

SYNERGISTIC USE OF LOTOS-EUROS MODEL AND OMI NO₂ TROPOSPHERIC COLUMN TO EVALUATE THE NO_x EMISSION TRENDS OVER EUROPE

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2 Methodology

Abstract – Emission estimates from inverse modelling has drawn a lot of attention in the recent years. In this study we use OMI tropospheric NO₂ columns to set up an inverse modeling method and estimate the NO_x emission trends over Europe. In this paper, a case study to test the method and evaluate the scaling factors for the emission is presented. Currently, the inversion method has been tested on a small test period and preliminary results seem satisfactory. However, Interpretation of the results remains difficult and arte still under investigation.

1 Introduction

The quality of available information on the emission of atmospheric pollutants is an important factor to represent the current state and predict future changes of the atmospheric composition. Nitrogen oxides play a key role in atmospheric chemistry, indeed they are the precursors of tropospheric ozone, aerosol nitrate and of the hydroxyl radical. In 2007, the fourth assessment report of the IPCC, [IPCC, 2007] estimated that two third of the global Nitrogen oxides (NO_x) is from fossil fuel combustion and is mainly located in urban areas (over Europe this percentage is close to 90%). The goal of this study is to estimate the trends of NO_x emission in Europe.

The short lifetime of NO_x combined with the heterogeneous distribution of its sinks and sources imply that the NO_x tropospheric concentration is highly variable in both space and time. Recent studies using spaceborne instruments have pointed out that the tropospheric column of nitrogen dioxide contains valuable information about its sources, transport and sinks (Martin et al 2003,2006) and a clear signature at the city scale (Beirle et al 2003). The timeseries of nitrogen dioxides (NO₂) derived from satellites instruments have also been used to study long-term changes in anthropogenic emissions of NO_x. Hence, a general consensus has been reached that satellite remote sensing provides viable mean to measurement-based characterization of NO_x and NO₂ on a regional to global scale

In this study, we aim to develop an efficient method to estimate the change in NO_x emission during recent years using data-assimilation of satellite tropospheric NO₂ measurements in the LOTOS-EUROS chemistry transport model.

The tropospheric NO₂ columns retrieved from OMI measurements are combined with the LOTOS-EUROS chemistry transport model using an Ensemble Kalman Filter (EnKF). With this system a multi-year analysis is performed using a constant a priori NO_x emission database. The difference observed in the NO_x emission scaling factors resulting from the EnKF should represent the interannual changes in the NO_x emissions. Figure 1 is a schematic description of the inversion method.

LOTOS-EUROS [Schaap et al., 2007] is an operational 3D chemistry transport model which simulates the air pollution in the lower troposphere over Europe.

The OMI tropospheric NO₂ column product has been validated in several studies [Boersma et al, 2008, 2009b]. More recently a comparison of OMI NO₂ tropospheric columns with an ensemble of global and European regional air quality models was performed (Huijnen et al, 2010).

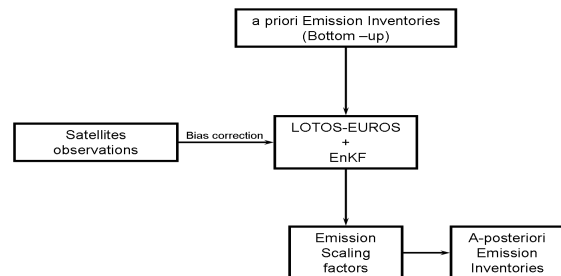


Figure 1: Schematic description of the protocol

Figure 2 provides box whisker plots of the tropospheric NO₂ column product over Europe retrieved from OMI measurements. The bottom and top of the boxes represent the 25th and 75th percentile (the lower and upper quartiles, respectively), and the line near the middle of the box represents the median. In this study we decided to use the whisker to illustrate the 5th and 95th percentile. Eastern Europe presents an obvious annual pattern with high NO₂ concentrations in the winter months and low values for the summer. The transition between winter and summer is less obvious for Western Europe.

