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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.

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We are grateful to the Referee for the positive evaluation of our paper and for the useful comments which were carefully addressed in the revised manuscript. Below we describe our point-to-point responses to the referee's comments.

Referee's comment: One major issue seems to be the (lack of) distinction between NO_x emissions and NO_2 observations. It seems that the authors use NO_x emission data (reported "as NO_x", "as N", "as NO_2"?) in conjunction with NO_2 measurements. They should explain how the uncertainty in the NO/NO_2 partitioning (which can change with season and local time) influences their results, and it should be made clear to the reader that the difference between NO_x emissions and NO_2 observations is not problematic in this context (if this is actually the case). This is important, e.g., in the

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discussion of Eq. 7, and on p.21/l.14.

The NO_x emissions are conventionally reported in inventories in grams of NO_2 (although actually most of them are coming in the atmosphere in the form of NO and then are being oxidized to NO_2). We agree that the distinction between NO_x emissions and NO_2 observations (as well as the distinction between the species NO_x and NO_2) was not sufficiently clear in our manuscript. Partly, it was so because we wanted to simplify our discussion, presuming (implicitly) that not only NO_2 columns but also NO_x columns could (in principle) be retrieved from satellite measurements by using (to different extents) modeled data. In the revised version, we tried to clarify this distinction. In particular, we state (in Abstract and Introduction) that one of the proxy species considered in this study is NO_2 (rather than NO_x). Accordingly, NO_x emissions are referred throughout the revised manuscript not as emissions *of* the proxy species, but rather as emissions *for* (or corresponding to) the proxy species. To clarify the role of a chemistry transport model in our study, we note (in Sect. 3.2, first paragraph) that Eq. (3) is used specifically to express the modeled relationships between NO_2 measurements and NO_x emissions, as well as between CO measurements and CO emissions.

The uncertainties in the NO/NO_2 partitioning can indeed influence our results. Due to the very complex nature of this uncertainty (which may, in particular, be due to errors in the chemical reaction rates and in the boundary conditions, as well as due to inaccuracies in the reduced chemical mechanism used in our model), we cannot and did not attempt to evaluate this uncertainty explicitly. However, we expect that particularly because the NO/NO_2 partitioning changes with season and geographical location, its uncertainty is mostly included into the confidence intervals evaluated with the subsampling technique described in Sect. 3.4.

In the revised manuscript, the discussion of possible effects of model errors on our emission estimates is extended. In particular we note (in Sect. 3.4) that it is 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. As an example, we mention that systematic underestimations of the NO_x emissions may be due to persistent positive biases in the ozone formation rate and in boundary conditions for tropospheric ozone concentration (as ozone concentration accounts for partitioning of NO_x between NO and NO_2) as well as due to other numerous factors (such as e.g. underestimation of the hydrocarbon emissions or of the ozone photolysis rate) that may result in underestimation of concentration of hydroxyl radical providing a major sink for NO_x and determining its atmospheric lifetime. We conclude that more accurate evaluation of 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 by using our inverse modeling method requires further research (involving, e.g., multi-model inversions) that goes beyond the scope of this study. Corresponding caveats are also provided in Sect 3.5 and in the final section of the manuscript (Summary and Conclusions).

Eq. (7) does not involve any emission estimates that were obtained using a model. In a corresponding discussion, we have additionally clarified the distinction between the emission estimates obtained from a bottom-up emission inventory and optimal emission estimates inferred from the measurements by using the modeled relationships between the column amounts of a given proxy species and corresponding emissions.

Referee's comment: The authors should comment on to what extent limiting themselves to measurements over land produces a bias in their estimates, as NO2 and CO emitted over land and transported over the ocean is not considered (due to the limitation to land pixels) while the emissions are included in the emission totals. The same holds for emissions outside the study area (i.e., Ireland and Eastern Europe), which can be transported to the study area and thus be included in the measurements but

not in the emission totals.

First of all, we would like to emphasize that our inversion method involves extrapolating information on pieces of the emission signature in the atmosphere, based on an atmospheric transport model, rather than simple estimation of the atmospheric budget of a proxy species. So, when removing ocean data and data outside the modeling domain, we just reduce the number of elements for such an extrapolation. Specifically, the fact that we analyzed the measurements and emissions only over land (and only over the study region) means that the measurements of NO_2 and CO emitted over land but transported over the ocean were not used to constrain the corresponding emissions. This limitation affected the amount of data used in the analysis (and thus the size of the vectors $\mathbf{c}_m$ and $\boldsymbol{c}_o$). However, we do not see any reason to expect that this limitation could result in any biases in our emission estimates, which would not be covered by their uncertainty intervals (evaluated as explained in Sect. 3.4). Likewise, we do not expect that any biases in our emission estimates can be caused by NO_x and CO emissions outside of the study region. Indeed, on the scales considered, it seems reasonable to regard temporal and spatial variations of NO_2 and CO originating from any sources (including ship emission) outside of the study region as model errors on top of the modeled variations of NO_2 and CO originating from inside of the study region. Accordingly, we do not distinguish such variations from other errors and treat their systematic and random parts in the same ways as explained in Sect. 3.2 (see Eq. 6) and in Sect. 3.4, respectively. A corresponding discussion is provided in the revised manuscript (see Sect.3.2, the last paragraph).

