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Modeling results of atmospheric dispersion of NO_2 in an urban area using METI–LIS and comparison with coincident mobile DOAS measurements

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ABSTRACT

Synergetic use of in–situ measurements, remote sensing observations and model simulations can provide valuable information about atmospheric chemistry and air quality. In this work we present for the first time a qualitative comparison between modeled NO_2 concentrations at ground level using dispersion model METI–LIS and tropospheric NO_2 columns obtained by mobile DOAS technique. Experimental and modeling results are presented for a Romanian city, Braila (45.26°N, 27.95°E). In–situ observations of NO_2 and meteorological data from four ground stations belonging to the local environmental agency were used to predict the concentration of NO_2 at ground level by atmospheric dispersion modeling on two days when mobile DOAS measurements were available. The mobile DOAS observations were carried out using a UV–VIS spectrometer mounted on board a car. The tropospheric Vertical Column Density (VCD) of NO_2 is deduced from DOAS observations. The VCD was obtained using complementary ground and space observations. The correlation between model and DOAS observations is described by a correlation coefficient of 0.33. Also, model results based on averaged in–situ measurements for a period of 5 years (2008–2012) are used for an overview of the background NO_2 evolution in time and space for the selected urban area.

Keywords: *Air quality, urban pollution, nitrogen dioxide, model simulations, remote sensing*

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1. Introduction

Nitrogen dioxide (NO_2) is one of the major atmospheric pollutants, with important impact on the air quality. NO_2 is formed naturally in the atmosphere by lightning and microbial activity in soil by the oxidation of ammonium nitrate (Lee et al., 1997; Dunlea et al., 2007). NO_2 also affects the formation of ground–level ozone (O_3), and the concentration of NO_2 is determined by its emission and by its formation via chemical reaction involving NO with O_3 (Song et al., 2011; Shon et al., 2012). NO_2 can react with OH or rain droplets leading to acid precipitation. Anthropogenic emissions originate from fossil fuel combustion, power generation and transport (Ohara et al., 2007). Part of the anthropogenic NO_2 results from the NO caused by tailpipe emissions. Yao et al. (2005) point out that the direct contribution of NO_2 emissions to atmospheric NO_2 pollution is low, since the main component of NO_X emissions is NO, oxidizing to NO_2. In addition, other atmospheric contributions come from non–combustion processes: nitric acid manufacture, welding processes and the use of explosives, etc. (Hanna and Carey, 2010). NO_2 can persist in the atmosphere for several hours to one day (Beirle et al., 2003), lifetime of NO_2 being longer in wintertime due to less OH and increased volume of emissions.

Traffic activity has a significant contribution to NO_2 concentration levels in the urban area (Palmgren et al., 1996; Constantin et al., 2012; Soret et al., 2013; Lee et al., 2014) and is considered to be responsible for over half of NO_X emissions and represents a higher proportion in urban areas (Donnelly et al., 2011). This phenomenon occurs as a result of the increase of primary NO_2 emissions from diesel–fuelled passenger cars (Carslaw et al., 2011; Constantin et al., 2012; Ramachandran et al., 2013) and the non–linear, photochemical reaction of traffic–emitted NO to NO_2 (Sjodin et al., 1996; Meng et al., 2008). NO_2 concentrations near urban traffic routes are often higher than the maximum admitted levels due to the continuous input from road vehicles (Carslaw and Beevers, 2004; Anttila et al., 2011). For instance, NO_2 concentrations in Europe exceeded the annual maximum value in many traffic urban areas in 2009 (EEA, 2009). Since 2010, a new EU limit of the average annual ambient air NO_2 levels has been set up to 40 $\mu g/m^3$ or alternatively the maximum value of 200 $\mu g/m^3$ should not exceed more than 18 hours per year (EC, 2008).

This study uses time series of hourly NO_2 concentrations for the time interval 2008–2012 obtained for the city of Braila from the Local Environmental Protection Agency (EPA). Braila city is considered to be moderately polluted (Constantin et al., 2013a). According to data provided by Braila EPA, more than 50% of NO_2 emissions originate from transportation. While the NO_2 industrial emissions decreased during the last years, due to economic recession, the total NO_2 emission were almost constant. The increasing number of vehicles in Braila city, from 58 335 units in 2008 at 70 700 units in 2012 (DDLVR, 2014), has an important role

in the total amount of NO_2. In this paper the spatial and temporal distribution of NO_2 is analyzed using ground–level NO_2 concentrations obtained from measurements, a Gaussian dispersion model, METI–LIS (METI, 2006) and remote sensing observations.

Dispersion models estimate the circulation of pollutants in air and are widely used to calculate the spatial distribution of a pollutant concentration. Dispersion models have been categorized as statistical, deterministic, mathematical and physical modeling (Khare and Sharma, 2002; Odman and Hu, 2010; Carbonell et al., 2013). For the NO_2 model simulations we use the Gaussian dispersion model METI–LIS (Ministry of Economy, Trade and Industry Low Rise Industrial Source). A detailed description of the model and program initialization can be found in the next section.

The aim of the article is two–fold: to evaluate the temporal and spatial evolution of NO_2 concentration using dispersion modeling, by METI–LIS model results for a period of 5 years and to qualitatively compare model results with mobile DOAS measurements. On the one hand, in–situ measurements are performed on a continuous basis, which gives the opportunity to obtain also an overview of the diurnal and seasonal variation of the background NO_2, by averaging over the whole 5–years period. On the other hand, DOAS mobile measurements can be performed only for limited periods of time, thus NO_2 values for only those particular days are needed for the qualitative comparisons.

