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Application of Satellite Observations and Air Quality Modelling to Validation of NOx Anthropogenic EMEP Emissions Inventory over Central Europe

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Abstract: One of the most important minor species in the atmosphere is nitrogen dioxide (NO_2). The primary objective of the presented research was to propose a method to adjust emission inventories (emission fluxes) using tropospheric NO_2 columns observed by OMI and SCIAMACHY instruments. Modified emission fluxes were used in a chemical weather model GEM-AQ. The GEM-AQ model results were compared with the monthly averaged satellite-derived column amount of NO_2 over Europe for the 2008–2010 observing period. It was shown that the observed and modelled spatial distribution of high values of the NO_2 column is highly correlated with the distribution of major anthropogenic sources in the modelling domain. The presented findings highlight the importance of the anthropogenic sources in the overall budget of NO_2 in the polluted troposphere. Regions for which modelling results showed underestimation or overestimation compared with observations were constant for the whole analysis period. Thus, the NO_2 column observations could be used for correcting emission estimates. The proposed emission correction method is based on the differences in modelled and satellite-derived NO_2 columns. Modelling was done for 2011 using the original and adjusted emission inventories and compared with observed NO_2 columns. The analysis was extended to compare modelling results with surface NO_2 observations from selected air quality stations in Poland. A significant improvement in modelling results was obtained over regions with large overestimations in the control run for which the original emission fluxes were used.

Keywords: SCIAMACHY; OMI; NO_2 column; GEM-AQ

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1. Introduction

Nitrogen dioxide (NO_2) is one of the most important minor species in the Earth's atmosphere. Emissions of NOx (NO_2 + NO) are estimated, and concentrations are measured by national environmental agencies. Observations and modelling results allow us to assess the impact of air pollution on the environment, specifically on human health [1–7].

Nitrogen oxides (NO_2 and NO) in the atmosphere are oxidised, and their lifetime is several hours [8], resulting in high spatial and temporal variability. High concentrations are mainly limited to the boundary layer, i.e., about 1–2 km, with a rapid decline at higher altitudes. As a result of the decreasing temperature, the decrease is explicit for NO_2 due to the declining NO_2/NO ratio [9].

The development and application of satellite-based remote sensing methods allow monitoring atmospheric chemical composition [10]. NO_2 is among several chemical constituents that can be measured using space-borne instruments such as GOME (Global Ozone Monitoring Experiment), GOME-2 (Second Global Ozone Monitoring Experiment), SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY), OMI (Ozone Monitoring Instrument), and TROPOMI (TROPOspheric Monitoring Instrument).

Over the last decade, observations of NO_2 tropospheric columns were often used to validate air quality model results in the regional and local scales. Many publications focused on identifying regions with significant and systematic differences between satellite observations and modelling results. The identified differences were attributed to incorrect estimations of the emission fluxes provided in emission inventories used for model simulations [11–15].

Konovalov et al. [16] used satellite data from GOME [17] and SCIAMACHY [18] and a chemical transport model CHIMERE to improve inventories of NOx emissions over Western Europe. It was estimated that the uncertainty of the original (a priori) EMEP (European Monitoring and Evaluation Programme) emission inventory (www.emep.int/, accessed on 3 November 2021) for NO_2 was about 1.9 molec $\times$ cm^{-2} $\times$ s^{-1} $\times$ 10^{11}. The work of Blond et al. [19] showed that the combination of satellite and ground-based observations could improve air quality forecasts and allow for the correction of emission fluxes over Western Europe. Miyazaki et al. [20] presented a data assimilation system based on the NO_2 column derived from satellite observations that allowed estimating daily global emissions of NO_2 at the spatial resolution of 2.8 degrees.

Vinken et al. [21] compared the tropospheric column from the OMI [22] instrument with results from the GEOS-Chem model. The comparison showed a significant overestimation (about 60%) of emissions in the EMEP inventory for 2005 in the Mediterranean region that could significantly impact model simulations. For the North Sea, it was shown that the EMEP emission fluxes were overestimated by 35%. In comparison, the emission values were overestimated by 131% and 128% for the Baltic Sea and the Bay of Biscay, respectively. Kawka et al. [23] used satellite observation from Sentinel 5P to verify the location of large industrial point sources in Poland.

Tong et al. [24] examined the long-term trends of NO_2 concentrations based on satellite observations from OMI and ground observations. It was shown that a wide spatial range and the availability of near real-time satellite observations of NO_2 have significant potential to improve the quality of NOx emissions inventory used for air quality forecasting over the United States.

A similar study was conducted over Central Europe from 2008 to 2010 by Szymankiewicz et al. [25]. It was demonstrated that the spatial distribution of observed and modelled high values of the NO_2 column is highly correlated with the distribution of significant anthropogenic source regions. This relationship underlines the importance of the anthropogenic sources in the overall budget of NO_2 in the troposphere. Locations of regions for which modelling simulations gave underestimation or overestimation as compared to satellite observations were invariant for the whole analysis period. This finding was consistent with the hypothesis that the tropospheric NO_2 column was highly correlated with anthropogenic emissions over urban areas.

The presented work aims to continue analysis on the correction of emission inventories to improve model performance in NO_2 tropospheric columns and surface concentrations. The primary objective was to develop a method to modify emission inventories (strength of emission sources) using NO_2 column observations from two satellites, OMI and SCIAMACHY, and modelling results from a chemical weather model GEM-AQ (Global Environmental Multiscale model with Air Quality processes) [26].

2. Materials and Methods

The proposed emission correction method is based on the differences in modelled and satellite-derived NO_2 tropospheric columns assessed in the earlier study for 2008–2010 [25]. Model simulations were done for 2011 using the original and modified emission inventories to create an independent dataset. Modelling results from these two scenarios were compared with observed NO_2 tropospheric columns. The analysis was extended to compare model results with surface NO_2 observations from selected stations from Poland.

2.1. Emission Trend and Emission Correction Method

As a first step, the analysis of emission inventories was undertaken. Nitrogen oxides are mainly formed during high-temperature combustion processes. Transport, the energy production process, and the chemical industry contribute the most to anthropogenic emissions. The largest share of NO_X emissions in Europe (EU 27) is from road transport, energy production, municipal and domestic sources, and industry. Relative contributions from each sector may vary from country to country (Table 1). The relative contributions depend on the number and type of cars, industry, and fuel used in household combustion. In this paper, further analysis is focused on the Silesia region and the Netherlands.

Table 1. Contribution of NOx emissions by individual SNAP category (Selected Nomenclature for sources of Air Pollution).

No	SNAP Category	SNAP Description	EU27	Poland	Denmark	Netherland
1	01	Energy exploration and production	18%	33%	19%	13%
2	02	Commercial, intuitional and households	5%	10%	6%	12%
3	03	Energy in use	8%	9%	4%	11%
4	04 & 05	Industrial processes	3%	2%	0%	0%
5	06	Solvent and product use	0%	0%	0%	0%
6	07	Road transport	32%	33%	37%	38%
7	08	Non-road transport	33%	12%	34%	23%
8	09	Waste	0%	0%	0%	0%
9	10	Agriculture	1%	1%	0%	3%

For EU27 countries, a decreasing trend of NOx emission is estimated in the EMEP inventory (Figure 1). A considerable reduction was in the transport sector (over 30%) due to EURO norms and energy processes (25%) caused by emission regulations for large sources [27].

