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Assimilation of OMI NO_2 retrievals into a regional chemistry-transport model for improving air quality forecasts over Europe

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ABSTRACT

This paper presents the assimilation of satellite NO_2 observations into a chemistry-transport model (CTM). The NO_2 columns from Ozone Monitoring Instrument (OMI) aboard NASA Aura satellite are used during November–December 2005. These satellite observations are assimilated in an air quality model from Polyphemus, in order to better forecast NO_2 in Europe. The optimal-interpolation method is applied to produce analyzed columns, and these analyzed columns are mapped to model concentrations assuming that the model vertical profile is perfect. Good consistency is seen in the comparisons of model simulations, satellite data and ground observations before assimilation. The model results with and without assimilation are then compared with ground observations for evaluating the assimilation effects. It is found that the assimilation can improve the NO_2 forecasts by reducing their discrepancies against ground observations, indicating a better NO_2 forecast obtained with OMI observations. Such improvements are seen in a cold season rather than in a warm one probably due to the longer lifetime of NO_x and the initial condition changes having more impacts in winter. This also suggests that the assimilation of the short-lived species like NO_2 is a complicate problem.

1. Introduction

Nitrogen dioxide (NO_2) plays an important role in tropospheric chemistry (Logan, 1983). Anthropogenic NO_x emissions are key precursors for tropospheric ozone (O_3) (Murphy et al., 1993) and can affect the formation of particulate ammonium nitrate (NH_4NO_3) in the presence of ammonia (NH_3) (Logan, 1983). NO_2 also has a direct impact on the public health. NO_2 exposure may affect lung function and increase the risk of respiratory symptoms (Panella et al., 2000; Smith et al., 2000). Strong associations exist between NO_2 and nonaccidental mortality in daily time series studies (Steib et al., 2003; Burnett et al., 2004). Therefore, a better knowledge of NO_2 concentration is important to many issues related to air quality. In such context this study carries out assimilations of new observations with the aim of better forecasting NO_2 in a CTM.

Data assimilation is effective in the optimal estimation and forecast of atmosphere (Kalnay, 2003). By using observations of parts of the state it can estimate the less known model parameters or the state itself. Data assimilation techniques have been applied in the numerical weather prediction, stratosphere and oceans for a long time (Daley, 1993; Talagrand, 1997, 2003; Bouttier and Courtier, 1999; Ménard et al., 2000). Recently this method was introduced to air quality study (Elbern et al., 2000; Elbern and Schmidt, 2001, 2002; Segers et al., 2005). Pollutant measurements of diverse nature (ground, airborne, *etc.*) have been assimilated for different species (Elbern et al., 2007; Matsui et al., 2004; Richards et al., 2006). For nitrogen dioxide satellite data were used to improve the estimation of NO_x emissions (Konovalov et al., 2006; Napelenok et al., 2008). Few studies have been made to directly adjust the NO_2 concentrations with data assimilation techniques, due to the difficult modeling of NO_2.

Satellite observations of tropospheric NO_2 columns began in 1995 with the Global Ozone Monitoring Experiment (GOME) spectrometer aboard the European Research Satellite (ERS-2) (Burrows et al., 1999), and was followed by the Scanning Imaging Absorption spectroMeter for Atmospheric ChartograpHY (SCIAMACHY) (Bovensmann et al., 1999). Studies on GOME and SCIAMACHY observations have demonstrated the capacity of observing air pollution from space (Leue et al., 2001; Richter et al., 2005). More recently, the Ozone Monitoring Instrument (OMI) aboard the Earth Observing System (EOS) Aura satellite was launched in 2004 (Levelt et al., 2006). OMI provides high quality measurements of NO_2 columns with better spatial resolution

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and in daily global coverage, which are important advantages compared to earlier instruments of GOME or SCIAMACHY. The OMI NO_2 data have been validated against surface, in-situ, aircraft and other satellite observations (Boersma et al., 2008a,b; Brinksma et al., 2008; Kramer et al., 2008; Lamasal et al., 2008) and show good consistency with those observations. The OMI data are then used in this study.

Polyphemus is a modeling system for forecast and assessment of air quality (Mallet et al., 2007). It can deal with applications on photochemistry and aerosol dynamics at continental scales using Eulerian models. Polyphemus has been involved in operational testing for photochemistry forecasts on the French Prév'air platform from summer 2004 (http://www.prevair.org/). In this study the system is used with a similar configuration to forecast NO_2 concentrations in Europe. A cold season (November–December 2005) is studied for its importance regarding public exposure–more NO_2 concentrations are seen than in a warm period (Uno et al., 2006).

In Section 2 we introduce the Polyphemus system, its configuration in the present case and the comparisons between model simulations and ground observations. The NO_2 column observations from OMI are evaluated and their comparisons with model results are then detailed in Section 3. In Section 4 the assimilation method is introduced, with the choice of its parameter values explained, and the assimilation results are then presented. Discussions of a summer case and the conclusions can be found in Section 5.

