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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Regional nitrogen oxides emission trends in East Asia observed from space" *by* B. Mijling et al.

Anonymous Referee #2

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General comments:

This study applies a computationally cheap inverse estimation method in combination with the CHIMERE chemistry transport model to NO2 column observations from the GOME-2 satellite instrument in order to investigate East Asian NOx emissions, their trends, distribution, and seasonality.

Although there have been several previous studies combining satellite observations with (inverse) modeling to estimate emissions in this region, this study distinguishes itself from others by covering an extended period of time (5 years from 2007-2011) at high resolution (0.25°x0.25°), made possible by the computational efficiency of the

method. The study convincingly demonstrates that the "DECSO" inversion method, presented in more detail in a previous paper, produces very reasonable results and is suitable to study regional emission trends.

The paper provides a concise and thorough analysis of emission trends over China, South Korea and North Korea. It reveals very interesting and relevant differences in the evolution in different Chinese provinces and sheds further light onto the effectiveness of air quality measures during the Beijing Olympic Games in summer 2008. A further nice feature of the hybrid Eulerian-Lagrangian inversion approach is that it allows estimating the contribution of different regions to the NO2 columns at a given location, for example the contribution of emissions from provinces surrounding Beijing to NO2 over Beijing. The paper is very relevant, well written and concise. I thus recommend publication after responding to the following

Minor points:

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1. The paper reveals that NOx 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 NOx 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 NOx 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.

2. Section 2 describes the DECSO inversion method. nly 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

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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.

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 NOx 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.

Small points and technical corrections:

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

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

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

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

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.

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P17527, line 4: For regions dominated by industrial and power plant emissions the question of the vertical distribution of NOx emissions becomes relevant. If in the model all NOx emissions are released at the surface but in reality are released from stacks to higher levels, the model may underestimate the amount of NO2 at elevated levels where the satellite is more sensitive. Such an effect could lead to an overestimation of NOx emissions, e.g. over Mongolia where DECSO is significantly higher than EDGAR.

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.

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

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.

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.

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

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 cy-

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cles as highly uncertain and therefore recommend adding a word of caution to these numbers.

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