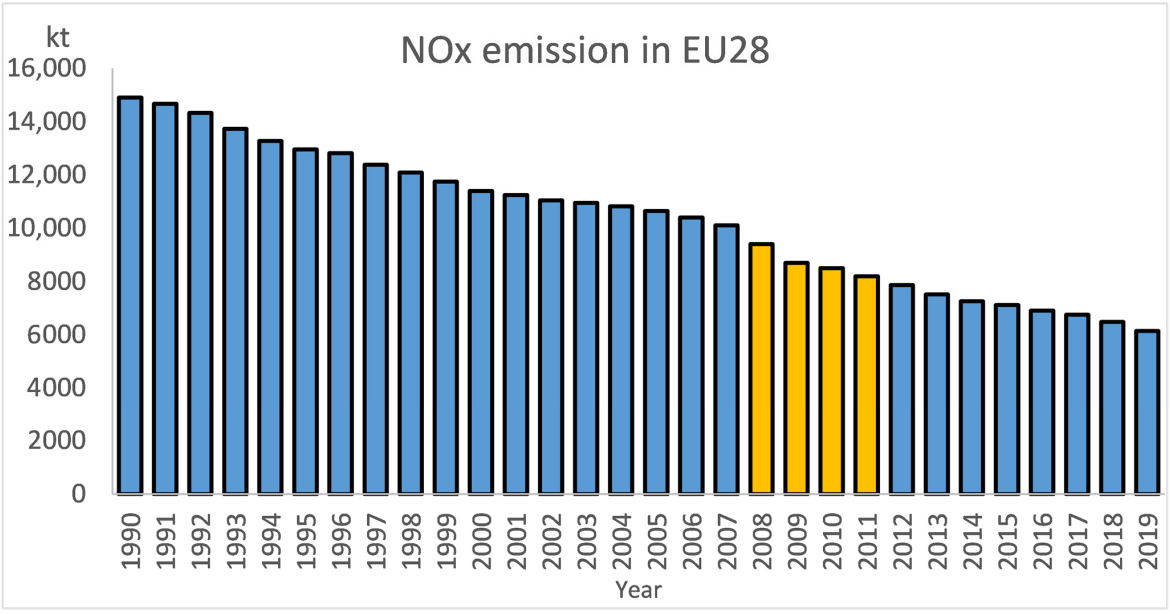

Figure 1. Emissions of nitrogen oxides NOx as NO_2 for EU28 from EMEP. Yellow colours of bars represent the years of analysis in the presented work, and blue bars represent other years.

In each sector for the 2008–2011 period, we observe a stable tendency, allowing the implementation of coefficients calculated based on differences between 2008 and 2010 as a

representative for 2011 (Figure 2). In the 2008–2010 period, the average emissions in the SNAP01 sector were 284 Gg. In the SNAP07 sector, the emissions were 273 Gg, and there was a slight decrease (about 1.1%) with respect to the total emission budget. Emissions from other vehicles and equipment (SNAP08) were approximately 102 Gg, which accounted for approximately 12% of the total emissions change from 2008 to 2010.

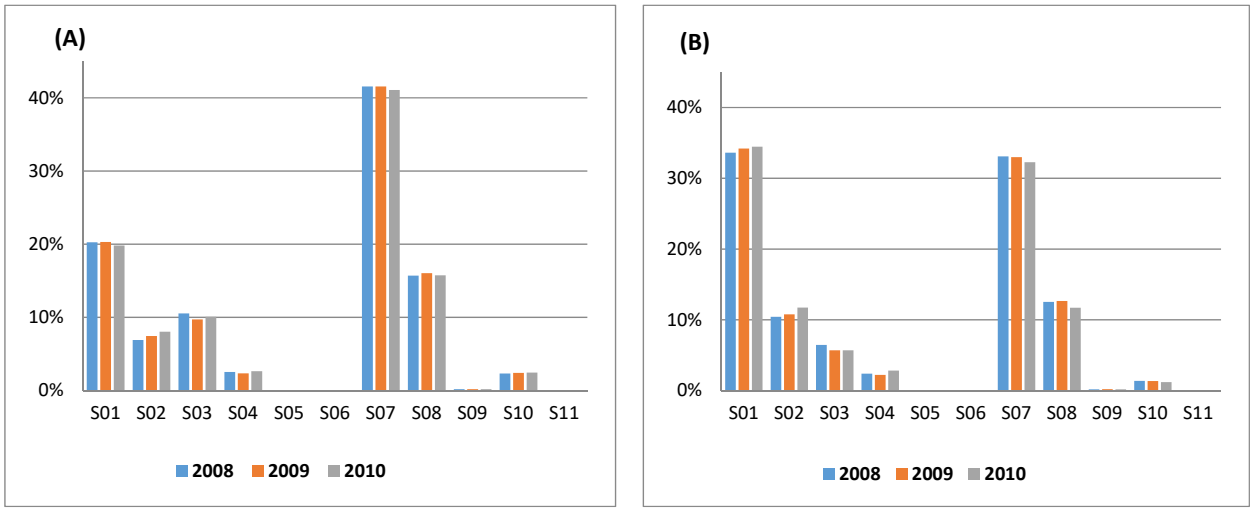

Figure 2. Emissions of nitrogen oxides NOx as NO_2 for EU28 (**A**) and Poland (**B**) divided into different SNAP97 sectors, based on the EMEP database for the period 2008–2010.

2.2. Model Configuration and Emission Processing

The GEM-AQ model configuration and emission inventory are based on the previous work by Szymankiewicz et al. [25]. The model was run on a global variable resolution grid. The uniform resolution of 0.125 deg (≈15 km) partially covered Central Europe (Figure 3). The model resolution in the core part was comparable to the resolution of the SCIAMACHY and OMI monthly products. The model top was set up at 10 hPa, with 28 layers defined in hybrid-sigma coordinate.

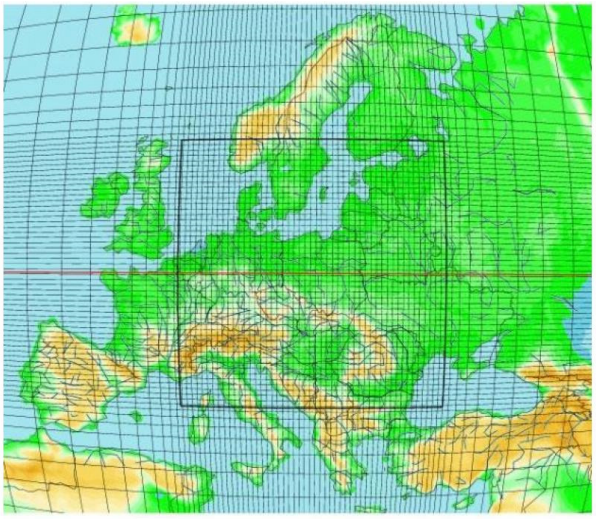

Figure 3. Core grid with a uniform resolution of 0.125 deg (red line—numerical equator).

The EMEP emission inventory prepared for Europe (www.emep.int/, accessed on 3 November 2021) at the resolution 0.5 × 0.5 deg (the study was conducted before the switch to EMEP grid 0.1 deg) was used. Emissions were relocated to the GEM-AQ model

resolution (0.125° × 0.125°) according to GIS (Geographical Spatial Information System) proxy data.

As a base for the temporal distribution of emissions in the GEM-AQ model, the daily and monthly time profiles from the CityDelta project were adopted [28]. Temporal profiles (slightly modified), depending on the SNAP category and the local time zone, were used in many European projects (e.g., COST728, AQMEII, EURODELTA) [29,30].

Although each SNAP97 sector has a specific time profile, the proposed method does not consider dynamic changes in the NOx emission fluxes.

In the GEM-AQ model, emissions for each sector are assigned to an effective plume height [31,32]. The vertical distribution of emissions indicates the height to which emission plum can be raised, considering the mixing processes occurring in the atmosphere [33,34].

