

Interactive comment on “Inverse modelling of the spatial distribution of NO_x emissions on a continental scale using satellite data” by I. B. Konovalov et al.

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We are thankful to the reviewer for the generally positive evaluation of our paper and helpful critical comments.

Most of the reviewer's comments concern possible uncertainties in our results, which are related to potential shortcomings of the model, measurements, and their combination in our inversion algorithm. Another part of the comments concerns our estimates of uncertainties in the model, input data and a posteriori emissions. In our opinion, both parts are related. Indeed, if we managed (as we believe so) to take into account implicitly or explicitly all kind of model and measurements errors, and if, in spite of

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those errors, the a posteriori emissions are less uncertain (on the average) than the a priori ones, then we would have achieved the main objective of the inversion. In this case, even though the identification of potential sources of errors in the model or in the measurements would be important from the point of view of further improvements in the inversion scheme, the existence of these errors does not invalidate our results.

Accordingly, we consider the reviewer's comments as evidence that the description of our method in the reviewed manuscript was, probably, too formal, and that presentation of the mathematical side of the problem hampered comprehension of general ideas of our approach. Hence, we have made a number of changes in the manuscript in order to clarify the description of the method, in particular concerning the treatment of uncertainties. On the other hand, we are aware that our study is rather different from other inverse modeling studies in the part concerning estimation of uncertainties. Due to this, comprehension of our approach inevitably requires some effort even from a reader who is well acquainted with previous inverse modeling studies. Here, we would like to remind general ideas of our method and to present arguments that major concerns of the reviewer have already been adequately addressed in our study. Some important details of our method are discussed also in the response to the other reviewer.

The principal distinctive feature of our method is that the parameters of the inverse modeling scheme which characterize uncertainties in observed and modeled NO₂ columns and a priori emissions are not assigned as a part of preliminary analysis or as a kind of expert estimates but are evaluated self-consistently within the inversion scheme. This innovation and reasons for introducing it are noted in the Introduction of the reviewed paper (p. 12646, lines 5-26), while the algorithm is described in detail in following sections (see Sections 3.2, 3.3.1, 3.3.2, 3.3.4 and 3.4.1). In other words, the uncertainties in NO₂ columns and a priori NO_x emissions are internal rather than external parameters of our algorithm. Accordingly, for example the knowledge of the accuracy of GOME measurements of NO₂ columns is not a prerequisite for the success of our method. The same holds for the uncertainty in the simulation of NO₂ columns

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with CHIMERE for a known set of NO_x emissions. Note also that we do not separate model and measurement errors, their random parts are combined in a single parameter σ_c (see, e.g., Eq. (4)). Also for that reason, individual knowledge of both model and measurement error is not necessary. This treatment of errors is clearly stated at page 12657: “the variance σ_c^2 represents here both the model and measurement errors and is equal to the sum of the variances of errors of the model and the observations taken separately (see, e.g., Tarantola, 1987; Enting, 2002) . . .”. The second type of random errors is related to emissions (σ_e). As noted in page 12654, we are working on three month averages in time and do not consider temporal evolution of observed characteristics or parameters. Accordingly, “random” refers here to a spatial sense. On the other hand, systematic errors in observations, model and emissions (average over the model domain) are regrouped in a term δ_c (also in Eq. 4). As it has been emphasized in the Introduction, neither errors of the model nor uncertainties in measured NO₂ columns, nor those in emissions are known a priori sufficiently well.