Experimental data and the methodology are described in Section 2, while the results and discussions are presented in Section 3. Conclusions are given in the last section.

2. Data and Methods

2.1. Observation technique: *in–situ*

Since 2008 the Romanian Ministry of Environment implemented a network of air quality monitoring stations. The National Air Quality Monitoring Network currently consists of 142 fixed monitoring stations, 41 laboratories for analysis endowed with necessary equipment and 42 centers for data processing (Directive 2008/50/EC; EC, 2008). Each monitoring traffic station records, on an hourly basis, meteorological data and concentration of atmospheric pollutants: sulfur dioxide (SO_2), nitrogen oxides (NO_X), carbon monoxide (CO), ozone (O_3), volatile organic compounds (VOCs) and particulate matter. The air quality monitoring network in Braila city consists of four monitoring stations (BR1, BR2, BR3, and BR4) which are localized in different zones (Figure 2) to measure atmospheric emissions and meteorological parameters. For the NO_2 measurements "Thermo scientific model 42i NO–NO_2–NO_X" analyzers, which are based on the most common used technique (chemiluminescence), recommended by the European legislation (European Standard, 2005) are used. This technique involves the reduction of NO_2 to NO using heated (300–350 °C) Molybdenum (Mo) surfaces. Briefly, it is based on the chemiluminescent reaction of NO with O_3 to form electronically excited NO_2, which fluoresces at visible and near infrared wavelengths (Dunlea et al., 2007).

2.2. Observation technique: remote sensing

The remote sensing measurements of the tropospheric NO_2 used in this paper are based on the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt, 1994; Platt and Stutz, 2008; Adame et al., 2012). DOAS is a technique which is widely used on board different mobile platforms like satellites (Bovensmann et al., 1999), aircrafts (Merlaud et al., 2012) or cars (Rivera et al., 2009). The experiment was performed using a mobile DOAS system installed in a car, operated in the framework of a collaboration with BIRA–IASB. The optic system consists mainly of a UV–VIS spectrometer which acquires spectra between 200–750 nm. More details about the mobile DOAS system can be found in Constantin et al. (2013b). The spectra registered during the experiments were analyzed with QDoas software (Fayt et al., 2011), which is dedicated to the spectra analysis measured by the respective instruments. The DOAS analysis gives the Differential Slant Column Density (DSCD) and the tropospheric Vertical Column Density (VCD) of NO_2 is the result of an algorithm which involves ground and space observations (Constantin et al., 2013b). The NO_2 VCD was obtained from the spectra recorded in the zenith geometry, using the procedure introduced by Constantin et al. (2013b). This retrieval procedure of NO_2 VCD is based on the DSCD resulted from the DOAS analysis and involves a Slant Column Density (SCD) reference calculated with the Langley–plot method, a stratospheric VCD obtained from satellite observations and an Air Mass Factor (AMF) calculated with the radiative transfer model (RTM) UVspec/DISORT (Mayer and Kylling, 2005). In brief, the tropospheric VCD can be expressed as:

$$VCD_{tropo} = \frac{DSCD_{meas} + SCD_{ref} - SCD_{strato}}{AMF_{tropo}} \quad (1)$$

2.3. Model Simulation: METI–LIS Model

The NO_2 concentration was simulated using the METI–LIS software (Kouchi et al., 2004), which is a computer–based model developed originally by the Japan Ministry of Economy, Trade and Industry (METI, 2006). The program METI–LIS, model ver. 2.03, is a Gaussian dispersion model and calculates concentrations in steps of one hour or less, therefore a minimum of meteorological data per each hour is necessary (Al Razi and Hiroshi, 2012). This model includes point source, line source, building downwash, terrain effects, and line source emissions. The METI–LIS model adopted a downwash scheme based on that of the US Environmental Protection Agency's (EPA) Industrial Source Complex (ISC) model, but the parameters in the dispersion widths describing the down–wash effect were improved by incorporating the results of wind tunnel experiments (Bowers and Anderson, 1981).

Essential input data are emission rate and other emission conditions such as location, height, gas volume and temperature, and meteorological factors at every hour during the averaging period. The measured hourly NO_2 data–set was processed to match the pattern required by the program and were used as inputs in the model. Wind direction and speed, temperature, solar radiation and atmospheric stability were the meteorological data required for our analysis. Considering that, the recommended format for wind–direction data are compass–point notation (north–north–east: 1; north: 16; calm: 0; missing measurements: 9 999). Wind measurements, clockwise from due north, were converted to the above format before input. Wind–speed data was expressed to 0.1 m/s accuracy. The daytime Pasquill stability category was determined by entering solar radiation data measured with a pyrheliometer and hourly averaged for the time interval. The night stability has been modeled on the assumption that it is solely dependent on wind speed, thus the model uses only wind–direction and wind–speed data for night stability calculations; cloud cover or other input data are not needed.

The purpose of METI–LIS model is to estimate a long–term, average distribution of pollutant concentrations in a relatively large area, such as Braila. Atmospheric concentration dispersion of chemical substances of a 50×50 m square spatial grid for a short or long term can be calculated by this model. Because there is very limited knowledge about the horizontal dispersion widths observed near roadways, this model sets the horizontal dispersion width to the target road width divided by 2.15, as given in the line–source calculation released by the US Environmental Protection Agency (Bowers et al., 1982).