2.3. Satellite Data

The average monthly tropospheric NO_2 column, at a resolution of 0.125° × 0.125° from SCIAMACHY (https://www.iup.uni-bremen.de/doas/scia_no2_data_tropos.htm/, accessed on 3 November 2021) and OMI (https://www.temis.nl/airpollution/no2.php/, accessed on 3 November 2021) spectrometers, were used in the analysis. The utilisation of average monthly products made it possible to obtain a very high coverage in the study area. It allowed for analysis of the monthly variability of the tropospheric NO_2 columns. In addition, the application of two independent data sources gave up to 6 satellite observations at each grid square for each month. Thus, it allowed for reducing areas with missing data due to cloud cover, mainly during winter.

2.4. Data from Surface Air Quality Monitoring Stations

For a comparative analysis of modelled NO_2 concentrations, 14 surface air quality stations from Poland were used. Only stations with a continuous and complete observational record in 2008–2011 for NO_2 were selected. Selected stations are located in Poland, where significant overestimations or underestimations of model results were found in [25] for 2008–2010. Station names, types, and locations are given in Table 2.

Table 2. Surface air quality stations used in the analysis.

No	Station Name	Station Type	Latitude	Longitude
1	Czerniawa	Background rural	16°56.52′ E	50°17.82′ N
2	Bory Tucholskie	Background rural	17°55.98′ E	53°39.00′ N
3	Widuchowa	Background rural	14°23.82′ E	53°10.32′ N
4	Krzyżówka	Background rural	22° 04.14′ E	54°09.00′ N
5	Granica KPN	Background rural	21°07.32′ E	49°38.64′ N
6	Szymbark	Background rural	15°16.02′ E	52°13.32′ N
7	Smolary Bytnickie	Background rural	19°33.84′ E	51°20.82′ N
8	Parzniewice	Background rural	16°25.32′ E	51°59.64′ N
9	Gajew	Background rural	19°13.98′ E	52°13.98′ N
10	Biały Słup	Background rural	22°59.76′ E	50°35.46′ N
11	Belsk	Background rural	20°52.02′ E	51°52.02′ N
12	Wrocław Bartnicza	Background suburban	17°08.46′ E	51°06.90′ N
13	Legionowo	Background suburban	20°30.84′ E	52°18.66′ N
14	Białystok	Urban	23°20.82′ E	53°13.32′ N

3. Results

To verify the developed emission correction factors, the GEM-AQ model was run for two scenarios in 2011. It should be underlined that emission correction factors were calculated using modelling results for the period 2008–2010 described in [25]; hence, the results obtained for 2011 were independent. The base emissions were used in the first scenario (BASE), and modified emissions were used in the second scenario (BIS).

Spatial analysis was carried out based on a monthly average of

- NO_2 concentrations in the lowest model layer; and

- Calculated tropospheric NO_2 column for the base scenario for 2011.

3.1. Emissions Correction Method

Emission corrections factors were calculated as differences between model results and observations of two spectrometers, OMI and SCIAMACHY, for 2008–2010. The ratio was calculated using six satellite observations and three model results (2008, 2009, 2010). The monthly average tropospheric NO_2 columns and the monthly average concentration of NO_2 in the lowest layer of the model were used in the analysis.

The following equation gives the method for calculating the correction factor for each grid cell. The summation is done for all available (n) satellite observations in a grid square.

$$F = \frac{GEM - \frac{\sum(SAT_{OMI} + SAT_{SCIA})}{n}}{GEM} \quad (1)$$

where

F—correction ratio;
GEM—tropospheric NO_2 column calculated by the GEM-AQ model;
SAT_{OMI}—tropospheric NO_2 column from OMI;
$SATS_{CIA}$—tropospheric NO_2 column from SCIAMACHY;
n—number of available satellite observations in a grid square.

The ratio was calculated from the largest possible number of satellite observations (up to six values). The use of two satellite instruments allowed for obtaining correction factors for almost the entire domain during the summer and for about 80% of the domain during the winter.

Figure 4 presents the spatial distribution of the emission correction factor for selected months in individual seasons. It was not possible to calculate the indicator, over the entire domain, for the winter months because of high cloud coverage. Negative values of the ratio indicate that the baseline emission should be increased, and the positive values of the calculated ratio indicate that the baseline emissions should be reduced. It should be noted that the negative values of the tropospheric NO_2 columns that are caused by using the DOMINO method [35] were not used in the calculations.

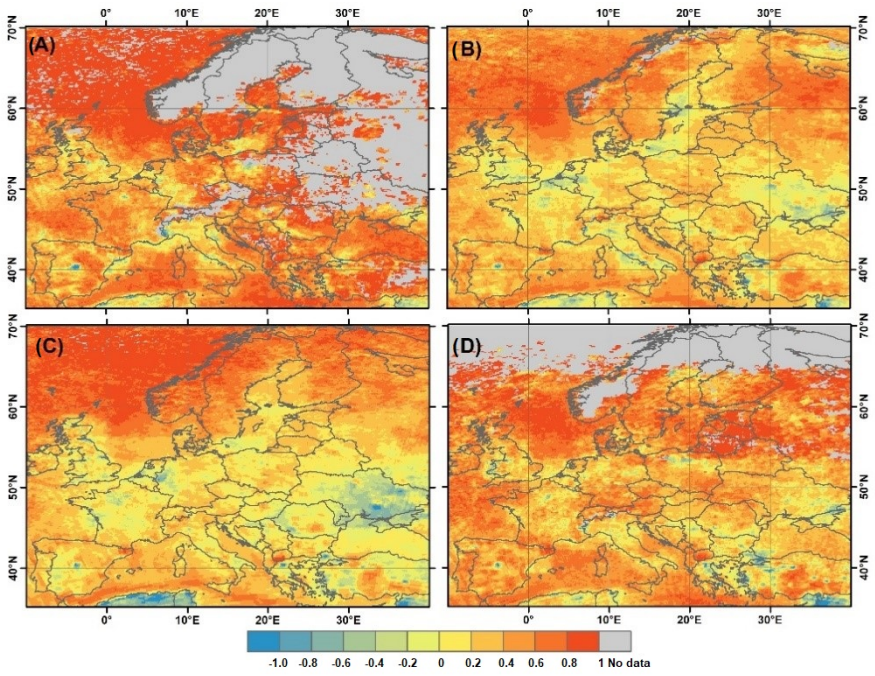

Figure 4. Correction ratio values (F) for NOx emission based on satellite data and GEM–AQ model results. (**A**) February, (**B**) May, (**C**) August, and (**D**) November.

The correction factor calculated based on the NO_2 tropospheric column was applied to NO_2 and NO emission fluxes. To process monthly emission fields, the annual emission budget from EMEP was monthly-distributed based on the temporal profile. Furthermore, the values of emission fluxes were modified by the correction factor, taking into account the magnitude of fluxes in each of the SNAP categories in a given grid square. Emission fluxes before and after modifications for July and December are shown in Figure 5.