The basic ideas behind the estimation of the uncertainties in our study are first to estimate the ratio φ of *uncertainties* in NO₂ columns σ_c and a priori NO_x emissions σ_e by minimizing the difference between measured and simulated data for NO₂ surface concentrations (see Eq. (7), (9) and the related discussion in Sec. 3.3.1). Indeed, the ratio φ indicates how much we trust in a priori emissions with respect to observation derived emissions. The combination of both yields optimized emissions (Tarantola, 1987; Enting, 2002). Second, the variance of the debiased difference between the measured and simulated surface NO₂ is used as an estimation of the combined uncertainties in NO₂ columns due to emission, model and observation uncertainty (see the left-hand side of Eq. 15). Third, uncertainties in NO₂ columns due to uncertainties in emissions are estimated by calculating the relationship between the uncertainties of the a priori emissions and respective perturbations of NO₂ columns (see the last term in the right-hand side of Eq. (17)). Formally, as soon as φ is estimated from equation (9), we have to solve just one equation (17) for the one unknown σ_e .

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The important assumption underlying this procedure is that the random (in a spatial sense) uncertainties in the model and the random uncertainties in the measured NO_2 columns are sufficiently independent. This would not be the case, if we used CHIMERE for calculations of air mass factors. This is why it was necessary to use a version of the satellite data that has been obtained with an essentially different model (MOZART). In principle, it could be possible to avoid dependence between satellite data and the model by simulating directly slant columns retrieved from satellite measurements. However, up to our knowledge, this method has not yet been realized in any inverse modeling study, and we believe that the important developments in inverse modeling algorithm necessary to apply would be worthy of a separate publication. We do not think that it is really necessary to go into details of vertical distribution of NO_2 columns from MOZART and CHIMERE in the framework of the present study. Indeed, the model inter-comparison is obviously not a main subject of the present paper, which is already rather lengthy. But most importantly, we do not see how this comparison could help us to get better solution of the considered inverse modeling problem in the light of the distinctive features of our approach mentioned above. Indeed, following to our approach, we take the NO_2 columns derived from satellite measurements and the NO_2 columns simulated by the model “as given”: we need only that these data were sufficiently independent (in the sense that the error covariance were, on the average, much smaller than the error themselves).

In accordance to the above reasoning, the omission of the upper troposphere, emissions from lighting and aircrafts in CHIMERE, and some inconsistency of sampling do not create any problem either. Indeed, a random part of the respective uncertainties in the simulated NO_2 columns is taken into account “automatically”, while systematic errors are simply subtracted from the respective values and cannot influence the results (please see more detailed discussion of this issue in our response to the second reviewer). Note that in this study we try to correct only random uncertainties in emissions, or, in other words, we try to improve spatial distribution of emissions rather than their total estimates for the considered domain.

Moreover, some shortcomings of our scheme noted by the reviewer cannot lead to serious extra uncertainties in our results. Specifically, the emissions from aircrafts represent only 1-2% of the total global NO_x emissions [e.g., Lee et al., 1997]. The contribution of aircraft NO_x emissions to European area total NO_x emissions is estimated to be in the same range of magnitude [see, e.g. Tarassón et al., 2004, Table 7]. Therefore, even though aircraft emissions may, in principle, be more important for some pixels, it is very unlikely that their omission may provide significant contribution to the overall random uncertainty of the a posteriori NO_x emissions, which remains rather significant (about 40 %). The global amount of NO_x produced by lightning is rather uncertain [see e.g., Labrador et al., 2005]. But even if the contribution of lightning NO_x emissions is considerable at a global scale, the fraction of lightning-produced NO_x in Europe is probably much smaller. Indeed, it is well known that major part of NO_x from lightning is produced in tropical regions, while the major part of man-made NO_x is produced in extra-tropical regions. In particular, the estimations of lightning NO_x production derived from satellite measurements [Boersma et al., 2005, Fig.5] show that the yearly average lightning-produced NO_2 columns amounts over Europe is less than $0.02 \cdot 10^{15}$ (molec/cm²), while the three-month average observed values of NO_2 columns (our study) is about $2.9 \cdot 10^{15}$ (molec/cm²). Even if the lightning-produced NO_x amount averaged for summer season is up to four times higher than the its yearly average, these numbers indicate that NO_x emissions from lightning are hardly of any importance in Europe, as long as we consider temporally averaged emissions.

