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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.

I. B. Konovalov et al.

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We are grateful to Dr. Houweling for the critical evaluation of our paper. Although we cannot accept some of his critical remarks, we believe that, on the whole, his comments allowed us to improve our paper and facilitate understanding of our method and results. In our response, we will try to clarify, how the method we set-up makes best use of available observations and of a chemistry-transport model to derive new information about "real world" emissions.

I. The reviewer argues that our paper "confuses uncertainty and variability of the signal". Moreover, he comes to a very serious conclusion that because of this shortcoming

our method of estimating uncertainties is not valid at all ("The authors claim that the uncertainty of the various sources of input data can be estimated from the data using their method, which in my opinion is not the case because the method does not allow separating the contributions of signal and error to the overall variance.)". We think that this remark is due to some misunderstanding of our concept in estimating uncertainties. Hence, along with answering to the reviewer's comment, we would like to recall the general "philosophy" of our method.

Our estimations of uncertainties in NO_2 columns and NO_2 concentrations are based on consideration of differences between their observed and modeled values (see Eqs. (15) and (18)). Commonly, such differences include systematic and random errors of the corresponding estimates. Throughout our paper, we consider only random (in the spatial sense) errors. The random part of errors is separated from the systematic errors by "debiasing" the differences between the observed and measured values, as specified in Eqs. (4), (10) and (11).

As far as the random part of errors is concerned, our method aims at deducing it from the variability of the (debiased) differences between simulations and different type of observations. Let us first consider in a general way, how differences of signals (observation or simulation) are related to errors. For this, let us look to the difference between two estimates of the same characteristic. We put $x_1=x_{true}+\Delta_1$ and $x_2=x_{true}+\Delta_2$, where x_1 and x_2 are two different estimates of the same characteristics x (e.g., NO_2 column) and Δ_1 and Δ_2 are their errors. We have immediately that x_1-$x_2 = \Delta_1 - \Delta_2$ and thus there is no "signal" in the right part. Therefore, we can see in particular, that in contrast to the review's statement, the right-hand part of Eq. (18) does not contain an explicit contribution of the signal. However, we see, that the *differences* in the estimation of x are related to the respective errors. As these errors are random (in a spatial sense), the variability of the difference x_1 - x_2 (for different locations) is linked to the sum of the errors Δ_1 and Δ_2. This concept is applied throughout the paper. Clearly, it links the variability in *differences* of signals to errors, but does not mismatch

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them. More specifically, we have to estimate two types of random errors, one related to a priori emissions (σ_e), the other related to NO_2 columns combining both model and observation related random errors (σ_c).

As also pointed out in our response to the other referee, the basic ideas behind the estimation of the uncertainties in our study are first to estimate the ratio φ of *uncertainties* in NO_2 columns σ_C and a priori NO_x emissions σ_e by minimizing the difference between measured and simulated data for NO_2 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 column NO_2 is used as an estimation of the combined uncertainties in NO_2 columns due to emission, model and observation uncertainty (see the left-hand side of Eq. 15). Third, uncertainties in NO_2 columns due to uncertainties in emissions are estimated by calculating the relationship between the uncertainties of the a priori emissions and respective perturbations of NO_2 columns (see the last term in the right-hand side of Eq. (17)). Formally, as soon as φ is estimated from the equation (9), we have to solve just one equation (17) for the one unknown σ_e.

Again, all equations involved in this derivation *only* include the variance in *differences* of signals, and never involve the variance of the signal alone. So there is clearly no mismatch between the variance of errors and signals.

II. Another major comment of the reviewer concerns our treatment of the bias in NO_2 columns and the meaning of our emission estimates. This is indeed a very serious methodological question, and we consider the reviewer's comment as evidence that this question should have been discussed in the paper more in detail. Accordingly, the discussion is extended in the revised version. Here, we would like to recall, first of all, how this problem is usually treated in other atmospheric inverse modeling studies, some of which are cited in our paper.

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It is recognized commonly that the differences between model and observations include the measurement errors, the errors of the model results due to uncertainties in sources and all other model errors. While the measurement errors are usually discussed in detail, only very tentative assumptions are commonly made regarding two other error sources. For the model error, it is usually assumed that the model results are either perfect, or contain some definite fraction of errors (e.g., 10% [Müller & Stavrakou, 2005] or 30% [Martin et al., 2003]), which is usually pre-specified with no or little discussion. Also, no difference is ever made between systematic and random (in space or in time) parts of the model errors. Indeed, while the measurement errors can, in principle, be assessed using independent ("reference") measurements, the estimation of model errors, which are not related to uncertainties in emission sources, is, to the best to our knowledge, is still unresolved problem. The comparison of the model with measurements is of little help here, since it necessarily assesses both the emission and the model part, and error compensation between both parts cannot be excluded.

