not contribute to concentrations of the proxy species and corresponding emissions were excluded from our calculations of the conversion factors).

Referee's comment: P9, L1-2: P9, L12-15: Do the authors mean that their motivation for solving for 2 categories of sources (EHI and TCO) is to reduced the aggregation error, because these sources have different temporal/spatial errors? If so, this could be made clearer and stated early, e.g. P9, L3 when the grouping into these categories is first mentioned.

Yes, our primary motivation for defining the two specific categories was to limit aggregation errors, but we expected also that consideration of these two categories would allow us to get more specific information on emission sources. The corresponding paragraph was revised accordingly.

Referee's comment: P12, Eq. 4: How well conditioned is this expression? The authors use no regularization method?

Indeed, we did not use any regularization method (or a priori constraints on the solution). The main reason is that the number of control parameters is very small (one or two) especially if considering the vast amount of atmospheric data which we use. This reason was briefly explained on P. 13 of the revised manuscript, and we tried to improve the corresponding discussion in the revised manuscript. Another reason is that we presume that each control variable is seen with some level of independence by subsets of these data given the geographical distribution of each. The results of our OSSEs based on the Monte Carlo method (see Sect. 3.5) show that the uncertainties in our emission estimates remain rather small in spite of very large uncertainties in the input data (see Sect. 2.4). In our understanding, this fact clearly indicates that the inverse problem considered is not ill-conditioned. This is mentioned in Sect. 3.5. The analysis of the relationship between the CO or NO$_x$ measurements and the emission estimates by using methods of linear algebra was beyond the scope of this complex and time-consuming study (and would be pointless in the important case where the control vector has only one component corresponding to the total NO$_x$ or CO emissions). Application of such methods to the inverse problem considered in our study is complicated due to the fact that the cost function given by Eq. (4) includes the "bias" term that depends on the model data (and thus on the emission estimates) in a rather complex manner (according to Eq. 6).

Referee's comment: P13, L15-16: The condition given, i.e., that the control vector is smaller than the measurement vector is not sufficient. A sufficient condition is rather the condition number of the matrix inverted. One could imagine a case where there are more measurements than unknown variables but where each measurement provides only a weak constraint (or even no constraint) on the unknown variables.

We fully agree with this critical remark. The corresponding text is corrected accordingly. Specifically, instead of the incorrect statement given in the reviewed manuscript, we mention that avoiding formal a priori constraints (or any other regularization) does not *necessary* result in ill- conditioning of an inverse problem, as long as the dimension of the control vector is much smaller than that of the measurement vector. We mention also that although satisfying this criterion alone cannot guarantee that the problem is well-conditioned, the numerical experiments presented below in Sect. 3.5 show that errors in our emission estimates due to probable errors in input data remain limited and thus the results of these experiments indicate that the problem considered in this study

is not ill-conditioned.

Referee's comment: P17, L18-25: Are the results of these 2 cases discussed? They are mentioned here but there is no conclusion given about the uncertainty in the posterior due to potential errors in the seasonal cycle.

Yes, the results of these two cases are discussed in Sect. 4.1 (in the 5th paragraph from the beginning of the section) and in Sect. 4.2 (in the last paragraph).

Referee's comment: P18, L3-7: How independent are the conversion factors among the three inventories? How are the factors determined? Do they rely on independent observations? This is important, as error in the conversion factor will translate directly into error in the CO2 emissions.

The NO$_x$-to-CO$_2$ (or CO-to-CO$_2$) emission conversion factors were calculated in our study as the ratios of the corresponding emission annual totals provided by the emission inventories according to Eq. (7). In Sect. 3.4, we explain that we used two different sets of estimates for the conversion factors, one of which (considered as the main option) is based entirely on the EDGARv4.2 emission inventory, while another is based on the data for CO$_2$ emissions from CDIAC and for NO$_x$ and CO emissions from the EMEP inventory. Therefore, we presume that the first question of the reviewer (*How independent are the conversion factors among the three inventories?)* can be reformulated as follows: are the differences between the CO$_2$ emission estimates given by the CDIAC and EDGARv4.2 inventories and the differences between the NO$_x$ (CO) emission estimates provided by the EDGARv4.2 and EMEP inventories sufficiently representative of uncertainties in the inventory data (as assumed in our study)? Although we cannot provide a mathematically precise answer to this question, the fact is that there are considerable differences in both the data sources and the methodologies used across the three inventories. Specifically, while the fossil fuel burning CO$_2$ emission data provided by CDIAC are based on the energy statistics published by the