We used the standard version of CHIMERE which enables simulating chemical and transport processes only up to 500 hPa pressure level. We consider this limitation as a reasonable compromise between sufficiently thorough simulations of boundary layer processes with high temporal and spatial resolutions and computational efficiency. Naturally, the later issue is especially important in an inverse modeling study. CHIMERE is not a perfect model but there are no perfect models. For example, the obvious shortcoming of typical global models is rather low spatial resolution.

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In practice, it is most important to insure that shortcomings of a model cannot invalidate results. The reviewer suggests that the model errors due to omission of the upper troposphere should be included in inversion. This, in fact, is already done in the general way discussed above (in particular, by debiasing NO₂ columns from observations and simulations before the inversion).

Moreover, random errors related to the omission of the upper troposphere are hardly significant. Indeed, we have found earlier [Konovalov et al., 2005, Fig. 4] that the spatial distribution of tropospheric NO₂ above the CHIMERE top is very homogeneous and cannot account for the observed features in the distribution of observed tropospheric NO₂ columns. Besides, the simple comparison of simulated and measured NO₂ columns (see Fig.2 in the reviewed manuscript) shows that the model is reasonably well reproduces the spatial structure of measured NO₂ columns. Quantitatively, this agreement is manifested in rather high spatial correlation between simulations and measurements ($R=0.86$, see Fig. 3), especially when our results are compared with the results obtained with global models (which, of course, include upper troposphere) [e.g., Savage et al., 2005].

The sampling of NO₂ columns from CHIMERE in this study is done in a simplified but yet sufficiently accurate way. As it is mentioned in Section 2.1 of the reviewed manuscript, “In order to be consistent with satellite data, which will be described in the next section, the modelled NO₂ columns for each model grid cell are taken between 10 and 11 h of local solar time and only on days with insignificant cloud cover. Since the total cloud cover is not used in the CHIMERE simulation, we use a selection criteria based on a threshold value of the radiation attenuation coefficient. Specifically, we disregarded days on which reduction of solar radiation due to clouds was larger than 30%.” This criterion is not perfect but still sufficiently reasonable. It has also been noted (p. 12649, lines 6-8) that “the sensitivity of simulated seasonally average NO₂ columns to the radiation attenuation coefficient threshold value has been tested and found to be rather insignificant”. The sensitivity is small because we consider season-

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ally average data. This averaging enables compensating of random inconsistencies between GOME and CHIMERE in different days. Typically, the considered period of June-August 2001 includes from 20 to 30 days of GOME daily observations per a given grid cell of CHIMERE and the same number from simulations. Besides, when this study had already been finished, we have considered the difference between seasonally averaged NO_2 columns extracted from CHIMERE using the simplified “radiation attenuation” criterion and NO_2 columns extracted exactly on the days for which GOME data have been provided for a given grid cell. The difference between these two versions of NO_2 columns from CHIMERE proved to be quite negligible when compared with the difference between the NO_2 columns from CHIMERE and GOME. The use of the simplified approach for extracting NO_2 data from CHIMERE allowed us to focus our attention on principal methodological problems which we faced in this study. The change of the data selection method would necessitate re-doing all calculations which, with available computer recourses, would take several months. We strongly believe that this step would be excessive.

We agree with the reviewer’s remark that our method accounts only partially for the transport. It may be useful to note with this respect that the results of special tests shown in Figs. 5 and 6 indicate that even if we took into account more distant transport, the results obtained within the framework of the present scheme would hardly be much more accurate.

