Comparison of Satellite NO_2 Observations with High Resolution Model Simulations over the Balkan Peninsula

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Abstract. High resolution model estimations of tropospheric NO_2 column amounts from the Comprehensive Air Quality Model (CAMx) were simulated for the Balkan Peninsula and were compared with satellite data for a period of one year, in order to study the characteristics of the spatial and temporal variability of pollution in the area. The Balkan area is considered a crossroad of different pollution sources and therefore has been divided in urban, industrial and rural regions, aiming to investigate the consistency of satellite retrievals and model predictions at high spatial resolution. Satellite measurements of tropospheric NO_2 are available daily at 13:30 LT since 2004 from OMI/Aura with a resolution of 13x24km. The anthropogenic emissions used in CAMx for the domain under study, was compiled employing bottom-up approaches (road transport sector, off-road machinery) as well as other national registries and international databases. High resolution GIS maps (road network, landuses, population) were also used in order to achieve high spatial resolution. In most of the cases the model reveals similar spatial patterns with the satellite data, while over certain areas discrepancies were found and investigated.

Keywords: Balkan, satellite, model, nitrogen dioxide, air pollution.
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INTRODUCTION

Nitrogen dioxide plays an important role in photochemistry. Measurements of nitrogen dioxide (NO_2) are important for the understanding of tropospheric and stratospheric chemistry, particularly in relation to ozone production and loss [e.g. Crutzen et al., 1979, Murphy et al., 1993, Finlayson-Pitts and Pitts, 2000]. On the one hand, tropospheric NO_2 is essential for maintaining the oxidizing capacity of the atmosphere. Photolysis of NO_2 during daytime is the major source of ozone (O_3) in the troposphere and photolysis of O_3 in turn initializes the production of the hydroxyl radical (OH), the main cleansing agent of the atmosphere (van Noije et al., 2006). On the other hand, NO_2 as well as O_3 are toxic to the biosphere and may cause respiratory problems for humans. Moreover, NO_2 may react with OH to form nitric acid (HNO_3), one of the main components of acid rain.

To date no clear assessment of the behavior of nitrogen dioxide over the Balkan Peninsula exists. Few studies have localized their results over this part of Europe. For instance, in Ladstätter-Weiβenmayer et al [2007] the synergistic use of GOME tropospheric column data (version 1, developed at the University of Bremen) with back-trajectory analysis and box model calculations enabled the detection of significant changes in pollutant tropospheric columns related to general air circulation patterns. It was found that when the Mediterranean is influenced by polluted air masses from Central Europe, the Balkans and the Black Sea, pollution leads to an increase in NO_2. Furthermore, the observed mean NO_2 tropospheric column densities (in 10^{15} molecules/cm^2) were determined to be: for Crete 1.1, for Athens 2.0, for Thessaloniki 2.3 and for Istanbul 2.4 for the month of May as a mean value of the years 1996 to 2002. A detailed analysis for Western Europe was presented by Blond et al. (2007), who compared tropospheric NO_2, from a vertically extended version (up to 200 hPa) of CHIMERE with high-resolution column observations from SCIAMACHY as retrieved by BIRA/KNMI. Konovalov et al. [2005] used GOME-based data products (version 2, developed at the University of Bremen), to evaluate the CHIMERE CTM over Western (10°W,

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18°E, 35°N and 60°N) and Eastern (18°E, 65°E, 40°N and 65°N) Europe. Their study indicated much lower levels of NO_2 in Eastern Europe (which includes the Balkan Peninsula) compared to Western Europe and no clear evidence could be found that either the performance of CHIMERE or the quality of NO_2 columns derived from GOME measurements performs poorer for Eastern than for Western Europe.

