Chapter 41
Synergistic Use of LOTOS-EUROS and NO2 Tropospheric Columns to Evaluate the NOX Emission Trends Over Europe

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Abstract The NOx-emission trend has been evaluated over Europe using the LOTOS-EUROS model and the NO2 tropospheric columns from OMI.

Keywords LOTOS-EUROS • Emission trends • NO2

41.1 Introduction

The quality of available information about sources (emissions) of atmospheric pollutants is a key parameter in any attempt to represent the current state or predict the future changes of the atmospheric composition. Besides natural sources and biomass burning, current estimates show that fossil fuel combustion is responsible for about 90 % of the total NOx emissions over Europe. In situ measurements in polluted areas have shown that the boundary layer contains more than two third of the tropospheric NO2. Hence, satellite remote sensing providing tropospheric NO2 columns is a suitable answer for the monitoring of pollution. Recent studies using spaceborne instruments have illustrated that the tropospheric column of nitrogen dioxide contains valuable information about its sources, transport and sinks. NO2 timeseries derived from satellites instruments have also been used to study long-term changes in anthropogenic emissions of NOx. The goal of this study is to estimate the trends in NOx emissions in Europe and subsequently identify the source sectors responsible. To this end we investigate the possibility to assess the trends in anthropogenic NOx emissions over Europe at high spatial resolution

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using OMI observation and the LOTOS-EUROS chemistry transport model. The study takes place within the framework of the ENERGEO and GlobEmission projects which respectively aims to set-up monitoring system of air pollutants using earth observation data and contributes to the verification and improvement of the UNECE/EMEP emission inventory over Europe.

41.2 NOx Emission Source Apportionment

In 2005, the anthropogenic NOx emissions over Europe were dominated by combustion processes in road transport with a 40 % share, followed by power plants, industry, off-road transport and the residential sector. In the last decades the abatement strategies over Europe have principally targeted combustion processes by power generation, road transport and industries. Moreover, it is anticipated that future changes in NOx emissions will be driven by certain sectors. As the OMI satellite observes the total column at local overpass time, It is important to know the sensitivity of the OMI signal to emission changes and investigate whether OMI is suitable to observe such trends over Europe. This is especially relevant considering the short life time of NOx in the atmosphere and the large variability of the temporal (or diurnal) variation in emission strengths for the different source sectors. To this end, the LOTOS-EUROS chemical transport model was ran for 2005 over Europe using the TNO-MACC emission inventory [4] for 2005. The model includes a new source apportionment module that tracks the contribution of specified sources through each modelled process and thus through the whole simulation. This means that for all oxidised nitrogen components at every time step and each grid cell the origin is calculated. Here the emissions were categorized per source sector and hour of emission. As the life time of NOx is short, it was chosen to focus the resolution for the time of emission to the morning hours prior to OMI overpass.

Figure 41.1 presents the modelled contribution per source sector and emission hour over two industrialized area in Europe at OMI overpass time (i.e. 1.30 p.m. local time). At first glance it is observed that 50 % of the OMI signal results from NOx emissions in the 3 h prior to OMI overpass. Besides, it can be observed that over the heavily industrialized area like the BENELUX the NOx emission are mainly driven by combustion processes in the road transport whereas over the Northern Iberian Peninsula the combustion processes in road transport energy sector and non-road transport have an equal share.

41.3 Trends Analysis

The tropospheric NO2 columns retrieved from OMI measurements, DOMINO [1, 2], are used in synergy with the LOTOS-EUROS [5] chemistry transport model. LOTOS-EUROS is an operational 3D chemistry transport model which simulates