Referee's comment: As the present study uses the DOMINO NO2 product (version 2), the reference to Bucsela et al., 2013 given on p.5/l. 29 for the AMF uncertainties seems to be off, as that study addresses the NASA OMI NO2 product and not the DOMINO product.

We agree with this remark. We presumed that the algorithms which were used to develop the NASA and KNMI data products were very similar, while, in fact, there are some noticeable differences between them. Accordingly, the reference to Bucsela et al., 2013 is replaced by the reference to Boersma et al., 2011.

Referee's comment: In the discussion p.10/l.32 and following, it should be stated if the same sampling (coming from cloud and intensity filtering and satellite coverage) is also applied to the CTM values. Also, it should be clearly stated that the satellite retrieval's averaging kernels are applied to all CTM profiles to get the modelled columns (at least I hope that this is the case!). Furthermore, the authors should state how they determined tropospheric columns from the CTM profiles (i.e., use of tropopause information).

Indeed, exactly the same sampling (based on the measurement information used in the satellite retrieval procedure) was applied to both the satellite data and the CTM values. Furthermore, the satellite retrieval's averaging kernels were applied to all CTM profiles that were used to get the modeled columns. In the revised version of our manuscript, the corresponding explanations (that were provided in Sect.2.2 and 2.4 of the reviewed manuscript) are extended and formulated more clearly. We also note (in Sect. 2.2) that in relatively rare cases (constituting less than 20 % of the total number of valid observations available for the study region and period) where the tropopause pressure was less than the pressure at the top of the model grid (200 hPa), the lack of the simulated data at altitudes exceeding the height of the upper model layer could result in some underestimation of the modeled tropospheric columns, but the effect of such underestimation on the results of our analysis is expected to be small, owing to application of a debiasing technique described in Sect. 3.2 and validated in Sect. 3.5.

Referee's comment: In p.11/l.8 and following, and the corresponding description in Sect. 3.2. it should be noted that the seasonal changes in the "bias" between observa-

tions and models strongly points towards systematic errors in the assumed seasonal cycle of emissions and the satellite retrievals (which mostly come from the assumed surface reflectance climatology and the emissions used in the a-priori NO2 profiles used for the AMF calculations in the DOMINO retrieval).

The corresponding remark is added in Sect. 2.2 of the revised manuscript. In particular, we note (in Sect. 2.2) that the seasonal changes in the monthly biases may partly be due to errors in the seasonal cycles of the emissions specified not only in CHIMERE but also in the global models that were used to obtain the a priori NO_2 and CO profiles for the corresponding retrieval procedures; such changes may also be indicative of some errors in the assumed seasonal variations of other parameters of the retrieval procedures, such as, e.g., surface reflectance or atmospheric scattering by clouds and aerosol in the case of the NO_2 retrievals and surface temperature, local emissivity, vertical distributions of atmospheric temperature and humidity in the case of the CO retrievals.

Referee's comment: In the context of Eq. 3 (and in general), it would help if the authors clearly stated that their emission estimates are annual totals for the whole study region, divided by sector and species.

The components of the control vector involved in Eq. 3 are explained more clearly (as suggested by the referee) in the first paragraph of Sect. 3.2 of the revised manuscript. Note that our idea was to provide (in Sect. 3.2 and Sect. 3.3) first a description of our procedure for a general case (an arbitrary region and the arbitrary numbers of emission source categories and proxy species), since we believe that the method proposed can be used in other similar studies. Some details specific to the given study are provided in the end of Sect. 3.2 and in Sect. 3.4).

Referee's comment: Furthermore, the authors should explain how they derive the Jacobian matrix S.

The estimation method used in this study requires the knowledge of the product of the Jacobean matrix, S, and of the emission allocation vector, a, while the knowledge of the Jacobean matrix itself is not needed. The corresponding remark is added in the second paragraph from the end of Sect.3.2.

Referee's comment: In Eq. 4, it should read "argmin" instead of "agrmin".

We are sorry for this misprint. The equation is corrected in the revised manuscript.

Referee's comment: Furthermore, in p.12/l.30, the authors should explain if and how their results are biased towards summer observations, as a result of more available satellite measurements in summer (due to cloud/intensity filtering).

