

We appreciate the referee #1 for giving valuable comments. We respond to each specific comment below and indicate where the changes we have made in the manuscript. The comments and questions from the referee are in blue italic font.

*General comments:*

*Additional descriptions are needed in section 2 so that an independent author could reproduce the results from the information in the paper. For example, the prior emission inventory used and its error statistics (Pf) should be carefully enumerated. The authors refer to Mijling and van der A (2012) and to Ding et al. (2015) for detailed information, but the prior emissions used in those two studies are not identical.*

The NO<sub>x</sub> emissions derived with DECSO become independent from the a priori emissions after the spin-up time of this algorithm, which is about 3 months. Different a priori emission inventories used in Mijling and van der A (2012) and Ding et al. (2015) have little influence on the final emission results. To explain our method, we add the following sentences to line 6 of page 3:

“.....domain is taking into account. The transport is calculated using an ensemble of 150 isotopic 2-D trajectories for each grid cell. The inversion method used in DECSO is based on an extended Kalman filter. The extended Kalman filter is applied on emissions which uses a persistent emission forecast model. The emissions and their error covariance derived from DECSO are independent from the a priori emission inventory after a spin-up time of about 3 months.”

On line 11 page 3, we add:

“..Range Weather Forecasts (ECWMF). The land use information we use for CHIMERE is from the GlobCover Land Cover database in 2009.”

*The high resolution of the CHIMERE model system is a significant advantage over other attempts to relate OMI NO<sub>2</sub> measurements to emissions. However, the authors should acknowledge that there are known biases in the OMI product they are using that stem from low resolution elements of the retrieval. These will bias the resulting emissions by amounts that are comparable to the changes the authors derive. There will also be biases from the 25km resolution of the model that are not negligible especially for sources that are small compared to the grid scale. While it is not necessary to do new calculations, it is necessary that the paper discuss the results presented in light of this related research.*

We agree with the referee that the NO<sub>2</sub> observations suffer from biases due to the limited resolution of the a priori information used in the retrieval cases with high NO<sub>2</sub> concentration gradients (Heckel et al., 2011; Russell et al., 2011; Laughner et al., 2016). However, in China, there are few isolated emission sources with large concentration gradients. Therefore, several validation studies of the DOMINO v2 product with MAX-DOAS measurements show no significant biases in China on average (Irie et al., 2012; Wang et al., 2016). Ma et al. (2013) evaluated the gradient smoothing effect in the DOMINO v2 retrieval product in China and found no significant bias in winter and an upper limit for the bias in summer of 11-26%. Other studies show that the NO<sub>2</sub> observations suffer from biases in scenes with high aerosol concentrations in China (Lin et al., 2014; Kuhlmann et al., 2015; Chimot et al., 2016). Large biases coming from for example the high aerosol cases are avoided by the emission update constraint explained in section 3.2.

We add the discussion on line 25 page 9:

“Direct validation of emission estimates is not possible due to the lack of independent observations. The accuracy of the emission estimates depends largely on the accuracy of the NO<sub>2</sub> satellite observations and of the CTM used in the inversion. Any random errors in the observations or the model are described in the Kalman Filter and result in an error estimate for the emissions. Biases are more difficult to quantify. A bias in the CTM can occur due to its limited resolution (Valin et al., 2011). In DECSO, the grid NO<sub>2</sub> columns simulated by the CTM are projected via a high resolution grid onto the footprint of satellite observations, which avoids the bias caused by different resolutions of the CTM and satellite observations. The NO<sub>2</sub> observations can suffer from biases in scenes with high aerosol concentrations (Lin et al., 2014; Kuhlmann et al., 2015; Chimot et al., 2016) or the limited resolution of the a priori information used in the retrieval cases with high NO<sub>2</sub> concentration gradients (Heckel et al., 2011; Russell et al., 2011; Laughner et al., 2016). In China, however, there are few isolated emission sources with large concentration gradients. Therefore, several validation studies of the DOMINO v2 product with MAX-DOAS measurements show no significant biases in China on average (Irie et al., 2012; Wang et al., 2016). Ma et al. (2013) evaluated gradient smoothing effect in the DOMINO v2 retrieval product in China and found no significant bias in winter and an upper limit for the bias in summer of 11-26%. Large biases coming from for example the high aerosol cases are avoided by the emission update constraint explained in section 3.2.”