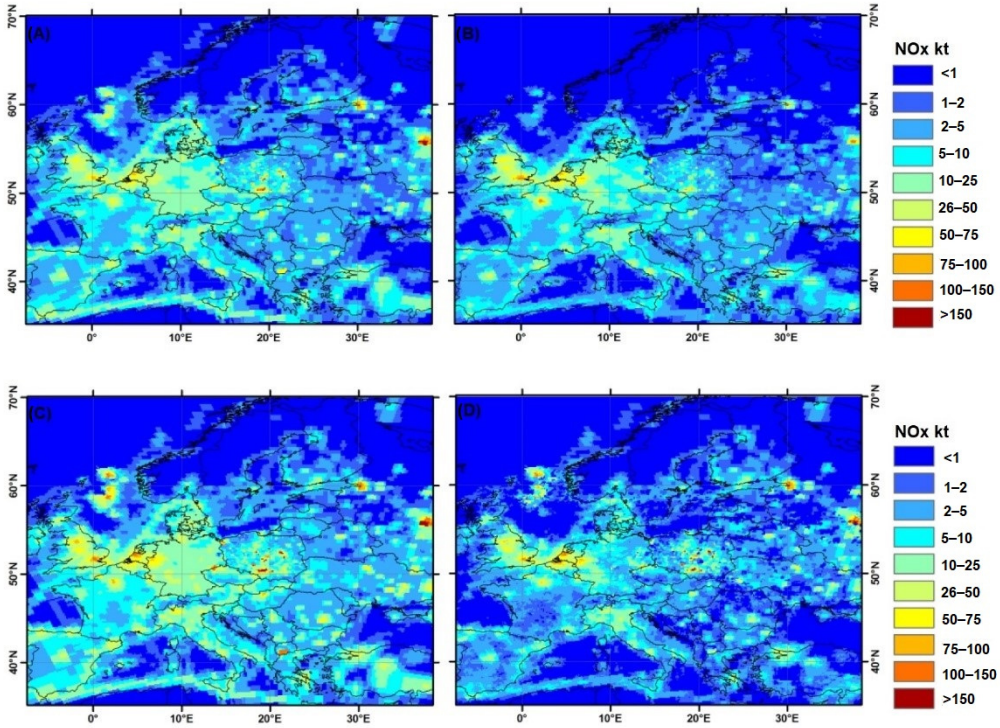

Figure 5. Comparison of NOx emission fluxes in 2010 for July (upper panels) and December (lower panels) before (left panels) and after (right panels) modifications.

After modification, more concentrated and higher fluxes of NOx emission were observed around agglomerations (average difference from 20 to 50 kt). Over the rural areas and the Mediterranean Sea, emissions were significantly lower. For the Mediterranean basin, the total emissions are reduced by almost half (from 51 to 28 Mt). In addition, some high values over inactive power plants have high decrees. For example, the region closest to the power plant in Bitola (Macedonia) registered a six-fold reduction (from 1000 to 150 kt) of the total emissions.

3.2. Modelled vs. Observed NO_2 Column

Figures 6–8 show a comparison between tropospheric columns observed by SCIAMACHY and OMI instruments for two simulations—with and without modification of NOx emissions for January, June, and August 2011, respectively.

In January, the spatial pattern is incomplete due to the cloud coverage, especially in northern and eastern Europe (Figure 8). Over the rest, a slight reduction of the positive bias can be found over the south of Europe: Spain, southern France, north Italy, and the Mediterranean Sea (from 10 to 25 $\times$ 10^{15} molec/cm^2). In the case of negative bias, no significant improvement was found.

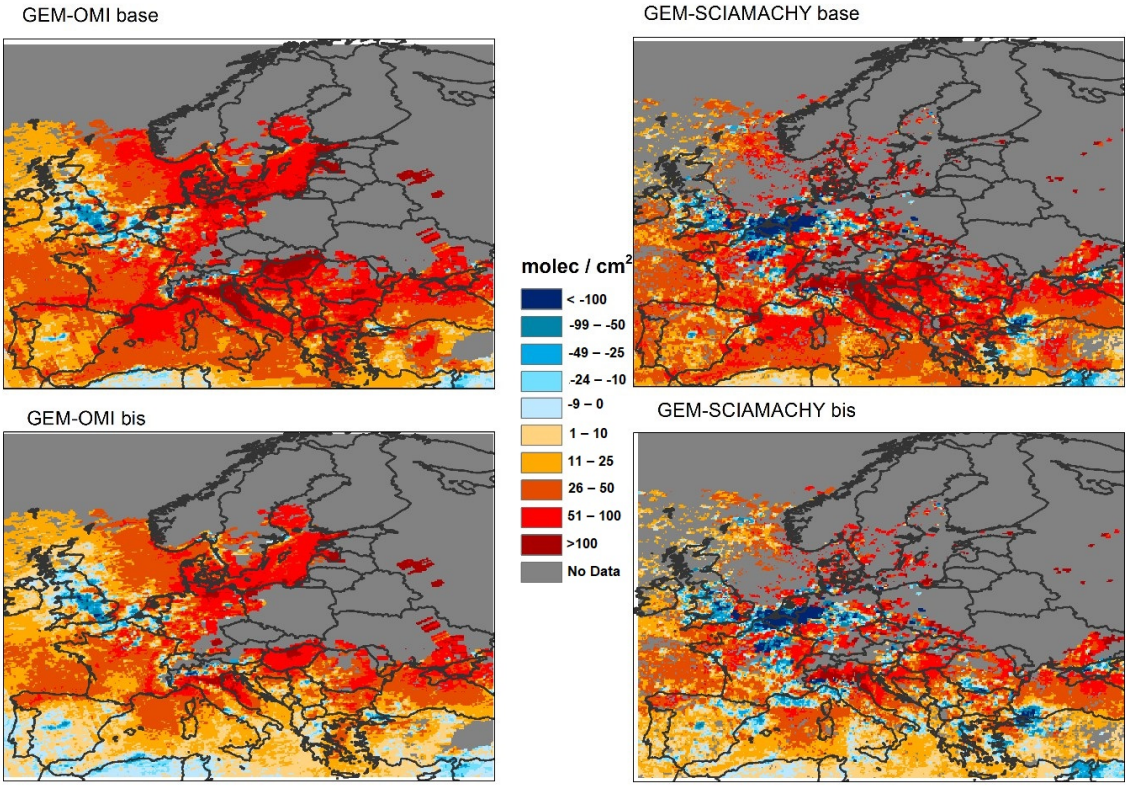

Figure 6. Differences (10^{15} molec/cm^2) in the tropospheric NO_2 columns derived from GEM-AQ for the baseline emissions (upper row) and the modified emissions (bottom row) and satellite observations (OMI left, right SCIAMACHY) for January 2011.