As it has already been noted above, we do not assign the model errors "by definition", but try to estimate them together with the errors of measurements based on the measurements themselves. It has also been noted above that in this way we are able to estimate only the random part of the errors, and that our treatment of the bias assumes the subtraction of the systematic errors from the difference between measured and modeled NO_2 columns. Accordingly, we try to correct only those uncertainties in the a priori emissions that cause random differences between the measured and modeled NO_2 columns. In other words, we deal mostly with random uncertainties in emissions, but cannot say anything definite about systematic uncertainties. Nonetheless, by reducing the random part of uncertainties in emissions, we also reduce inevitably their total uncertainties, since the systematic and random errors are additive. Hence, the a posteriori estimates are expected, on the average, to be closer to the unknown true values (e.g., in the RMSE sense) than the a priori emissions. Moreover, it seems reasonable to expect that the systematic uncertainty in the a priori emissions, which is

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closely related to the uncertainty in total emissions for the considered European domain, is much smaller than the uncertainty in emission data for a single pixel. If it is indeed so, then while interpreting our results, it is possible to neglect the systematic part of emission uncertainties. In any case, we believe that our inversion results in substantially improved spatial structure of NO_x emission. The improvement of spatial structure of NO_x emissions was, in accordance to the title of our manuscript, the main purpose of our study.

Because, as it is argued above, consideration of the systematic difference between the measured and simulated NO_2 columns does not supply us with any important information that could be used for inversion, we do not discuss them in detail. Nonetheless, the reader can get an idea about this difference in Fig. 2, where we show the distribution of measured and modeled NO_2 columns, and especially in Fig. 3, where we present scatter-plots of the NO_2 columns and show also average values of the measured and modeled NO_2 columns. Specifically, the average values of measured and simulated NO_2 columns are 2.9 and 2.1 (*10^{15}) molecules/cm^2. As it is suggested in Section 2.3 of the reviewed manuscript, CHIMERE gives smaller values mostly due to the fact that it does not take into account the upper troposphere. Indeed, an estimation made in our previous study cited in the manuscript [Konovalov et al., 2005] shows that the average NO_2 column amount in the upper troposphere (above 500 hPa pressure level) is about 0.5*10^{15}molecules/cm^2. So taking this into account, the systematic difference between average observed and simulated NO_2 columns is only about 10%. As we argued above, we have no information whether this bias is due to observations, model or emissions. But, it is clearly smaller, than the spatial corrections applied for individual grid cells.

With respect to our statement that improving the agreement between measured and simulated NO_2 columns does not necessarily signify the improvement of emissions, the reviewer notes that this statement suggests that “the inversion is actually not meant to improve emission estimates”. Here the reviewer has taken our statement out of con-

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text. It is clear that a better agreement between simulated and observed NO_2 columns cannot be taken as an indication of improvement of a posteriori emissions, because observed NO_2 columns have been used as constraining data. It only gives an indication of the efficiency of the method. In order to assess the improvement of emissions, independent data have to be used, such as NO_2 EMEP data. This is done in section 4.2. In general, the purpose of inversion is indeed to improve emission estimates, but it is not usually equivalent to simple fitting of the model to observations, since both the model and observations contain different errors that must not be attributed to uncertainties in emissions (see e.g., Enting, 2001). In the revised manuscript, we have reformulated the concerned paragraph in an attempt to avoid misunderstanding.

III. The reviewer notes that "hardly any quantitative information on these fluxes [that is, on the a posteriori emissions] is provided that would allow comparison with other studies". In fact, we discuss mainly uncertainties of emission estimates rather than their absolute values. Apparently, the reviewer has not noticed that one of the major quantitative results of our study, which is emphasized both in Abstract and Conclusions is the observation-based estimation of uncertainty in the spatial distributions of NO_x emissions. Values of these uncertainties and their spatial distribution have been presented in Fig.7c. To the best of our knowledge, such kind of estimation is performed for the first time and we believe that it can present significant interest for both atmospheric modelers and providers of emission data. These results can be easily compared with other similar studies, as soon as they will be available.

In addition, we do present quantitative information on a priori and a posteriori fluxes in Fig. 7a, b (as noted by the reviewer) and also in Fig.1. More quantitative information can be easily available upon request. A corresponding offer is introduced in the revised manuscript. Besides, several major cities are marked in the plots: we hope that this will facilitate inter-comparison. We have also re-plotted most of the figures such that to show explicitly the result for each grid cell of the model and thus to avoid possible artifacts caused by interpolation. We are sorry for omission of units in Fig.7a,b, but

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we hope that this omission, which is of course corrected in the revised version, did not preclude understanding of the results in the published manuscript, since the fluxes are presented in the similar format in Fig.1, where the units are given.