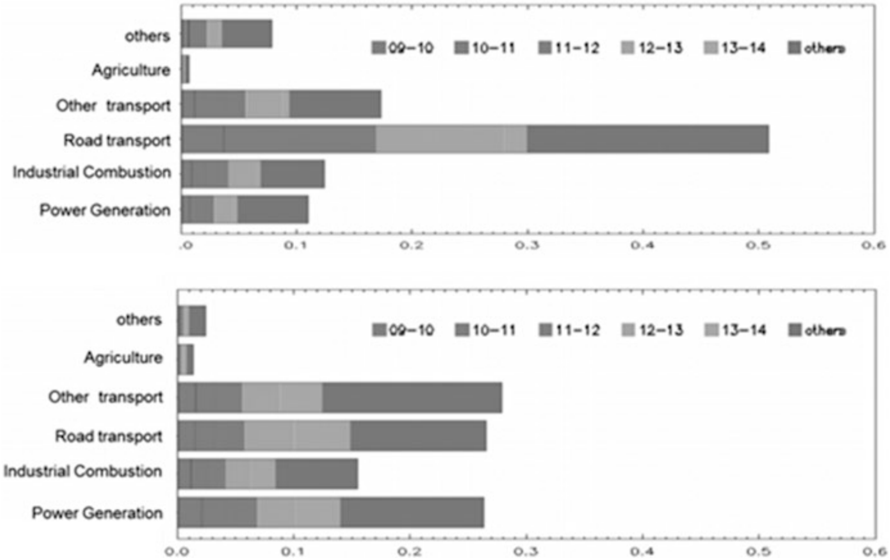

Fig. 41.1 Barchart of the contribution of various source sectors for different emission time to the total NOx emission

the air pollution in the lower troposphere over Europe. The OMI tropospheric NO¬2 column product has been validated in several studies [1, 2] and recently a comparison of OMI NO2 tropospheric columns with an ensemble of global and European regional air quality models was performed [3]. This last study concluded that the vertical column of the regional air quality models ensemble median shows a spatial distribution which agrees well with the tropospheric OMI NO2 tropospheric columns. The discrepancies observed between the tropospheric OMI NO2 tropospheric column and its modelled counterparts should occur mainly in the higher part of the atmosphere due to a lack of sources such as lightning and incorrect estimates of the lifetime of NO2 at higher altitude. In this study, a multi-year simulation is performed for 2005–2010 by LOTOS-EUROS using a constant a priory NOx emission database [4], to model the NO2 tropospheric columns at the overpass location and time of OMI. As we are using a fixed NOx emission database the inter-annual and seasonal variability present in these columns will solely represent the changes in the meteorology, transport and chemistry. In a second step the NO2 tropospheric columns from both OMI and LOTOS-EUROS were meshed into a 0.25×0.5 grid and for each pixel the time series of the monthly mean NO2 tropospheric columns are fitted using a model with a linear trend and a seasonal component which accounts for the annual cycle of NO2 [8]. This model has been validated for mean NO2 tropospheric columns using GOME and SCIAMACHY data at a global scale and over China [6, 7]. Figure 41.2 shows an example of a measured time series and the fitted function. The monthly averaged NO2 tropospheric column is plotted as a function of the month number starting in

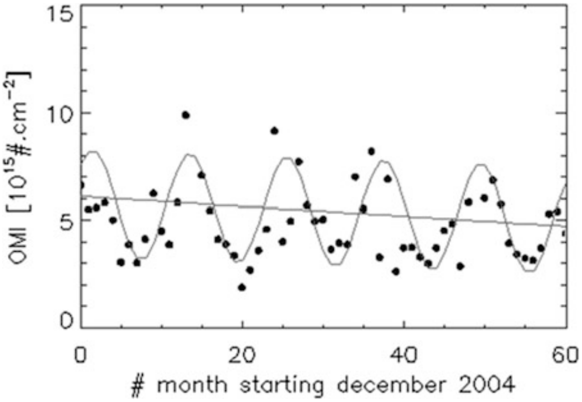

Fig. 41.2 Example of a timeseries for one grid cell near Paris (France). The Y axis shows the monthly mean NO2 tropospheric column, and the X axis shows the month index starting January 2005. The *dots* represent the measurement while the *straight* and *sinusoidal lines* represent respectively the linear decrease and seasonal component of the fitting result

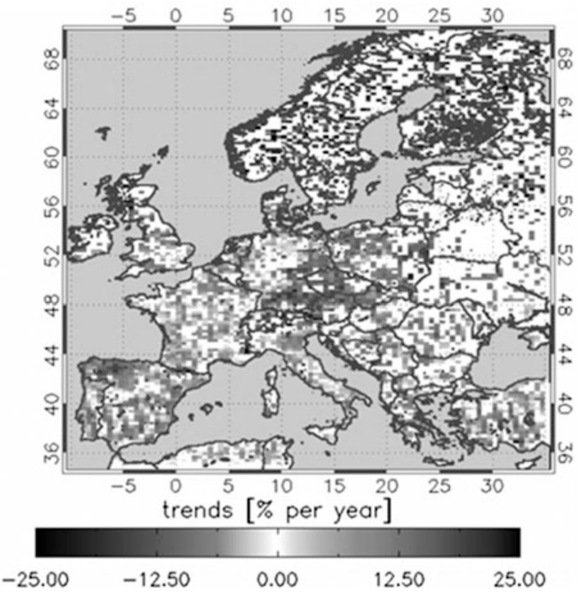

Fig. 41.3 Linear trends per year for tropospheric NO2 for 2005–2009 derived from OMI observations over Europe

