

We thank both reviewers for their careful evaluation of the presented work. Most of their comments and remarks have been processed in the manuscript, which we believe has gained in clarity and scientific soundness. Below is a point-by-point reply (in **black font**) to this round's reviewer's comments (in **blue font**).

Response to Reviewer #2

1. The paper reveals that NO_x emissions are increasing dramatically over China, in some provinces by more than 20%/yr. Air pollution is a serious health and environment issue in China and the continuing increase in NO_x emissions is contributing to the problem in a major way. Of course, this publication should not make a political statement, but it should at least mention the great relevance of the high NO₂ concentrations for public health in China and that the increasing trends are a matter of concern. There is no single line in the introduction, the abstract or the conclusions making a connection between high NO_x concentrations and environmental and health issues apart from mentioning that NO₂ is an air pollutant. By neglecting these aspects the paper doesn't sufficiently value its relevance.

We elaborated this aspect in the beginning of the introduction: "The unprecedented increase in energy demand, industrial production, urbanization and car ownership has resulted in significantly increasing air pollutant emissions. Being responsible for respiratory and cardiovascular problems, these high levels of air pollution affects the health of many people living in this region."

2. Section 2 describes the DECSO inversion method. Only the computation of the sensitivity matrix H is described, but no details are given on the Kalman filter. Although the DECSO method was described in more detail elsewhere, a few more sentences are needed to explain the setup of the Kalman filter. As the reader only learns in later sections, the filter sequentially adjusts the emissions initialized at the beginning of the simulation (Jan 2007) to obtain a better match between observations and model, but it is not clear how strong this constraint is since no information is provided on relative uncertainties assigned to the observations and the model, nor on the uncertainty assigned to the initial emission field. This information is needed. It would also be good to mention already in Section 2 that the filter needs a few months of spinup at the beginning and that the filter likely creates some time lag in the emission estimates, because unlike a Kalman smoother, it only assimilates past observations.

A similar point was also made by reviewer #1. We added information on the response time of the Kalman filter to Section 2 and Section 4.2. More information is added in the Introduction on the model and Kalman Filter set-up is added in the introduction.

3. I think there is an error in Equation (1): The exponent should be $\exp(-(T-t)/\tau)$ rather than $\exp(-t/\tau)$ because, to my understanding, it should reflect the exponential chemical depletion of NO_x from its emission at time t to the observation at time T. In the present form of the equation, emissions close to time T would have the lowest weight, emissions at the beginning of the interval (i.e. 24 hours ago) would have the highest weight.

The equation is not inconsistent. However, the confusion of the referee might be caused by an error in the definition of Ω . This should read: “ $\Omega_{ij}(t)$ describes the transport of NO_x from cell j to i during $[T-t, T]$ ”. Bear in mind that the variable t in the integral indicates the time **before** observation time T .

For example, at $t=0$, the moment when satellite data is assimilated, the factors in the integral are:

- $f(T)$ representing emission strength at observation time T ;
- $\Omega(0)$, representing the transport during $[T, T]$, i.e. no transport has taken place;
- $\exp(0)$, indicating that the freshly emitted NO_x has not decayed yet.

At $t=T$, 24h before assimilation will take place, the factors in the integral are:

- $f(0)$, representing the emission strength 24h before assimilation;
- $\Omega(T)$, representing the transport over the grid during the $[0, T]$ interval;
- $\exp(-T/\tau)$, indicating the NO_x decay factor during 24h based on a lifetime τ .

Alternatively, by substituting $t'=T-t$, the equation can be rewritten to:

$$H_{ij} = \frac{dc_i^{\text{NO}_2}}{de_j^{\text{NO}_x}} = \gamma_i \frac{a_j}{a_i} \int_0^T \exp(-(T-t')/\tau_j) \Omega'_{ij}(t') f_j(t') dt'$$

in which $\Omega'_{ij}(t)$ represents the transport of NO_x from cell j to i during $[t', T]$. In the new text, we substituted this new expression for the old version for clarity.