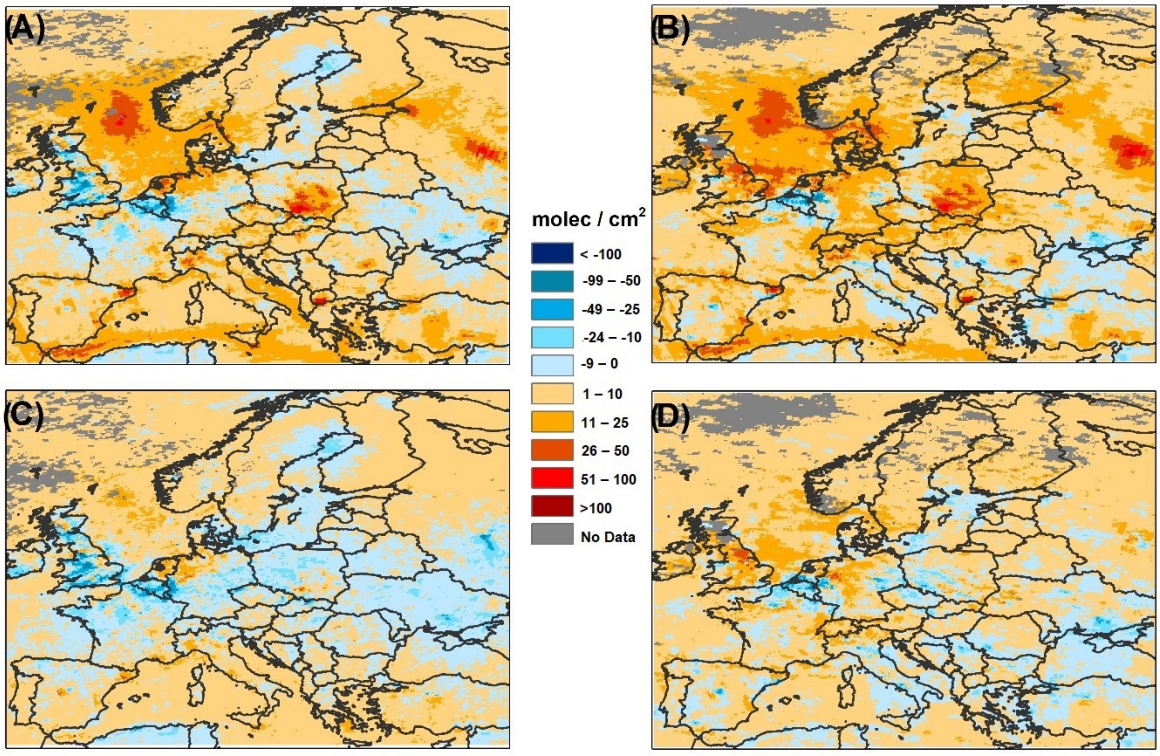

Figure 7. Differences (10^{15} molec/cm^2) in the tropospheric NO_2 columns derived from GEM-AQ for the baseline emissions (upper row) and the modified emissions (bottom row) and satellite observations (OMI left, right SCIAMACHY) for June 2011.

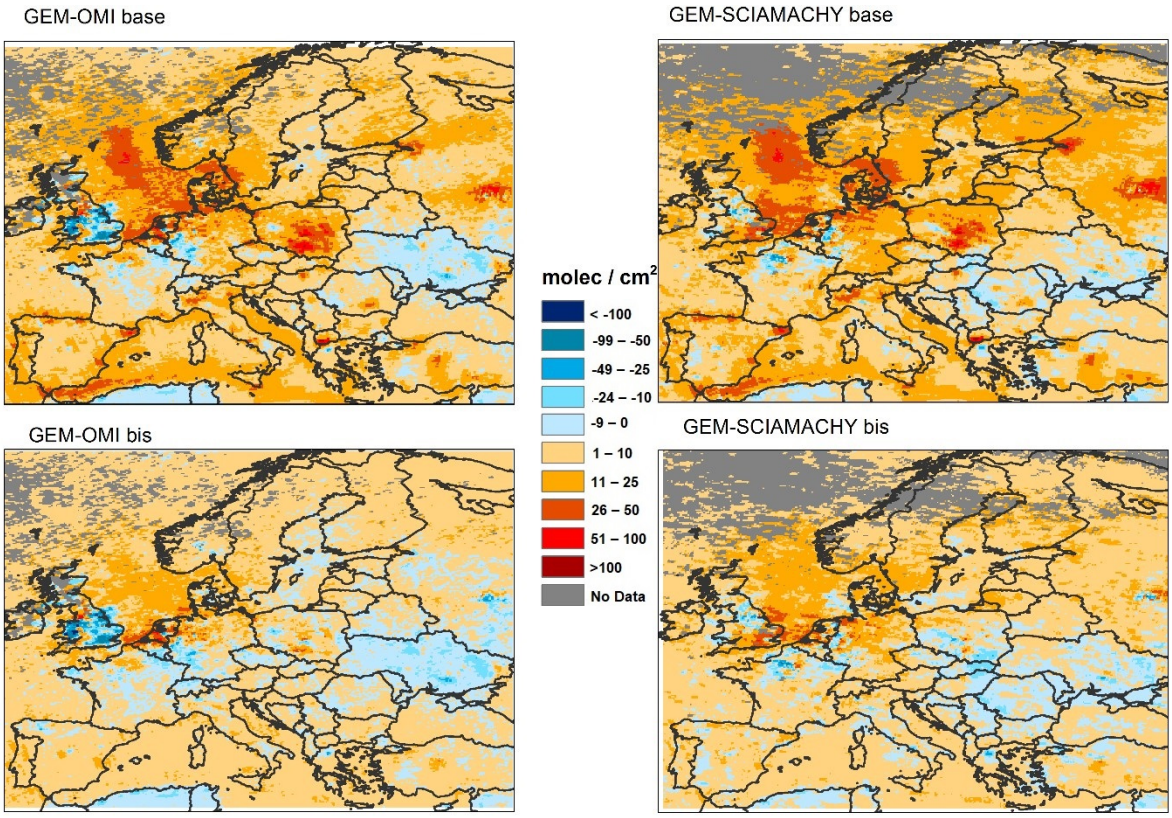

Figure 8. Differences (10^{15} molec/cm^2) in the tropospheric NO_2 columns derived from GEM-AQ for the baseline emissions (upper row) and the modified emissions (bottom row) and satellite observations (OMI left, right SCIAMACHY) for August 2011.

For June, the improvement is significant, especially in northern and central Europe. The overestimation over the North Sea, Poland, and the several agglomerations were reduced (from over 100 to 10–20 $\times$ 10^{15} molec/cm^2), sometimes resulting in a slight underestimation (about 10–20 $\times$ 10^{15} molec/cm^2). Similar to January, no significant changes in bias were detected for regions in western Europe characterised with underestimation.

For August, the general pattern of the differences between model and satellite observation was similar to those obtained for June.

Although in areas with significant underestimations, i.e., in the Benelux countries, an improvement of modelling results was not achieved. However, the underestimation was reduced from about 100 $\times$ 10^{15} molec/cm^2 to approximately 40 $\times$ 10^{15} molec/cm^2.

3.3. Chemical Regime

The insufficient increase of the modelled NO_2 column value despite the significant increment of the emission flux may be related to the non-linearity between the concentration of NO_2 and other photochemical species in the atmosphere and the ozone concentrations. An analysis of photochemical regimes was carried out to verify this hypothesis. In the scientific literature, the concept of chemical regimes refers mainly to near-surface ozone concentrations [36–38]. However, since NOx is a major ozone precursor, and the highest NOx concentrations are detected in the lowest 2 km (e.g., [8,39]), a regime-based concept was developed to analyse differences in chemical regimes in terms of the spatial pattern over Europe.

An indicator based on the ratio of O_3 and NO_2 was proposed. The partial columns of ozone (O_3) and nitrogen dioxide (NO_2) were calculated for the seven lowest model layers, i.e., up to about 1.400 m, representing the average height of the atmospheric boundary layer. The partial column ratio O_3/NO_2 was calculated for each month.

Figure 9 shows the average differences between the modelled tropospheric NO_2 and the satellite observations averaged for two instruments for both simulations, as well as the O_3/NO_2 partial column ratio for the boundary layer for June 2011.