January 2005. The fitting model is applied to each grid cell over Europe, which allow for a spatial distribution of each of the fitting parameters. Figure 41.3 presents the annual linear trends per year for tropospheric NO2 for 2005–2010 derived from OMI observations over Europe due to change in the NOx emission. A clear negative trend is observed over Europe. Significant negatives trends, ranging between −8 and −4 % are found over the dominant anthropogenic source regions of Western Europe such as Western Germany, Benelux, Po-Valley. The largest decreases coincide with well-known industrialized areas such as the Benelux, the Po-Valley in Italy and the Ruhr area in Germany. This can be explained by the fact that over Europe the decrease of NOx emissions is mainly due to a better control of road transport and

Table 41.1 Comparison table for the average trends (% per year) derived for three heavily industrialized area over Europe

	Area		
Study	Benelux	Po Valley	Northern Spain
Konovalov et al. [4]	−3.7	−2.7	−1.3
Zhou et al. [9]	−6.	−6.	−20 to −10
This study	−4.1	−6.1	−7.5 with hotspot −20

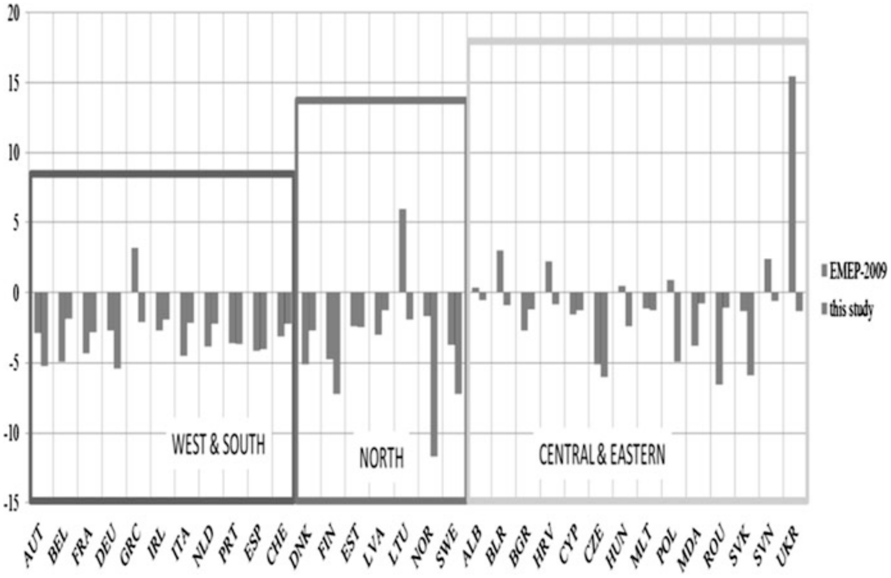

Fig. 41.4 Barchart of the yearly trends derived per country within the framework of this study (*left bars*) and the trends derived from officially reported data by countries (*right bars*)

power plants. A comparison of our results with recent trend studies using satellite is presented Table 41.1 for the BENELUX, Po Valley and Northern Spain. In general, a good agreement is observed. However, discrepancy occurs between the study carried out by Konovalov et al. [4] and the other two studies for Northern Spain. In fact, the strongest decreases occurred over Northern Spain (−20 to −10 %) and we believe it to be strongly related to emission abatement strategies targeting power plants implemented since beginning of 2008.

The trends derived for each country from this study are presented in Fig. 41.4 and compared to official reported data by countries. In general, the reported and retrieved trends are of similar amplitudes and providing confidence in the trends deduced in this study. However, a dichotomy between European countries appears and the reported and estimated NOx emissions tends compare better over Western Europe than over Central and Eastern Europe. This dichotomy may illustrate the differences between countries in re-updating their emission inventories but may also reflect that the dominant NOx source in Western Europe is road transport with a well-documented and, here confirmed, decrease in emissions due to implementing new technologies, whereas trends in other source sectors in eastern Europe are less known. The difference in sensitivity of the OMI instrument to source sectors may also play a role here and is under investigation. Besides, the large differences observed for Finland, Norway and Sweden are believed to be due to surface reflectance contamination and are under further investigations.