*Model error is an important aspect in emission inversion. As meteorological variables are not updated in DECSO, the sensitivity of observation to emissions (H) can suffer from model transport errors, which will bias the emission estimates. Discussion of the implication of model transport errors and its treatment on the emission inversion here.*

We use the meteorological data from ECWMF with the time interval of 3h in the CTM CHIMERE. In the calculation of emission trajectory analysis, we interpolate the meteo data into half an hour time steps. The meteorological variables are updated. The errors of meteorological data only leads to random errors, which are covered by the Kalman filter. The threshold of H matrix described in section 3.4 helps to reduce the effect of the transport errors caused by errors in the meteorological data.

We explained this in the paper on page 3 line 6:

“...in which the transport of NO<sub>2</sub> over the model domain is taking into account. The transport is calculated using an ensemble of 150 isotopic 2-D trajectories for each grid cell. For the trajectory analysis, we use the operational meteorological forecast of the European Centre for Medium-Range Weather Forecasts (ECWMF) interpolated into half an hour time steps.”

*The paper describes the resetting of the threshold of matrix H that represents the sensitivity of observation to emissions. The authors update the minimum value for H matrix elements from 0.05 to 0.1 hour. The motivation is that the H elements for observations located at the edge of the plume are usually small. Without explanations from a tracer transport perspective, I'm not convinced that setting a minimum is the appropriate approach to solve this problem. H elements calculated from the simplified 2D trajectories represent the contribution from model emission grid to the observations. Setting the 0.1 hour threshold could arbitrarily enhance this sensitivity for some emission points, and overcorrects the emissions which observations are not sensitive to. Some tests showing these effects are negligible and that a choice of 0.1 is optimal should be added.*

The H matrix plays an important role in the calculation of the Kalman Gain. The low value in the H matrix can lead to high values in the Kalman Gain analogue to the effect of

low Eigen values in any inversion. Therefore, the Kalman gain becomes sensitive to errors in the low values of H matrix elements. We have tested several threshold values of the sensitivity matrix H by comparing the results over some isolated hot spots. When the threshold is too high, we are running into numerical problems. 0.1 is the optimal choice based on these tests.

On line 33 page 6, we add:

“..... In this study we set the threshold value to 0.1 hour based on several tests using different threshold values”

*The model assumption of persistent emissions is inconsistent with the behavior of biogenic, fire and lightning emissions. Additional discussion of this issue is needed.*

The persistent emission model is for day to day evaluation. Emissions from any lasting changes are captured but with a delay which has been discussed in Ding et al. (2015). Fast, temporary changes of emissions (less than 1 day) cannot be captured or are underestimated by DECSO. Lightning emissions for example are not detected. Fires are usually detected since they last for several days.

*For example, there is some knowledge of the mechanisms of biogenic emissions and models are available that represent processes. These processes vary strongly in response to temperature and soil moisture. e.g. Hudman, et al.: A mechanistic model of global soil nitric oxide emissions: Implementation and space-based constraints, Atmos. Chem. Phys. Disc., 12, 3555-3594, 2012.*

*Do the derived biogenic emissions behave as expected in response to temperature or rainfall?*

We could refine our persistency model with such information, but this would mean that we add a priori information based on land use, temperature and soil moisture. This would add additional complications and we still would miss for example the commissioning of a new power plant. Therefore, we prefer using a very simple model without a priori information that is able to follow changes on a time scale of days or longer.

In Mijling et al. (2013), we see that the periodicity of total NO<sub>x</sub> emissions in Mongolia follow the rain/temperature cycle.

*It does not appear that lightning NO<sub>x</sub> emissions are represented in the model. Is it possible that the effects of lightning are interpreted as biogenic emissions?*

*Is it possible that fires are interpreted as biogenic emissions?*

We are not able to distinguish the emissions of different source categories; so all emissions derived are represented as surface total emissions. If the emissions changes are due to lightning, in DECSO, it is considered to the surface level.