Regarding equation (2), it has been noted in the reviewed manuscript (see lines 19-23 on page 12651): “This artificial convolution is intended to replicate the convolution of real NO_2 columns within the GOME window. We approximate the shape of the GOME window on the longitudinal plane by the Gauss function with half-width equal to three grid cells of CHIMERE and assume that a signal outside of the range of seven cells is negligible.” In our opinion, this notice clearly explains the meaning of all numbers involved in Eq. (2). Specifically, the “3” is an assumed half-width of the GOME window, the numbers from 0 to 6 in summations define indexes of the grid cell inside the window

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(the total number of such grid cells thus equals seven), and $9 (=3^2)$ is a parameter of the Gaussian window defining its half-width. Nonetheless, in order to emphasize that all those numbers are directly related to a single parameter (the half-width) of the window, the equation (2) is rewritten in the revised manuscript in a more general form.

The ground based measurements may indeed be biased. Moreover, there are probably large representativeness errors, as discussed in Section 2.4. All these errors are treated similarly to the errors in NO_2 columns, as it has been discussed in Sections 3.3.1, 3.3.2 and 3.4.1 of the reviewed manuscript. Specifically, we perform an observation-based estimation of the random part of errors and subtract the systematic errors (see Eq. (11)&(18)). The random errors are then taken into account in the Monte-Carlo uncertainty calculations as indicated in Sec. 3.4.2 (see Eq. (20)), while the contribution of systematic errors is excluded.

The standard deviations in Table 1 describe the random (in the spatial sense) part of uncertainties in the a priori and a posteriori emissions, as well as in total uncertainties of measured and simulated NO_2 columns. The meaning of these parameters has been explicitly defined by means of Eq. (4) and subsequent remarks. Moreover, all the characteristics that we consider are the averages over three months summer months (see Section 3.1, lines 13, 14 on page 12654), and we do not consider temporal evolution of our estimates. The reviewer's question whether the standard deviations in Table 1 refer to temporal or spatial variations is thus rather difficult to understand in the context of our study.

In view of the above discussion, we cannot quite agree with the reviewer's remark that our study is "only test of the method" and that the reported reduction in emission uncertainties is not real. Of course, the results of our study, as well as results of any other inverse modeling study, are conditional of some assumptions. But in contrast to other inverse modeling studies, the main assumptions that are made in our study are of qualitative, rather than of quantitative character. For example, we assume that errors are distributed in accordance to the lognormal distributions, but we do not make

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definite assumptions about magnitudes of the errors. That is, although our results may indeed be uncertain to some degree, we hope that our estimations are not too far from the reality. It probably should have been mentioned in Conclusion that the reported reduction in emission uncertainties only concerns the random part of uncertainties. The systematic part, although probably not very large (please see the response to the second referee), remains uncorrected. The corresponding part of conclusion is accordingly corrected in the revised manuscript.

We agree with the reviewer's remark concerning the presentation of the emission rates in Fig. 1, 7, and 8. The respective changes are made in the revised manuscript.

The reviewer asks also about statistical significance of the ratios of the biases given in Fig. 11. While it is not possible to answer the reviewer's question for a single site, it is possible to infer the statistical significance of the comparison of model results with the ensemble of stations. As it has been noted in the legend to Fig. 11, "The a posteriori emissions enable the reduction of biases at 15 stations out of 21". This result is already statistically significant. Indeed, a simple Monte Carlo experiment (whose results, unfortunately, have not been mentioned in the reviewed manuscript) has shown that if the result of inversion were equivalent to mere re-sampling of errors in NO_2 concentrations from the same probability distribution (in other words, if the errors of emissions had not been actually improved but simply redistributed), then the number of stations for which the bias was improved would be in the range from 7 to 14 with the probability 0.90. Therefore, we can conclude that the obtained improvement is statistically significant with the probability of error less than 10 percent.

Finally, we would like to note that the inverse modeling of NO_x emissions using satellite data is a very new area of research. It is obvious that because the related problems are very complex and "multidimensional", it cannot be expected that all problems will be definitely resolved in the framework of just one study. Nonetheless, we believe that our study proposes a considerable novel contribution to the field of inverse modeling using satellite observations, and we hope that this paper will be interesting for the audience

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