The results of our regional study could not be presented in the same format as results of other studies which so far are performed mainly with global models. The reason is that both the scope and method of our study are very different. In particular, the reviewer suggests to add a table "with a priori and posterior emissions per country. . . ". We agree that it would be indeed important to validate and improve the national totals for emissions, but unfortunately, due to the limitations of our method that have been discussed above, we cannot claim that our estimate of total emissions *per country*, are better than a priori ones, especially for such big states as Germany or France. Instead, we have added a similar table for several major cities. While this kind of information is also practically interesting, it will be completely consistent with the scope of our study. Besides, such data will be easy to use for inter-comparison with regional emission estimates.

IV. The reviewer further doubts the usefulness of the deconvolution of GOME NO_2 columns with SCIAMACHY data for a different year. In fact, any spatial averaging always results in suppressing highs and increasing lows in a spatial distribution. Likewise, the GOME data tend to underestimate NO_2 columns over isolated megacities and to overestimate them in their rural surroundings. In the inversion, this leads (we checked it) to clear systematic artifacts ("a dipole" effect) in corrections of the a priori emissions. The a priori emissions themselves are of little help here, unless they are used (by means of the model) for the deconvolution of the GOME data. But this way leads to statistical dependence between simulated and "measured" columns and thus should be avoided. Nonetheless, in order to check the sensitivity of the method to the data used for the deconvolution (CHIMERE or SCIAMACHY output), we presented the results obtained in both ways (see Fig.7c and Fig. 8a). It is easy to see that they are very similar. The consistency of these results suggests that uncertainties of

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SCIAMACHY data do not introduce considerable extra uncertainties in the inversion.

The inversion obtained with SCIAMACHY data taken alone is qualitatively rather similar but quantitatively different (see Fig 8b). The differences are mainly associated with the estimation of uncertainties and are discussed briefly in page 12669, lines 7-11 (here and below we refer to the published version of the manuscript). However, one has to keep in mind that when deconvoluting GOME data, we only use information on fine-scale features of NO_2 columns from SCIAMACHY. The fact that SCIAMACHY data correspond to a different year may indeed be a reason of some uncertainties in the composite columns on the fine scales. However, this extra uncertainty is implicitly taken into account in our estimation of the overall random uncertainty of the GOME/SCIAMACHY composite and simulated NO_2 columns.

V. The last major comment concerns the use of the units in the reported emissions and uncertainties. We have agreed already that the omission of units in Fig. 7 should be corrected. In all other instances, our estimates are dimensionless. We assumed this to be obvious because we consider mainly logarithmic values. For example, the difference between two logarithms (e.g., $\ln(x_1)$-$\ln(x_2)$=$\ln(x_1/x_2)$) does not depend on the units of the respective values (x_1 and x_2) as far as their units are common. Nevertheless, in order to avoid any problems in understanding, a specific explanation is introduced in the revised manuscript.

Specific comments:

Page 6. SNAP sectors are explained, as suggested.

Page 11. We do not know a true reason for the large difference between the model and measurements in Sniezka station. There could be either some problem with accuracy of the measurements, or with their representativeness (e.g, due to the presence of strong NO_x sources nearby).

Page 26. This reviewer's remark suggests that the respective argumentation was not

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quite clear and thus it is improved in the revised manuscript. In fact, we did not actually mean that "the RMSE improves from 0.3 to 0.25". For better understanding, it may be useful to look again to the respective fragment of the original text: "Indeed, on the one hand, using our model we have found that if the uncertainty of NO_x emissions was about 0.6 and if other sources of errors of NO_2 concentrations were absent, the corresponding RMSE for near surface NO_2 concentrations would be about 0.3 (instead of 0.63)". Now, let us assume that errors (related or not to emissions) are normally distributed and independent, then squares of errors are additive. Then RMSE due to non-emission related errors would be 0.55. Thus the observed reduction of RMSE from 0.63 to 0.56 implies that errors caused by uncertainties in a posteriori emissions should be very small. The corresponding RMSE would be less than 0.1. This corresponds indeed to a strong reduction in the part of error related to emissions (with respect to the initial value of 0.30).

Figure 6b. The blue curve in Fig. 6b is calculated indeed with perfect data. However, the model approximation (12), which is used for the inversion, is not perfect, due to linearization of the model. Accordingly, the presence of the minimum reflects the errors of the approximation. The minimum at φ=0.2 suggests that the standard deviation of these uncertainties equals to 0.12 (=0.2*0.6).

Technical corrections:

Page 18. The use of the "a priori" is mended.

Page 19. This misprint is already corrected in the published version of the manuscript.

Page 19, "emisions". This misprint is corrected.

Page 19: "The RRMSE expressed in terms of absolute emissions'=RMSE". We cannot agree with this suggestion. In accordance to definition (14), the RRMSE is the ratio of the RMSE of retrievals to the RMSE of the a priori guess.

Page 21. The sentence of the question is reworded.

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Page 22, an absent reference to Fig.4. There is indeed a misprint in page 12653, line 5: instead of “shown in Fig.3” should be read: “shown in Fig.4”.

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