A difference in the numbers of observations in summer and winter can result in a bias in our estimates if the assumed seasonal cycle of emissions is incorrect. For example, if the seasonal cycle overestimates the emissions in summer and underestimates in winter, then, taking into account that more satellite observations are available in summer than in winter, our annual estimates can be biased negatively. We attempted to take into account possible errors in our estimates due to errors in the temporal allocation of the emissions in the uncertainty analysis as explained in Sect. 3.4. A corresponding remark is added in Sect. 3.2 (see the paragraph before Eq. 6).

Referee's comment: In the discussion of Eq. 6, it might help the reader if the authors would clearly state that $\Delta_i{}^s$ is the average difference between modelled and observed columns for the month m in which observation i lies. (Or did I understand this wrong? In that case, the authors should clarify their explanation of what exactly they did.)

Indeed, $\Delta_i{}^s$ is the average difference between the modelled and observed columns for the month m in which observation *i* lies. An explanation provided before Eq. (6) is revised accordingly.

Referee's comment: In Eq. 10, the authors should explicitly define .

The explanation that was provided after Eq. (10) in the reviewed manuscript is revised and is defined explicitly.

Referee's comment: On p.15/l.17, I believe it should read (missing hat).

We are sorry for this misprint which is corrected in the revised manuscript.

Referee's comment: On p.17/l.31 it might be instructive if the authors gave the sample sizes (number of daily values going into the calculations) resulting from the subsampling.

The requested numbers are provided in Sect. 3.4 of the revised manuscript.

Referee's comment: The authors should discuss to what extent limiting themselves to only one alternative emission inventory (EDGAR and CDIAC for NO2 and CO) might be problematic after all, in principle there are more alternatives, and the authors could in principle use an ensemble of alternative inventories.

Ideally, it would indeed be best to consider an ensemble of several independent inventories providing the data on spatial distributions of emissions of all the species (NO_x, CO and CO_2) involved in our analysis. We tried but, apart from the data of the EDGAR inventory, we could not find publicly open inventory data satisfying these criteria and available for the region and period considered. In particular, we examined several in-

ventories presented in the GEIA data base (http://eccad.sedoo.fr/). So, in this study, in view of the limited practical availability of necessary inventory data, the "ensemble" approach could not be fully realized. A corresponding remark is added in Sect. 3.4 of the revised manuscript. Limitations of a simpler and slightly different approach used in this study are in detail discussed in Sect. 3.4.

Referee's comment: On p.20/l.24, the authors should explain how a Cholesky decomposition (of what?) is used to create error samples.

The requested explanation is added in Sect. 3.5 of the revised manuscript. In particular, we explain that samples of the errors with the covariance structure given by Eq. (16) were generated from a Gaussian distribution by using a standard method (Press et al., 1992) involving the Cholesky decomposition of the correlation matrices that were specified, in our case, using the covariance functions given by Eqs. (16) and (17). The Cholesky decomposition of a correlation matrix gives a lower-triangular matrix, **L**; applying this matrix to a vector of uncorrelated samples of Gaussian noise, $\boldsymbol{u}$, gives a vector, $\mathbf{L}\boldsymbol{u}$, with the components satisfying the original correlation matrix.

Referee's comment: In p.21/l.6, it seems that the word "not" is missing in "Note that not only anthropogenic. . . ".

Indeed, the word "not" was missing. A corresponding correction is made in the revised manuscript.

Referee's comment: The authors should explicitly state if they consider the CO_2 intensive cement production as part of the TCO or EHI sector.

We stated in Sect. 2.3 that CO_2 emissions from cement production are not considered

in our study. A reason is that, unlike FF burning, cement production is not associated with considerable emissions of either NO_x or CO, and so satellite measurements of the corresponding proxy species cannot provide strong constraints on CO_2 emissions from cement production.

Referee's comment: Comparing the "more than 60%" in p.28/l.24 to Fig. 10, it seems to me that this is a bit overestimated; from the figure alone it looks more like 50% to me.

Actually, we say about 60% relative to our estimates, not relative to the EDGAR data. A clarifying remark is added in the revised manuscript.

Referee's comment: Fig. 2 should explicitly state the units on the y-axis (at least use the word "normalized" in the caption).

It is indicated in the revised manuscript, that the values shown in Fig 2 are the normalized monthly NO_x and CO emissions and are unitless.

Referee's comment: Figs. 10+11 should be more specific in the units on the y-axis: NO_x emissions in Tg NO_x (which NO/NO_2 ratio?), or Tg N, or Tg NO_2, . . . ? The same holds for CO and CO_2 emissions.

We indicated that NO_x emissions on the y-axis of Figs 10 and 11 are given in Tg NO_2 and CO_2 emission are given in Pg CO_2.

Referee's comment: The bar for EDGAR in Fig. 11 should be the same color as the bar for EDGAR in Fig. 10.

Actually, the both figures were plotted using exactly the same color settings. To avoid a

possible impression (which may be due to differences in surroundings) that green color used in the bar for EDGAR in Fig. 10 is darker than that in Fig. 11, the figures are re-plotted using a different color scheme.

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