The Eastern Mediterranean is a known cross road of air masses where anthropogenic pollution emissions converge with natural ones [e.g. Lelieveld et al., 2002, Mihalopoulos, 2007]. The ground-based stations that exist in the region are managed by the various local authorities, such as municipalities, prefectures, and so on. As a result, such stations exist either only in the capitals of states and in largely populated cities, or near airport. As an example, Figure 1 (left panel) demonstrates the total number of air quality monitoring stations per country used to monitor regional air pollution in Europe. According to the European Topic Center on Air Quality there is limited knowledge of surface measurements in the Balkan Peninsula. Since the presently available ground-based stations do not have a proper spatial distribution in South Eastern Europe and there are not many scientific studies that focus on NO_2 variability over the Balkan Peninsula, the present study aims at providing more details on the temporal and spatial distribution of tropospheric NO_2 columns over the area through satellite observations and model simulations and to examine the satellites' consistency over areas with moderate loading of tropospheric NO_2.

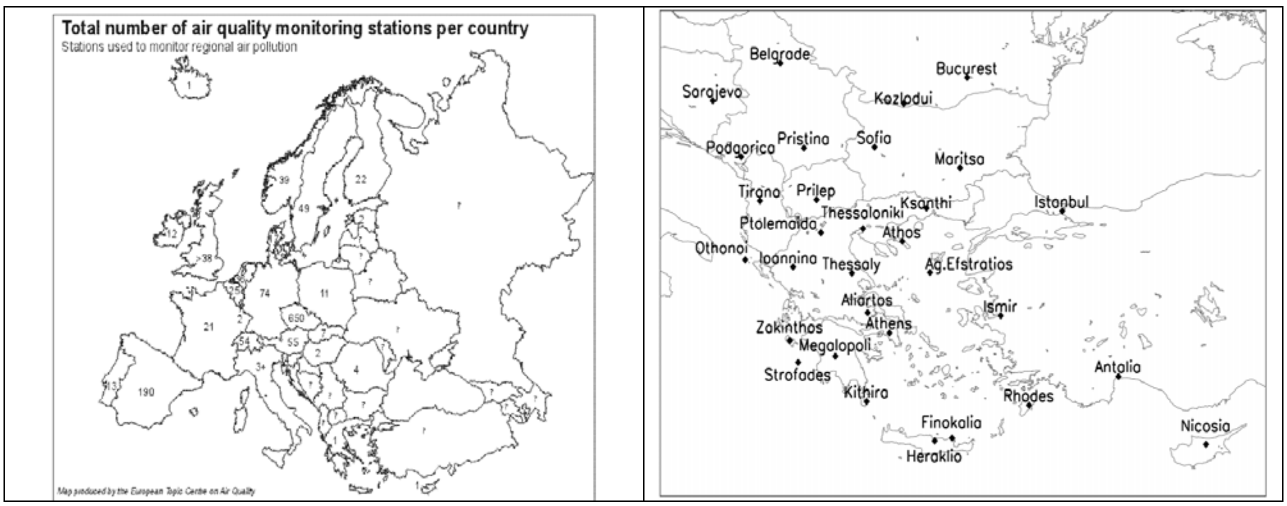


FIGURE 1. Number of air quality monitoring stations per country (left) and Spatial distribution of the geolocations considered (right).

In the next sections we give a description of the instruments, the algorithms and the photochemical model we used in our analysis. Then we investigate the long-term variability of tropospheric NO_2 over several Balkan geolocations as derived from satellite retrievals and model simulations in order to assess the ability of satellite sensors to detect pollution and to evaluate the photochemical model.

DATA SOURCES AND METHODOLOGY

Thirty-two geolocations around the Balkan Peninsula were chosen as focal point for this study and can be seen in right panel of Figure 2. They were selected according to the following criteria: their spatial distribution around the region, polluted sites such as industrial and commercial centers or capitals, unpolluted sites that can provide background values and sites that may help the detection of potential transboundary transport of NO_2 inside the Balkan Peninsula. For all these locations overpass files for the tropospheric NO_2 columns were generated from level-2 data of GOME, SCIAMACHY, OMI and GOME-2. The extraction criteria and the main characteristics of the instruments and the algorithms are discussed in the following paragraphs.