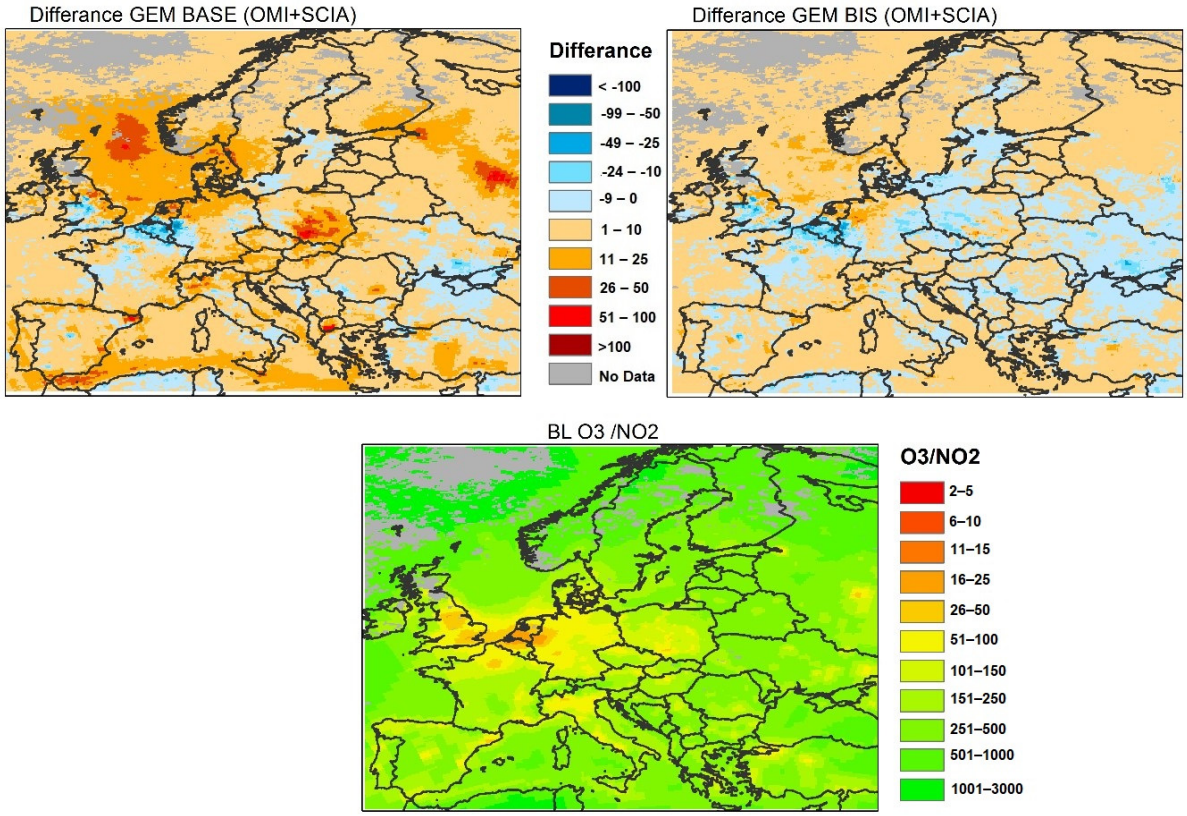

Figure 9. The difference between NO_2 tropospheric columns derived from the model results and observations from satellite (OMI + SCIAMACHY) for the Base and Bis scenarios and ratio O_3/NO_2 column for June 2011.

The photochemical regime index was the lowest over the region, with the strongest NO_2 column underestimation for all months. This may indicate that in NOx-saturated regions, the differences between models and observations are not sensitive to the relatively small emissions perturbations and should be further analysed in the contacts of the photochemical cycle formulation.

3.4. Modelled NO_2—Tropospheric Column and Surface Concentrations

Although the tropospheric NO_2 column does not fully reflect the near-surface emission sources, the regions for which the highest surface concentrations of NO_2 were observed (over 40 $\mu g/m^3$) coincide with the areas characterised by the highest values (over 100×10^{15} molec/cm^2) of the tropospheric NO_2 column calculated from model results. This relationship is particularly evident in the winter months. Surface NO_2 concentration and NO_2 column calculated from model results are high mostly, over large European agglomerations (e.g., Moscow, Paris, Madrid) and the Benelux countries (Figure 10).

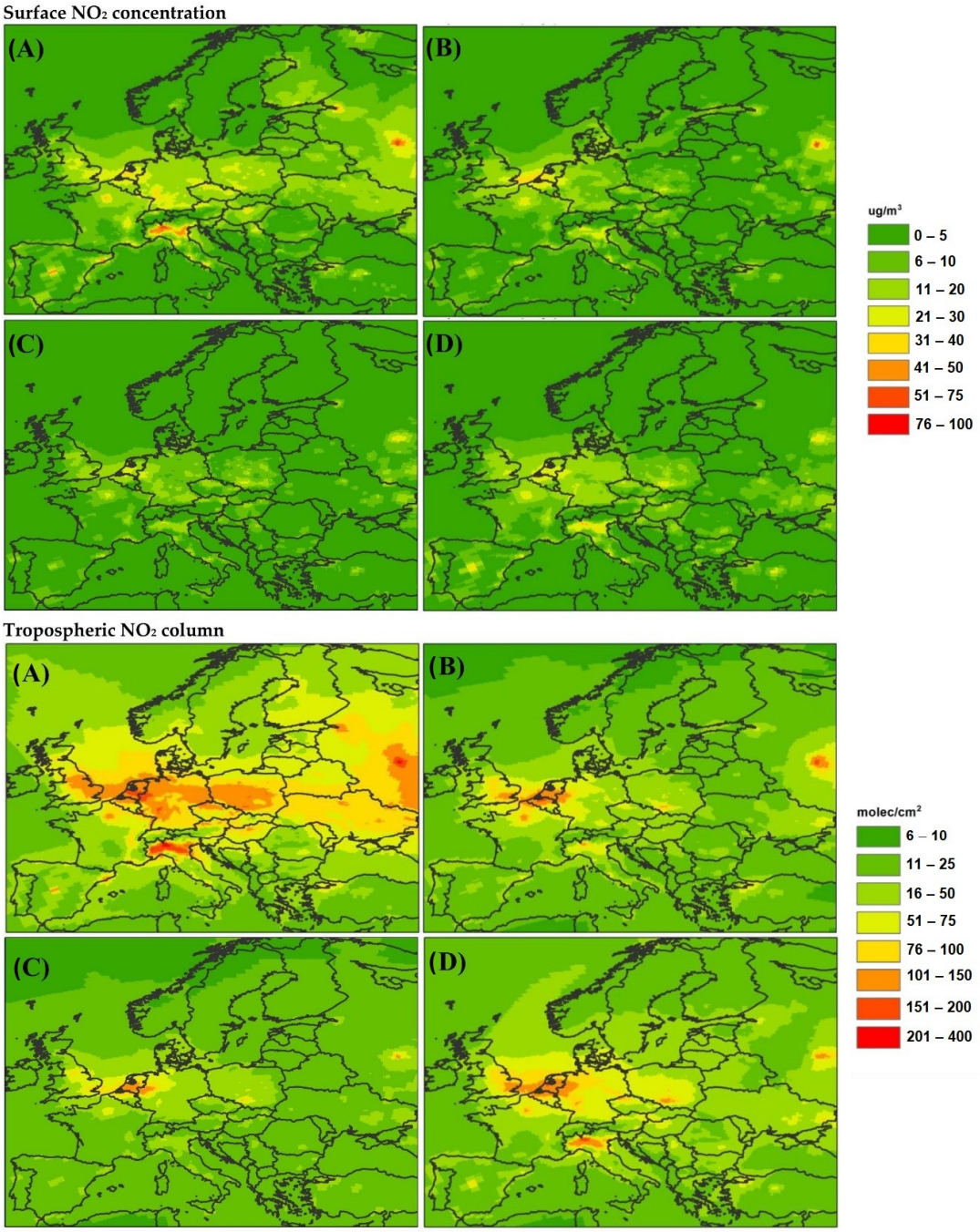

Figure 10. Surface NO_2 concentration fields (top panel) and tropospheric NO_2 column (bottom panel) for 2010 were calculated for a selected month by the GEM-AQ. (**A**) February, (**B**) April, (**C**) August, (**D**) October.

The relationship between the modelled surface NO_2 concentrations and the value of the modelled tropospheric NO_2 column is shown in Figure 11. A similar high correlation (>0.5) between OMI observations and ground-based in situ measurements was also recorded in important European Cities during 2005–2014 [40].