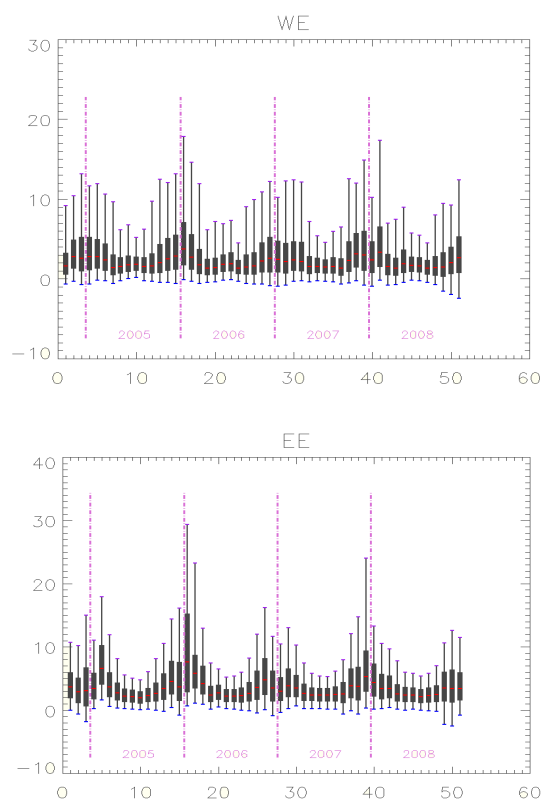


Figure 2: Box whisker plot of the NO_2 column from the DOMINO product over Western Europe (upper panel), Eastern Europe (Lower panel) for October 2004 to December 2008.

3 Preliminary results

Figure 3 represents the composite map of the NO_2 column retrieved and modeled (data have been plotted solely for the OMI pixel locations) for March 27th, 2007. The NO_2 columns from OMI and the free model run show good agreement. Similar hotspot patterns are observed. Both composite maps show a strong horizontal gradient with high NO_2 in the vicinity of highly industrialized and/ or densely populated areas such as German-Ruhr or the Rotterdam-Antwerp areas and low values around remote places. However, OMI tends to overestimate the tropospheric NO_2 column with respect to LOTOS-EUROS. Recently, a comparison study between the OMI NO_2 product and 11 regional air quality models (Huijnen et al, 2010) has shown a similar tendency for OMI to overestimate the tropospheric NO_2 column compared to the models by 27% in winter and up to 45% in summer, although the spatial pattern retrieved by OMI agreed with all the regional air quality models. This is also in agreement with Boersma et al [2008] where it is shown that the current OMI tropospheric NO_2 column is biased high up to 40% in polluted regions and especially in summer. Currently improvements are being made to improve the DOMINO product

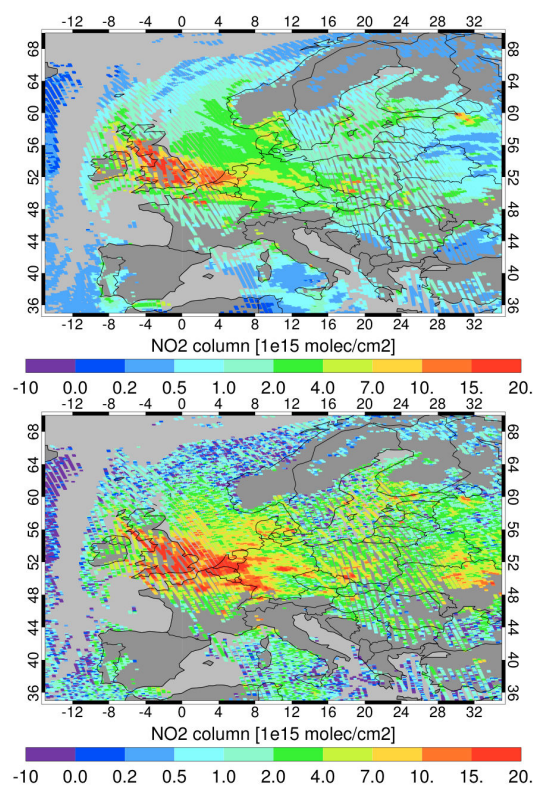
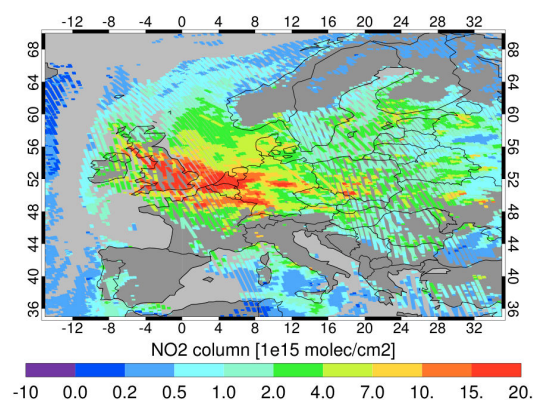


Figure 3: Composite map of the NO_2 column modeled by LOTOS-EUROS (Upper panel), and retrieved by OMI (Lower Panel) over Europe for March 27th 2007.