We emphasize it by changing the text on line 9 page 8:

“ DECSO v5 provides independent information about total surface NO<sub>x</sub> emissions in the region since it is difficult to distinguish source types of the emissions.”

*Details:*

*There are several other studies using Kalman filter and related methods to estimate NO<sub>x</sub> emissions The authors should cite them.*

We add the reference in the introduction on page 1 line 33:

“...This algorithm takes the transport of NO<sub>x</sub> from its source into account by including 2-D trajectory analysis in the sensitivity of NO<sub>2</sub> concentration on NO<sub>x</sub> emissions. Other NO<sub>x</sub> emission studies based on a Kalman Filter often use the decoupled direct method to calculate the sensitivity (Napelenok et al., 2008; Tang et al., 2013) or an ensemble Kalman Filter (Miyazaki et al., 2012).”

*The equations, data and method should be provided for the calculation of soil emissions in Guangxi using emission rates from Li et al. (2007).*

We calculate the emissions by multiplying emission rates with the area of the forests in the province.

We change the text on page 7 line 34:

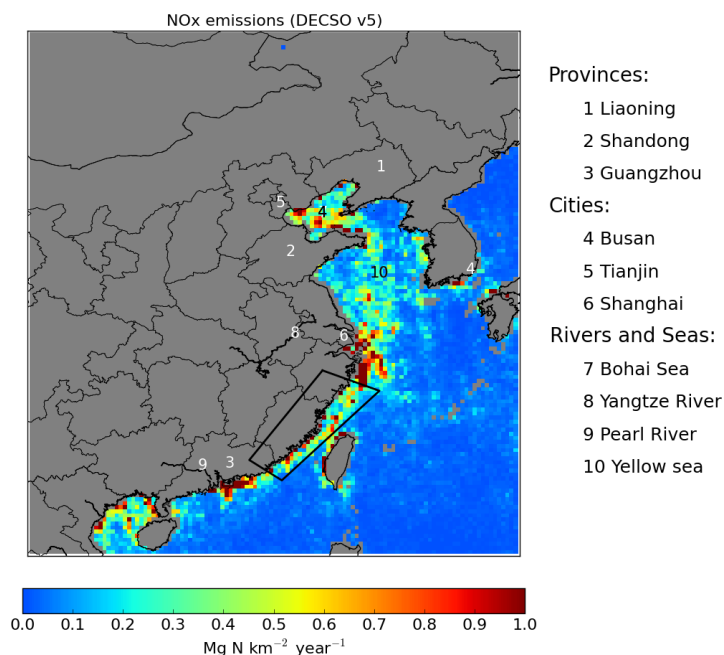
“About 60% of the province is covered by forest ([www.forestry.gov.cn](http://www.forestry.gov.cn)), which is about  $14 \times 10^5$  km<sup>2</sup>.”

On page 8 line 8, we change the text into:

“By using their range of emission rates, we calculate that the soil NO emissions in Guangxi in a range from 30 to 822 Gg N per year by multiplication with the forest area, the upper limit being...”

*Figure 7: locations mentioned in section 4.2 should be marked in Figure 7 for readers who are not familiar with locations in Asia.*

We add markers in Figure 7 mentioned in section 4.2 and change the figure 7 and change Figure 7 in the paper.



*The authors should define “total emission” in this paper because it actually only includes anthropogenic and biogenic components. The general total emissions should include lightning NO<sub>x</sub> also. Suggestion is to rephrase it as total emissions from surface.*

We define the total emissions by changing the text on line 4 page 7:

“By excluding biogenic NO<sub>x</sub> emissions in CHIMERE, we derive the total surface NO<sub>x</sub> emissions from satellite observations. The total emissions from surface include anthropogenic, biogenic and fire emissions.”

*Support for the statement: "the errors caused by few observations on the edge of the emission plumes have been decreased" by updating H should be elaborated.*

See the discussion above

*There are independent measurements from the national in situ observation network collected and maintained by the China National Environmental Monitoring Center (CNEMC).*

There are indeed independent measurements from the national in-situ observation network. Since the validation of emissions with direct observations is difficult unless a CTM is involved, the results are highly related to the choice of the CTM. We will address the validation of emissions in future work.

## References

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