Satellite Retrievals

The main data set used in this study are NO_2 vertical tropospheric column densities retrieved by KNMI (Royal Netherlands Meteorological Institute) and BIRA/IASB (Belgian Institute for Space Astronomy) which are publicly available on a day-by-day basis via ESA's TEMIS project (http://www.temis.nl). The satellite instruments use din this study, GOME/ERS-2, SCIAMACHY/Envisat, OMI/Aura and GOME-2/MetOp, span a period of over twelve years. The main features of the four instruments, satellite platforms and data versions used in this study are summarized in Table i).

TABLE i). Sources and characteristics of satellite tropospheric NO_2 data used in this study

Instrument	Start Date	End Date	Equator Cross Time	Horizontal Resolutition Here
GOME (ERS2) [Version 1.04]	03/1996	06/2003	10:30 LT	320x40 km^2
SCIAMACHY (Envisat) [Version 1.10]	01/2003	10/2008	10:00 LT	60x30 km^2
OMI (Aura) [Version 1.02]	01/2004	10/2008	13:30 LT	13x24 km^2
GOME2 (MetOp) [Version 1.1]	04/2007	10/2008	09:30 LT	80x40 km^2

The TEMIS NO_2 vertical tropospheric column for GOME, SCIAMACHY and GOME-2 tropospheric NO_2 columns are all products of the same retrieval algorithm. The retrieval of tropospheric NO_2 is performed in three steps: first the total slant NO_2 column density is retrieved by BIRA/IASB using a Differential Optical Absorption Spectroscopy (DOAS) technique [e.g. Platt, 1994], then the stratospheric contribution is deduced by assimilating the total slant column data in the TM4 chemistry model driven by meteorological analysis from the European Center for Medium-range Weather Forecasts (ECMWF) and subsequently the vertical tropospheric column is derived, applying a tropospheric air mass factor correction. More details on the retrieval can be found in Boersma et al. (2004) and in Blond et al. (2007). The KNMI/BIRA tropospheric NO_2 retrievals have been also validated in several studies (eg. Schaub et al., 2006, Blond et al., 2007, and Lambert et al., 2007)

The OMI retrievals were developed at KNMI within the DOMINO (Dutch OMI NO_2) project. The DOMINO product is available from www.temis.nl. The DOMINO retrieval algorithm is described elaborately in Boersma et al. (2007) and recent updates can be found in the DOMINO Product Specification Document (http://www.temis.nl/docs/OMI_NO2_HE5_1.0.2.pdf). The DOMINO tropospheric NO_2 columns have been validated versus independent measurements during various campaigns (Boersma et al., 2008, 2009).

For our spatial and temporal variability analysis only observations with a radiance reflectance of less than 50% from clouds were used which corresponds to a cloud fraction of less than about 20% [van der A et al, 2008]. In addition, only completely un-flagged retrievals were accepted. For GOME, SCIAMACHY and GOME2 we used only the measurements with flag=0 and for OMI those measurements with even flag values, that correspond to meaningful tropospheric retrievals. For all the satellite instruments the distance between the satellite's center field of view and the ground locus was set to 50km, in order to obtain spatially comparable measurements. This distance was the minimum we could use in our analysis to get adequate or coincident measurements from each satellite for the intercomparisons. Finally, we used only the forward scans for GOME, SCIAMACHY, and GOME-2 and from OMI pixels with CTP (Cross Track Position) 10 to 50 that corresponds to the OMI pixels which are closest to the near-nadir viewing position.