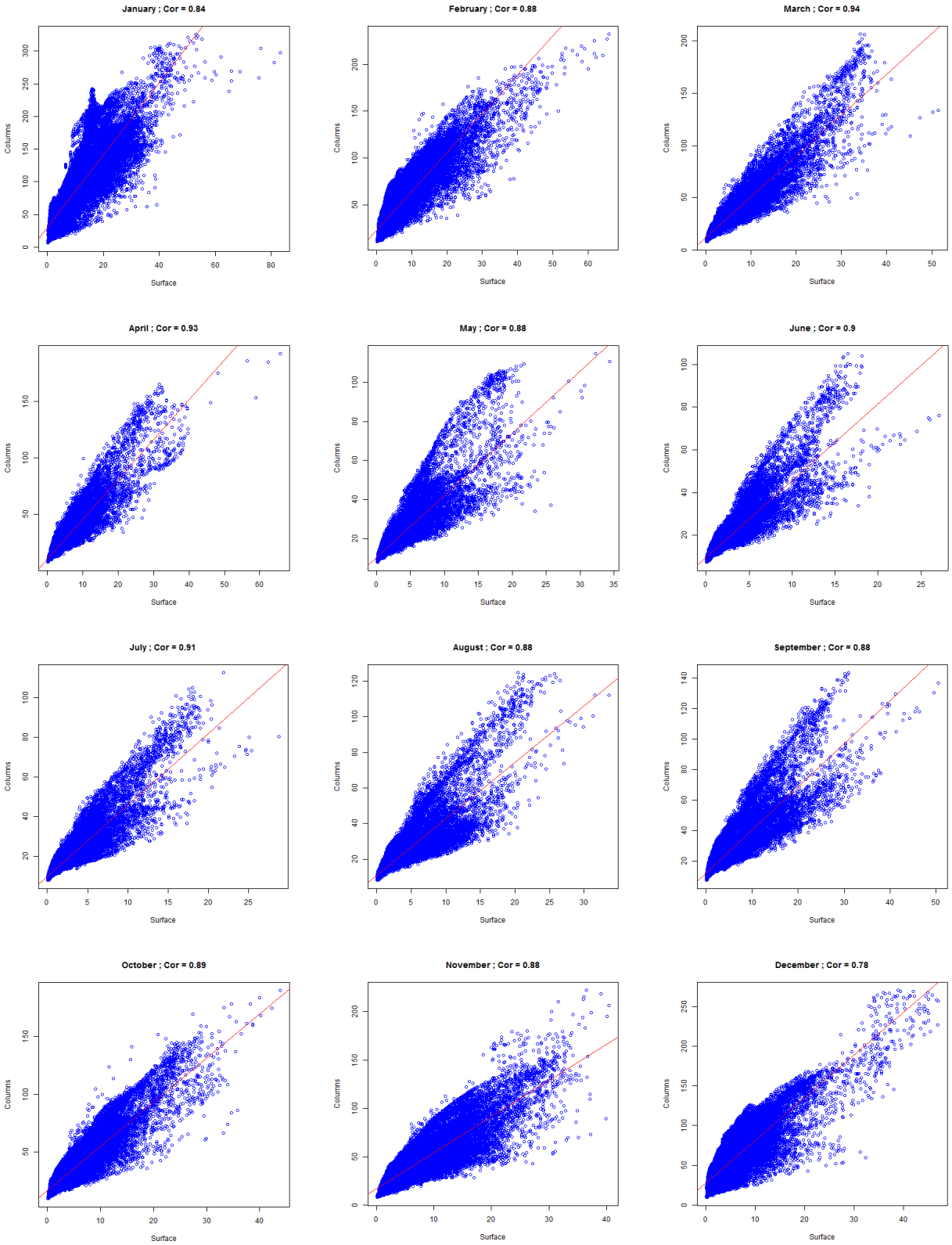

Figure 11. Relation between NO_2 tropospheric columns (Y axis) and surface concentrations NO_2 (X axis) from the GEM-AQ model (single point represent one grid point).

The observed dependency correlation coefficients between tropospheric NO_2 columns and surface NO_2 concentrations were calculated for all months in 2011 (Table 3). A high correlation in the range of 0.74 to 0.94 was calculated.

Table 3. Pearson correlation coefficients between tropospheric NO_2 column and surface concentrations for 2011.

Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
0.84	0.88	0.94	0.93	0.88	0.90	0.91	0.88	0.88	0.89	0.88	0.78

4. Discussion

High values of anthropogenic NOx emissions (over 40 $\mu g/m^3$) are mainly recorded in urbanised and highly industrialised areas. In Europe, the highest NOx emissions occur primarily in large cities, such as London, Paris, Moscow, St. Petersburg, Istanbul, and Madrid and high maritime traffic in the Mediterranean. Higher NOx emission flux values were observed on the border of three countries: Germany, the Netherlands, and Belgium. In Poland, the highest values occur in the Silesia, Łódź, and Warsaw agglomerations.

Applied correction factors lead to an overall reduction of the emission budget over Europe. Table 4 summarises the monthly total emission fluxes calculated for the whole domain. Table 5 presents the total emission fluxes in selected countries as an annual total calculated from monthly values. Generally, modified emission fluxes are lower than the base case (Table 4). Reduction varies from 18% to 41%. The smallest reduction was calculated for April (1.06×10^3 kt) and the highest was calculated for December (2.74×10^3 kt). However, based on the comparison for individual countries, for Belgium, the emission flux of NOx increased after correction (0.01×10^3 kt), and for the Netherlands, changes are relatively small (Table 5). A significant reduction of NOx emission budget followed by the improvement of the results indicates that there might be a systemic problem with the reporting of NOx emissions as the equivalent of NO_2.

Table 4. Emission of NOx (10^3 kt) for Europe before and after modification.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
Base	6.73	6.54	6.30	5.86	5.44	5.23	5.23	5.23	5.37	5.84	6.33	6.70
Mod.	4.67	3.94	4.40	4.80	3.96	3.72	3.90	3.96	4.09	3.90	3.79	3.96

Table 5. The monthly mean value of emission NOx (10^3 kt) before and after modification.

	Poland	France	Netherland	Belgium	Germany
Base	0.35	0.37	0.11	0.07	0.50
Mod.	0.22	0.31	0.10	0.08	0.40

The correlation coefficients between NO_2 columns derived from satellite observations (OMI and SCIAMACHY) and modelled by GEM-AQ for the base and modified emission scenarios were calculated to check if adjusted emissions improved the model performance. The correlation coefficient for columns from both spectrometers was enhanced by more than 0.1, and the extreme values were significantly reduced for the modified emission scenario.

The calculated correlations coefficients between the GEM-AQ base scenario and OMI-derived NO_2 tropospheric columns were higher than for SCIAMACHY for all months except for July and August 2011. The lower correlation may be because there were more data from OMI than SCIAMACHY for the algorithm training period 2008–2010. The correlation coefficient for SCIAMACHY was higher for the months when cloud cover over Europe was relatively low.

It can be seen that with the modified emissions, the average monthly NO_2 tropospheric column is better correlated with satellite data from SCIAMACHY and OMI (Table 6) for all months. For the summer months (June, July, August, and October), when a much larger number of observations were available to calculate the emission correction factors, the improvement of the correlation coefficient was ≈0.1. Significant improvement of the correlation coefficient was obtained for June and July (from 0.71 to 0.85) for SCIAMACHY.

Table 6. Correlation coefficients between NO_2 tropospheric column from satellite data and GEM-AQ base emission (BE) and modified emission (ME) scenarios for 2011 over the entire modelled domain.