Figure 4 illustrates the output of LOTOS-EUROS using the EnKF assimilation for the NO_2 tropospheric columns from OMI. The sole purpose of this run was to test the system. To this end the uncertainty on the OMI NO_2 tropospheric columns was set to be really low (50% of the real error) to allow a strong pull of the model toward the measurement. The upper panel is a composite map of the NO_2 tropospheric columns while the lower panel represents the scaling factor for the NO_x emissions i.e. the perturbation applied to the emissions in order to reproduce NO_2 tropospheric columns retrieved from OMI measurements.



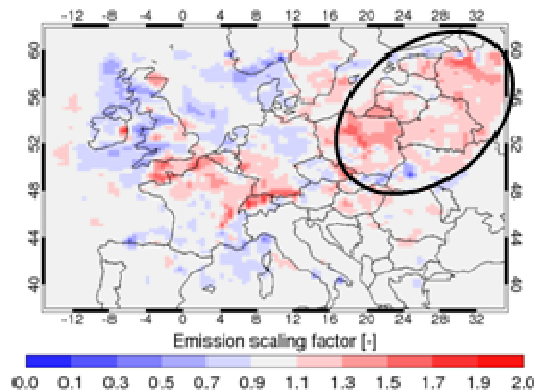


Figure 4: Europe for March 27th 2007. Upper Panel: Composite map of the NO₂ column simulated by LOTOS-EUROS after assimilation of OMI NO₂ column. Lower Panel: Composite map of the scaling for the NO_x emissions.

In order to reproduce the tropospheric NO₂ column retrieved from OMI the emissions were scaled with a factor up to 2. In general, the emissions over highly industrialized regions such as the German-Ruhr or the Rotterdam-Antwerp areas were increased while the emissions over remote regions were decreased. An important, increase of the emission is observed over Eastern Europe and was considered as unrealistic. After, further investigation it appears that this correspond to a contribution from the free troposphere which is not accounted for by LOTOS-EUROS. To overcome this artifact, a study is being carried out to investigate the best way to introduce the free troposphere concentrations from another chemical transport model into the LOTOS-EUROS model.

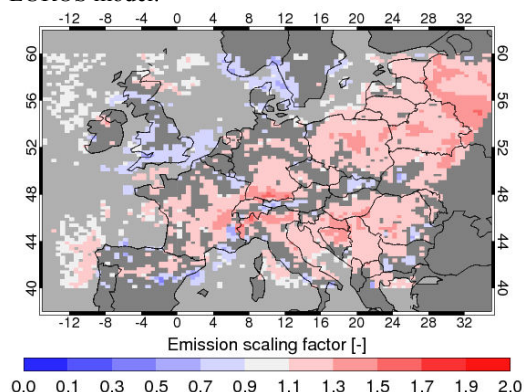


Figure 5: Composite map of the average scaling factor for the NO_x emissions over Europe for March 23rd to April 29th, 2007.

Figure 5 represents the composite map of the average scaling factor for the NO_x emissions over Europe for March 23rd to April 29th, 2007. We observe that for the period of interest the NO_x emission have been predominantly increased by a factor up to 2 in order to reproduce the OMI NO₂ tropospheric columns. The scaling factors present for Eastern Europe unrealistic pattern which has been attributed to NO₂ from free troposphere. Indeed LOTOS-EUROS at the moment does not account for it whereas the OMI NO₂ columns are contaminated by the NO₂ from the free troposphere. At the moment a method to account for this contribution is being implemented into LOTOS-EUROS

4 Outlook

The inversion method described here is a promising methodology to overcome the complexity and uncertainties associated to the bottom-up emissions estimates. Remote sensing observations can provide a top-down constraint which allow for a better estimate of the emissions and consequently of the emission trends.

In a following step, we expect to apply this method to the period spanning from October 2004 to December 2009 and subsequently assess the NO_x emission trends for these recent years.

5 References

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