Model Description

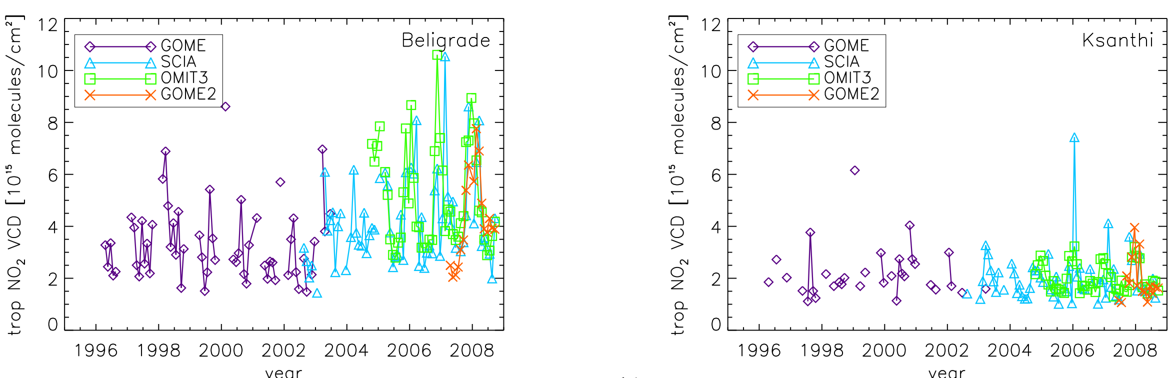
The Comprehensive Air Quality Model with extensions (CAMx) version 4.40 is a publicly available open-source computer modeling system for the integrated assessment of gaseous and particulate air pollution (www.camx.com). CAMx simulations presented in this study cover Europe with a spatial resolution of 50x50km in a domain identical to the one defined for the meteorological runs to avoid interpolation errors. The domain's vertical profile contains 12 layers of varying thickness, extending up to 450 hPa. The chemistry mechanism invoked is Carbon Bond version 4 (CB4). This mechanism includes 117 reactions – 11 of which are photolytic – and up to 67 species (37 gases, 12 radicals and up to 18 particulates). The meteorological fields were derived from REGional Climate Model runs (RegCM3, http://www.ictp.trieste.it/ ~pubregcm/RegCM3) which were forced by the ERA-40 reanalysis fields (2.5°x2.5°, L23 pressure levels) of the European Centre for Medium-Range Weather Forecasts (ECMWF). Details of the modeling system are presented elsewhere (Katragkou et al., 2009).

Organic biogenic emissions were calculated with the use of the RegCM3-CAMx interface, which extracts meteorological parameters from RegCM3 (temperature and radiation) and uses the available land use categories to calculate emission potentials and foliar biomass densities (Guenther et al., 1993). Anthropogenic emissions were calculated with data from the UNECE/EMEP data base (http://webdab.emep.int/) for European emissions (Vestreng et al., 2005) for the year 2000. Emissions from lightning and biomass burning activities were not considered in the model runs. The model run used in this study was performed in the frame of CECILIA EU project and at the moment the model is not yet optimized to include operationally these two mechanisms in the simulations. This is an ongoing effort. The boundary conditions were set to 1 ppb.

NO_2 tropospheric vertical column densities were extracted for the altitudes between 0-7 km and the time period from 01/01/1996 to 31/12/2001 for which the model run has been performed. We compared average 2-hour NO_2 predictions from CAMx with the GOME measurements, using an appropriate number of CAMx grid-cells in a way that they fit in with GOME's pixel spatial resolution.