41.4 Conclusion and Outlook

A trend was derived from synergistic use of LE and OMI NO2. The results were in agreement with recent studies and reported emission inventories per country from EMEP. Significant negative changes were found in highly industrialized areas over Western Europe i.e. ∼5–6 %/year. Strong decrease in were observed over a regions with many power plants in Northern Spain (up to 20 %) and over the Po-Valley (∼11 %). The method described here is a promising methodology to complement and evaluate trends in bottom up emission reportings. A strong point is the fact that the methodology using satellite date is in principle consistent throughout the entire domain. Besides, satellite data are available in near real time. Remote sensing observations can therefore provide a top-down constraint which allows for a near real time estimate of the emissions.

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References

1. Boersma KF, Jacob DJ, Bucsela EJ, Perring AE, Dirksen R, van der A RJ, Yantosca RM, Park RJ, Wenig MO, Bertram TH, Cohen RC (2006) Validation of OMI tropospheric NO2 observations during INTEX-B and application to constrain Nox emissions over the eastern United States and Mexico. Atmos Environ 42(19):4480–4497. doi:10.1026/j.atmosenv.2008.02.004, 2008. 4304, 4309
2. Boersma KF, Jacob DJ, Trainic M, Rudich Y, DeSmedt I, Dirksen R, Eskes HJ (2009) Validation of urban NO2 concentrations and their diurnal and seasonal variations observed from space (SCIAMACHY and OMI sensors) using in situ measurements in Israeli cities. Atmos Chem Phys 9:3867–3879
3. Huijnen V, Eskes HJ, Amstrup B, Bergstrom R, Boersma KF et al (2010) Comparison of OMI NO2 tropospheric columns with an ensemble of global and European regional air quality models. Atmos Chem Phys 10:3273–3296
4. Konovalov IB, Beekmann M, Burrows JP, Richter A (2008) Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions. Atmos Chem Phys 5(8):2623–2641

5. Kuenen J, van der Gon D , Visschedijk AH, van de Brugh H, van Gijlswijk R (2011) MACC European emission inventory for the years 2003–2007. Technical Report. report TNO-060-UT-2011-00588. TNO, Utrecht, the Netherlands
6. Schaap M, Renske MA, Timmermans MR, Boersen GAC, Builtjes PJH, Sauter FJ, Velders GJM, Beck JP (2007) The LOTOS–EUROS model: description, validation and latest developments. Int J Environ Pollut (IJEP) 32(2)
7. van der A RJ, Peters DHMU, Eskes H, Boersma KF, Van Roozendael M, De Smedt I, Kelder HM (2006) Detection of the trend and seasonal variation in tropospheric NO2 over China. J Geophys Res 111:D12317. doi:10.1029/2005JD006594
8. van der A RJ et al (2008) Trends, seasonal variability and dominant NOx source derived from a ten year record of NO2 measured from space. J Geophys Res 113:D04302. doi:10.1029/2007JD009021
9. Zhou Y, Brunner D, Hueglin C, Henne S, Staehelin J (2012) Changes in OMI tropospheric NO2 columns over Europe from 2004 to 2009 and the influence of meteorological variability. Atmos Environ 46:482–495. doi:10.1016/j.atmosenv.2011.09.024

Questions and Answers

Questioner Name: W. Lefebvre

Q: How does the changing NO2/NOx ratio affect your results?.
A: The downward trend observed in NOx emissions partly resulted in the development of new technologies that influence the ratio. Changes in NO2/NOX ratios during the period of interest may result in additional uncertainty in NOX emissions.

Questioner Name: J. Silver

Q: The comparison between OMI and LOTOS-EUROS showed a seasonal bias. Is this mainly due to the model or the observations?
A: Many studies shown this bias between chemistry transport model and observation. The discrepancies observed between the tropospheric OMI NO2 tropospheric column and its modelled counterparts should occur mainly in the higher part of the atmosphere due to a lack of sources such as lightning and incorrect estimates of the lifetime of NO2 at higher altitude. On the other hand, comparison studies between OMI and ground based observations have also shown that OMI was biased high.