2. Polyphemus system and comparisons with ground observations

2.1. Polyphemus system and experiment configuration

Polyphemus system can deal with applications of passive tracers, radioactive decay, photochemistry and aerosol dynamics from local to continental scales (Quélo et al., 2007; Sartelet et al., 2007; Korsakissok and Mallet, submitted for publication). It consists of four distinct components. i) Preprocessing of input fields (meteorological fields, emissions, …) in the first stage, which relies on the physical parameterizations available in the AtmoData library. ii) Eulerian chemistry-transport models (Polair3D and Castor) and local-scale plume and puff Gaussian models. iii) Drivers for handling the models, *e.g.*, a driver for Monte Carlo simulations, drivers for data assimilation. iv) Postprocessing tools (*i.e.*, the Python module AtmoPy) for statistical analysis of the results. Detailed descriptions of the system and source code are available at http://cerea.enpc.fr/polyphemus/. The version 1.3.1 of Polyphemus is used for study.

The configuration of the model can be described as follows. The land use data are obtained with 1 km resolution from GLCF (Global Land Cover Facility) data base (http://glcf.umiacs.umd.edu). Meteorological input data are computed from ECMWF (European Centre for Medium-Range Weather Forecasts) fields (12-h forecast cycles starting from analyses) with 60 vertical levels, a horizontal resolution of 0.36° by 0.36° and a time step of 3 h. The initial conditions and daily boundary conditions are extracted from outputs of the global Chemistry-Transport Model Mozart 2 (Horowitz et al., 2003). Anthropogenic emissions (*e.g.*, NO_X, SO_X, *etc.*) are provided by EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) inventory, aggregated according to Middleton et al. (1990). Biogenic emissions are parameterized with the method proposed in Simpson et al. (1999). Deposition velocities are from Zhang et al. (2003) for gaseous species.

The Chemistry-Transport Model (CTM) Polair3D (Boutahar et al., 2004) is in charge of the time integration of the reactive transport equation. The model domain covers the west and central Europe, from 10.5°W to 23°E and from 25°N to 58°N, with a resolution of 0.5° by 0.5° in horizontal directions. The simulation domain is shown in Fig. 1. In this study the maximum vertical height is 5000 m with 10 levels from the ground. The simulation time step is 10 min, with a time range from 1 November to 31 December 2005.

2.2. Comparisons with ground observations

NO_2 is more concentrated near the ground and the concentrations are much less at higher altitudes of the troposphere (Schaub et al., 2006). The ground concentrations are the key data for understanding the model performance in both the ground and its columns. In the following the model ground concentrations are compared with ground observations for validation.

The ground observations are from the EMEP network (http://tarantula.nilu.no/projects/ccc/emepdata.html). It can provide the hourly measurements of NO_2 near surface in Europe. The important advantages of EMEP measurements are the common quality control standards applied and their station locations; the latter makes the measurements representative of regional background conditions, relatively unaffected by local emissions. Several other monitoring networks (urban, suburban, *etc.*) are not used here, since they would pose serious problems concerning criteria of data selection and treatment of uncertainties. An obvious disadvantage of the EMEP network is the rather sparse distribution of stations for NO_2 (Sartelet et al., 2007).

The data at the same hours and locations are collected from model and EMEP stations respectively, and their mean ($\mu g\,m^{-3}$), correlation and root mean square error (RMSE) ($\mu g\,m^{-3}$) are computed on a daily basis using all the data. Totally 13 stations are found with data in the studied domain and the time period, with 673 pieces on the daily basis. RMSE evaluates the differences (*i.e.*, errors) between model and ground observations, and the standard deviation of these differences, referred to as STDDIF, is also computed in the following. The equations for computing these statistics are described in detail in the Appendix section Evaluation statistics. The RMSE is found to be 5.8 $\mu g\,m^{-3}$ between model and ground observations, and the STDDIF is 5.1. Their correlation is 0.7, indicating a good consistency between them. Model results are higher than ground observations on average: 12.5 $\mu g\,m^{-3}$ versus 8.1 $\mu g\,m^{-3}$. Konovalov et al. (2006) reported the effects of emission uncertainties (random part) on the modeling of NO_2 concentrations

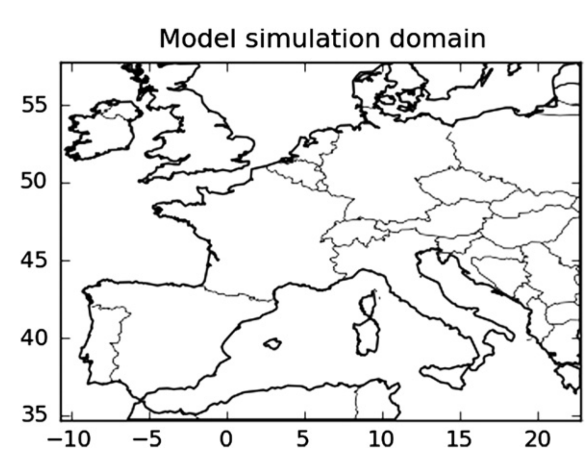

Fig. 1. The simulation domain in the study.