RESULTS AND DISCUSSION

For all the locations demonstrated in Figure 2 we compiled time series of monthly mean tropospheric NO_2 columns for each satellite instrument in order to examine to what extend we can determine certain characteristics of the temporal variability of tropospheric NO_2 over the Balkan Peninsuala. We focus our discussion on some of the geolocations which, as it can bee seen in Figure 3 are representative of mega, large, industrial and rural geolocations (Beligrade, Ksanthi, Kozlodui and Strofades respectively), according to their emission characteristics. SCIAMACHY, GOME-2 and OMI can reveal the characteristics of urban scale and industrial regions due to their finer horizontal resolutions and are quite consistent, while GOME is representative of a much larger area which smoothes out the effect of these local sources. Over the polluted sites there is a relative offset between SCIAMACHY and OMI measurements as shown here, as typical examples, for Kozlodui and Beligrade, which can be possibly attributed to the different local crossing time of each satellite (around three hours difference) and to the fact that the local NO_2 diurnal variability may quite possibly be different for each location. In a rural area like Strofades, with no large sources of NO_2 in the surroundings, the mean tropospheric NO_2 amounts are close to 1.0 x10^{15} molecules/cm^2. The seasonal variability of the monthly mean tropospheric NO_2 columns is similar in all cases, with maximum values during the winter and minimum during the summer. Figure 4 presents the time series of GOME and CAMx for the corresponding geolocations to the previous Figure. There is quite good correlation between the measurements and the modeling predictions. The discrepancies, especially in the summer months, may be probably credited to the missing CAMx emissions (biomass burning), to CAMx background conditions (no long range transport is considered from sources outside the modeling domain) and also to the fact that the model resolution is 50x50km, much finer than GOME's.



a) b)

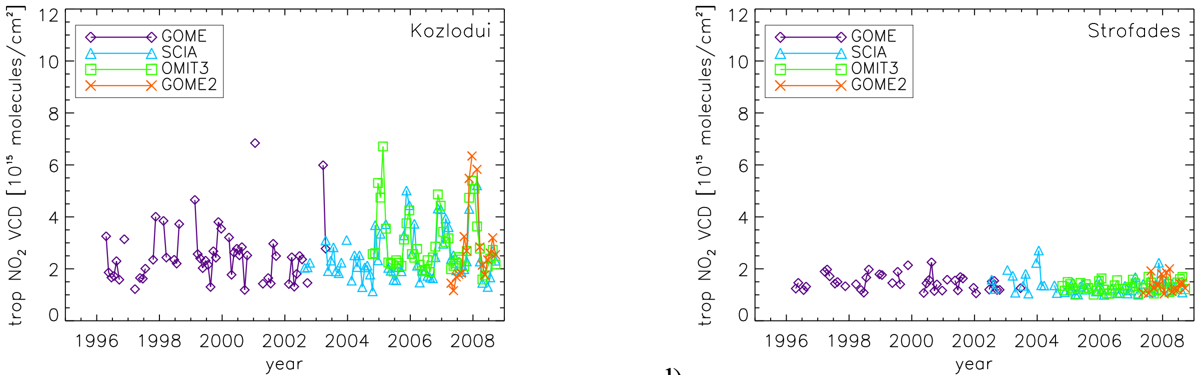


FIGURE 3. Time series analysis of satellite-measured NO_2 tropospheric columns for a) Beligrade, b) Ksanthi, c) Kozlodui and d) Strofades. The diamond, triangle, square and X symbols indicate the GOME, SCIAMACHY, OMI and GOME2 measurements respectively. The unit is 10^{15} molecules/cm^2.