All Area	Month											
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
OMI-GEM (BE)	0.64	0.68	0.86	0.83	0.80	0.74	0.69	0.73	0.77	0.78	0.78	0.60
OMI-GEM (ME)	0.68	0.74	0.92	0.89	0.88	0.85	0.78	0.82	0.88	0.87	0.83	0.65
SCIAMACHY-GEM (BE)	0.54	0.58	0.66	0.72	0.78	0.71	0.71	0.75	0.70	0.60	0.60	0.54
SCIAMACHY-GEM (ME)	0.61	0.58	0.69	0.82	0.85	0.85	0.85	0.85	0.83	0.70	0.66	0.53

The spatial distribution of the NO_2 tropospheric column calculated by the GEM-AQ model for the modified emission scenario shows a significant improvement compared to the observed patterns. The improvement of correlation over the entire area is approximately >0.1 for the summer months (Table 6). In addition, for Benelux and Silesia practically for all months, an improvement of correlation (from 0.01 to 1.14) for 2011 was achieved (Tables 7 and 8). A definite improvement of model results for the scenario with modified emissions was achieved over regions where emission fluxes were significantly reduced (North Sea, Mediterranean Sea, Silesia, Moscow, St. Petersburg). Systematic overestimations were reduced from $50–100 \cdot 10^{15}$ to $10 \cdot 10^{15}$ molec/cm^2.

Table 7. Correlation coefficients between NO_2 tropospheric column from satellite data and GEM-AQ base emission (BE) and modified emission (ME) scenarios for 2011 for the Benelux part of the modelling domain.

All Area	Month											
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
OMI-GEM (BE)	0.55	0.75	0.83	0.79	0.92	0.45	0.66	0.67	0.88	0.87	0.83	0.71
OMI-GEM (ME)	0.58	0.73	0.93	0.87	0.89	0.65	0.70	0.71	0.89	0.89	0.86	0.74
SCIAMACHY-GEM (BE)	0.65	0.68	0.60	0.63	0.89	0.69	0.77	0.83	0.71	0.66	0.76	0.42
SCIAMACHY-GEM (ME)	0.69	0.68	0.76	0.75	0.87	0.82	0.77	0.86	0.74	0.73	0.70	0.47

Table 8. Correlation coefficients between NO_2 tropospheric column from satellite data and GEM-AQ base emission (BE) and modified emission (ME) for 2011 for the Silesia part of the modelling domain.

All Area	Month											
	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
OMI-GEM (BE)	−0.50	−0.1	0.71	0.58	0.73	0.16	0.53	0.51	0.82	0.61	0.57	0.00
OMI-GEM (ME)	−0.50	−0.1	0.71	0.61	0.77	0.24	0.55	0.60	0.78	0.54	0.67	0.20
SCIAMACHY-GEM (BE)	0.23	0.25	0.52	0.43	0.59	0.33	0.41	0.56	0.80	0.25	0.53	0.52
SCIAMACHY-GEM (ME)	0.22	0.24	0.58	0.41	0.62	0.41	0.43	0.70	0.83	0.33	0.62	0.45

Although for Benelux, the correlation coefficient was generally higher than for Silesia (appropriately 0.42–0.93 and −0.5–0.83), and in most cases, correlation improved in the scenario with modified emissions, the negative bias change was relatively small. Qualitative analysis of the proposed indicator showed that the areas with a low O_3/NO_2 column ratio (below 10) coincide with regions characterised with the highest underestimation of the modelled NO_2 column compared to satellite observations.

The high spatial agreement of NO_2 tropospheric columns and surface concentrations suggests that modification of the anthropogenic emission fluxes may improve the distribution of NO_2 columns and surface concentrations as calculated by the GEM-AQ model.

For 2011, we compare concentrations of NO_2 with the model results for two scenarios (Base—with base emissions) and Bis (with modified emissions, using correction factors based on satellite data) for 14 stations (11 background rural, 2 background suburban, and 1 city station) in Poland. Statistical measures are given in Table 9. The analysis of the mean error shows that at most stations, on a yearly basis, there is a slight overestimation of the modelled NO_2 concentrations. The mean square error for all stations is approximately $\approx$10 μg/m^3.

Table 9. Statistical measures for two scenarios (Base—with base emissions) and Bis (with modified emissions, using correction factors based on satellite data) calculated for 2011.

Station	Mean		MRSE		Correlation	
	Base	Bis	Base	Bis	Base	Bis
Czerniawa	75.61	74.64	1.34	0.33	0.40	0.27
Bory Tucholskie	176.83	180.48	7.86	7.56	0.49	0.54
Widuchowa	51.09	52.35	1.09	−1.09	0.61	0.53
Krzyżówka	53.14	45.77	−1.33	−1.12	0.62	0.55
Granica KPN	101.28	149.02	8.22	5.16	0.57	0.61
Szymbark	49.12	37.64	0.90	−2.34	0.79	0.67
Smolary Bytnickie	166.23	204.09	5.63	4.84	0.70	0.79
Parzniewice	72.50	65.48	0.57	1.83	0.53	0.47
Gajew	64.53	52.92	4.77	3.69	0.77	0.69
Biały Słup	48.69	43.09	−0.19	−2.49	0.76	0.75
Belsk	58.05	66.81	3.20	0.91	0.60	0.56
Wrocław Bartnicza	61.03	74.31	4.72	1.49	0.13	0.22
Legionowo	48.32	64.32	2.33	−2.57	0.53	0.64
Białystok	54.33	50.47	0.13	−1.61	0.59	0.53

For most stations, a relatively high correlation coefficient was obtained in the range of 0.60–0.75, which shows that the long- and short-term variability for the modelled concentrations is correctly reproduced. The station with a weak correlation in relation to the daily average values is Wrocław Bartnicza, where the variability of NO_2 concentrations is controlled by the emission cycle, the phase of which was not correctly provided in the emissions inventory. Simulations with modified emissions gave slightly higher mean concentrations for all stations (change from 77.2 to 82.9 $\mu g/m^3$). The mean RMSE for all stations was almost three times lower for the Bis scenario (change from 2.8 to 1.0 $\mu g/m^3$).

5. Conclusions

A method for correcting nitrogen oxides anthropogenic emission fluxes was presented. The proposed emission correction method was developed using NO_2 column observations from two spectrometers: OMI (on board the AURA satellite) and SCIAMACHY (on board the ENVISAT satellite) and results from a chemical weather model GEM-AQ.

The presented study verified whether using emissions correction factors calculated based on systematic differences between model results and satellite observations for the reference period would improve the distribution of the modelled NO_2 columns and surface NO_2 concentrations in the independent simulation. Emission correction factors were developed from model results and satellite observations over the period 2008–2010. Verification of the prosed emission corrections factors was carried out using an independent set of NO_2 column observation and model simulations for 2011.

It was found that the modelled NO_2 tropospheric column is strongly correlated with surface NO_2 concentrations over the urban and polluted areas. The regions with the highest NO_2 column and the highest surface concentration coincide. It confirms the hypothesis that both fields are mostly dependent on anthropogenic emissions, and the developed correction factors may improve the distribution of NO_2 columns and surface concentrations. The developed method allows for modification of the emissions fluxes in areas where there were systematic differences between satellite observations and modelling results.

Application of the developed method could significantly improve model results in areas where the NO_2 column was overestimated, e.g., inactive power plants, the Mediterranean Sea, and the North Sea.

There is a need to develop emission correction factors for parts of the modelling domain where the NO_2 column was underestimated, e.g., Ukraine, Northern France, and England. Significant improvement after modification on NO_2 emissions fluxes was not achieved. The expanded factors might take into account NOx-O_3-VOC loss and production regimes.

Further development of emission correction factors and inverse modelling using satellite observations remains a very important area of research to refine estimates of anthropogenic emission fluxes. It is anticipated that future space observations and missions such as Sentinel 5p, 5, and 4 will provide additional information to support national policies in areas of air quality management and control.

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