P17521, lines 7-9: Unclear what kind of “emission estimates” are meant here, bottom-up or top-down estimates

To clarify these lines, we substituted these lines with:

“Satellite observations of air pollutants have a high temporal resolution, they are spatial consistent, and are rapidly available. Emission estimates based on observations of short-living species as NO_2 , SO_2 and PM allow for the monitoring of emission trends, giving important insight in the environmental impact of socio-economic events and the effectiveness of air quality policy.”

P17521, line 12: “spaceborn” -> “spaceborne”

Corrected

P17522, line 10: “relative high” -> “relatively high”

Corrected

P17523, line 10: “can be found in (Mijling and ..)” -> “can be found in Mijling and Van der A (2012).”

Corrected

P17524, line 21 and table 1: It is not quite clear whether all emission totals are for exactly the same domain or whether e.g. the DECSO estimates were corrected for the fact that it only covers 94% of all Chinese emissions.

The emissions in Table 1 are all given for the domain illustrated in Figure 1. The text at the beginning of Section 3 and the caption of the table have been adapted to avoid this confusion.

P17527, line 4: For regions dominated by industrial and power plant emissions the question of the vertical distribution of NO_x emissions becomes relevant. If in the model all NO_x emissions are released at the surface but in reality are released from stacks to higher levels, the model may underestimate the amount of NO_2 at elevated levels where the satellite is more sensitive. Such an effect could lead to an overestimation of NO_x emissions, e.g. over Mongolia where DECSO is significantly higher than EDGAR.

The reviewer is right that unrealistic emission injection heights can be a cause of error. If the bulk of modeled NO_2 is closer to the ground instead than in reality, the averaging kernel will amplify this signal to compensate for its insensitivity in lower atmospheric layers. The modeled NO_2 concentration will be overestimated, which results in an underestimation of the NO_x emission. This effect is not expected to be large, because the variation of the averaging kernel over a typical stack height (50–150m) is not so large [Eskes and Boersma, ACP 2003].

Another source of error due to incorrect emission injection heights will be caused by different transport directions and distances of the NO_x plume due to wind shear. Although this introduces errors in the sensitivity relations in matrix \mathbf{H} , it is unlikely that this will cause structural biases in time and area averaged emission estimates.

P17527, line 18: If the absolute error sigma was used as weights in the regression (Eq. 2), then this should be mentioned explicitly. What value of sigma was used? As a side remark: Instead of estimating a phase phi, Equation 2 could be written as a superposition of a sine and cosine, which would reduce the equation to a simple linear regression problem.

We took for sigma 10% of the mean value of the time series (added to the text). This is however arbitrary, because taking a fixed value of σ for each data point makes the regression independent for the exact value of σ . A different value of σ would basically multiply all weights by a same factor, which would result in the same fit.

P17528, line 27: “originate” -> “originated”

Corrected

P17529, line 17: As mentioned above, the fact that the method may introduce a time lag should already be stated in Section 2 presenting the DECSO algorithm.

This point was also made by the other reviewer. We included a discussion of the observed time lag in Section 4.2 with a reference to the Kalman response time, now discussed in Section 2.

P17530, line 20: Although Section 4.2 presents a nice way for separating biogenic from anthropogenic emissions, the results should probably be presented with a word of caution. E.g. Inner Mongolia was presented before as a province with a dominance of emissions from power plants and heavy industry. This kind of emissions is known to exhibit little to no seasonal variability. Thus, the lack of a seasonal cycle in Inner Mongolia could not only be due to opposing biogenic and anthropogenic emissions, but could also be due to the dominance of power plant emissions. Assuming a constant ratio between absolute level and amplitude of the seasonal cycle may not be valid for this province.

We added: “The biogenic emission estimates should be used with caution. The less pronounced seasonality of the dominant power plant emissions might distort the assumed ratio, and can partly explain the lack of seasonal cycle in Inner Mongolia by itself.”

P17534, line 17: “growing rates” -> “growth rates”

Corrected

P17535, line 1: As mentioned above, I do consider the estimates of the relative shares of anthropogenic and biogenic emissions based on the analysis of the seasonal cycles as highly uncertain and therefore recommend adding a word of caution to these numbers.

We weakened our findings in the Conclusions to “a rough estimate”.