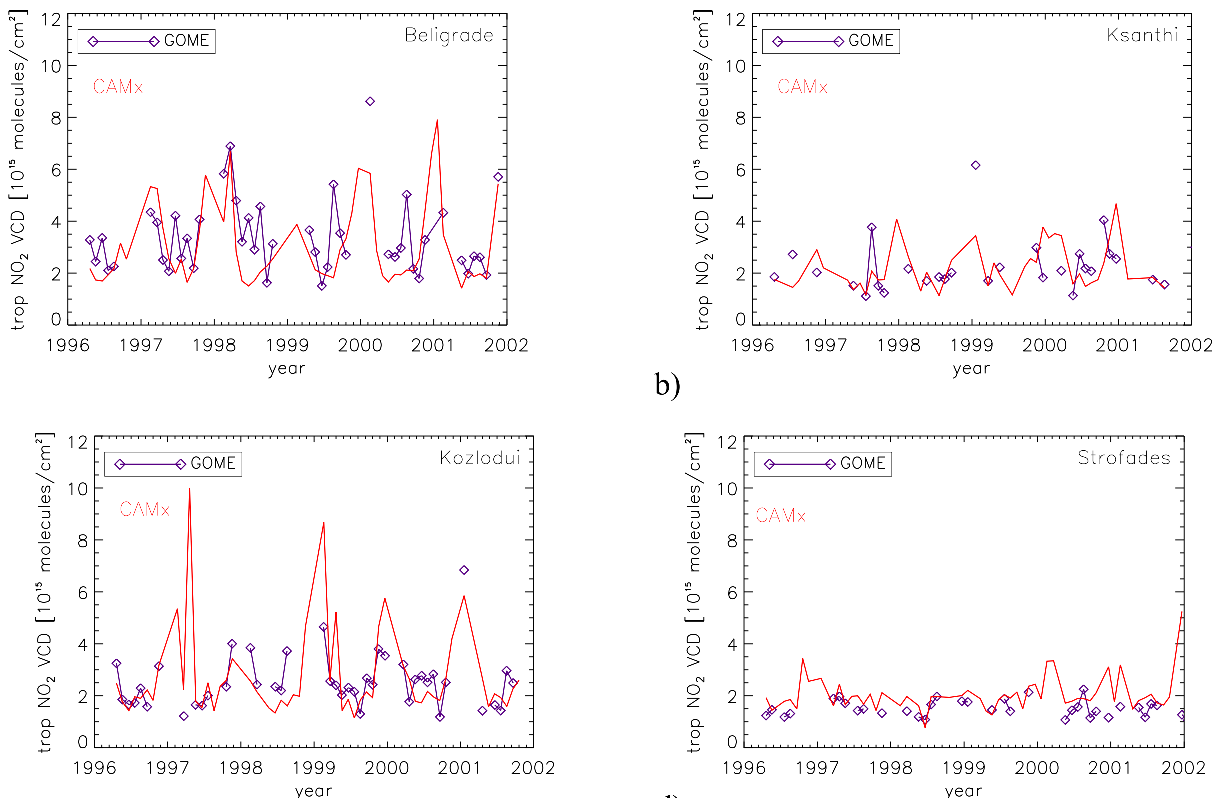


FIGURE 3. Time series analysis of satellite-measured NO_2 tropospheric columns for a) Beligrade, b) Ksanthi, c) Kozlodui and d) Strofades. The diamond symbols indicate the GOME measurements and the red solid line the CAMx predictions. The unit is 10^{15} molecules/cm^2.

CONCLUSIONS

We used 4 satellite instruments GOME/ERS-2, SCIAMACHY/Envisat, OMI/Aura and GOME-2/MetOp in order to examine the temporal variability of tropospheric NO2 vertical columns over Balkan regions deduced by

KNMI/BIRA retrieval algorithm. We have further evaluated modeling (CAMx) simulations by comparing them with GOME observations. The main findings of this work are summarized as follows:

There is a very good agreement between the time series of GOME, SSCIAMACHY, OMI and GOME2. The monthly mean NO_2 tropospheric column densities range, depending on the observing satellite, between 4.0 and 10.0 $x10^{15}$ molecules/cm^2, 2.0 and 4.0 $x10^{15}$ molecules/cm^2, 2.0 and 6.0 $x10^{15}$ molecules/cm^2 and 1.0 and 2.0 $x10^{15}$ molecules/cm^2 over mega, large, industrial and rural regions respectively. Over mega and large cities the observed levels are lower than the ones observed over the most polluted areas of Southeast Asia and Central Europe which often exceed 11.0 $x10^{15}$ molecules/cm^2. There is also quite good agreement between the observations and the modeling simulations especially during the winter months, while during summer there are discrepancies that are mainly attributed to the fact that biomass burning emissions are not included in CAMx. UV/visible satellite measurements of tropospheric species provide valuable long-term data sets which can be used to evaluate current emission inventories used by various groups, to provide estimates for average conditions over areas with limited ground-based data availability and in addition they have a great potential to detect longterm trends in regional scale pollution.

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