United Nations (UN, 2012), the EDGARv4.2 inventory uses energy activity data based on IEA (International Energy Agency) energy balances (IEA, 2012). The UN data used in CDIAC were compiled primarily from the annual energy questionnaire distributed by the United Nations Statistics Division and supplemented by official national statistical publications (UN, 2012), while the IEA data were compiled following harmonised definitions and comparable methodologies across countries and do not necessary represent complete data sets available to national experts (IEA, 2010). Similarly, the methodologies used in CDIAC and EDGARv4.2 to convert fuel consumption into CO_2 emissions have been developed independently and involve significantly different classifications of fuels and different sets of parameters. For example, while the key parameter involved in the EDGARv4.2 inventory is the net caloric value which is used to convert the activity data into the energy values (IEA, 2010) (that are then converted into quantities of carbon), CDIAC converts the quantity of fuel into the quantity of carbon directly by using the carbon content parameter (see Marland and Rotty, 1984 and IPCC, 2006 for more details on the methodologies used in the CDIAC and EDGARv4.2 inventories, respectively). The EMEP/CEIP inventory is based on emission reports provided by the national environmental agencies. Accordingly, compared to the EDGARv.4.2 inventory, the emission data provided by EMEP inventory may better account for statistical information and sources specific for a given country. The set of emission factors which EMEP recommends to use while preparing national emission inventories (EMEP/EEA, 2013) is substantially different from that used in the EDGAR v4.2 inventory (IPCC, 2006), particularly because it involves the different sector definitions. Taking all these differences into account, we believe that it is indeed safe to assume that the two kinds of the conversion factor estimates considered in our study are sufficiently independent. Nonetheless, it is also not quite impossible that, in some hypothetical cases (but hardly in our study region), different inventories can be biased in a similar way due to, e.g., the sources and technologies that are accounted for neither in international nor in national energy data bases. In such cases, the conversion factor uncertainty evaluated with our approach may be underestimated. So, in a general situation, a statistically significant difference between our "hybrid" CO_2 emission estimate and an estimate based on emission inventory data would strongly suggest that the latter is biased, although other, less probable, reasons, such as errors is the conversion factors or systematic uncertainties in the model representation of chemical processes should not be disregarded without special investigation.

Consistently with the above discussion, we mentioned in the revised manuscript (without going into details) that taking into account considerable differences in the data sources and methodologies used across the three inventories, we assume that the main and alternative conversion factor estimates are sufficiently independent. A similar assumption concerning reliability of the confidence intervals is also mentioned in Sect. 4.2. Finally, we remarked in Sect. 5, that further research is needed to ensure that the confidence intervals for our emission estimates actually take into account all possible error sources, including uncertainties in the conversion factors.

Referee's comment: P22, L31: It would help the reader to specify again what the analysis (in section 2.4) is being referred to here, the lower sensitivity of the IASI measurements to CO emissions?

We added an explanation. The corresponding revised sentence reads as follows: "Taking into account our preliminary analysis (see Sect. 2.4) indicating that the contribution of the anthropogenic CO emissions in the study region into the corresponding CO columns is relatively small and the results of the OSSEs (see Sect. 3.5), it is also not surprising that the uncertainties in our CO emission estimates are much larger than those in the NO_x emission estimates."

Referee's comment: *P23, L23: I would suggest either removing "robust" here or rephrasing the sentence to e.g. ". . .uses an approach, which is deemed more robust at the global scale", because the current formulation sounds somewhat contradictory,* *i.e., the EDGARv4.2 inventory is worse than EMEP in Europe but uses a more robust approach.*

The corresponding sentence has been rephrased following the referee's suggestion and reads as follows: "This is an expected result because the methodology used in the EMEP inventory is specific to national statistical data available from European countries, while the EDGAR v4.2 inventory uses another approach which is deemed to be robust at the global scale."

Referee's comment: P25, L24-27: I'm somewhat confused. The conversion factors were determined from the ratios of CO_2:NO_x and CO_2:CO in the inventories, so if the inversion results for NO_x and CO are not significantly different from EDGARv4.2 then how can the result for CO_2 be significantly different from EDGARv4.2?

Actually, the inversion results for NO_x are significantly different from EDGARv4.2, but this was not explicitly stated in the reviewed manuscript. This point is clarified in Sect. 4.1 of the revised manuscript.

The technical comments by referee were carefully considered. The corresponding changes are made in the revised manuscript.

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