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Improving ozone forecasts over Europe by synergistic use of the LOTOS-EUROS chemical transport model and in-situ measurements

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HIGHLIGHTS

► Traditional modeling system misses high ozone peak.
► We assess the benefit of the synergistic use of model and measurements.
► The added-value of the data assimilation (DA) of ozone in-situ data is investigated.
► Study of the scaling factors of DA shows that no information can be carried into the forecast.
► In practice, the ozone maxima were better reproduced for both reanalysis and forecasting.

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ABSTRACT

In this paper we investigate the added value of assimilation of ozone in-situ measurements for both re-analysis and forecasting purposes. Various simulations were performed using the LOTOS-EUROS chemical transport model and an Ensemble Kalman Filter (EnKF) to assimilate measured ozone surface concentrations over Europe, for spring and summer of 2007. The results for the re-analysis of ozone show a significant improvement in the LOTOS-EUROS performance score when compared to measurements. The average correlation coefficient for the daily maximum ozone concentration improves from 0.72 to 0.83. Similarly, the average Root Mean Square Error (RMSE) for the daily maximum ozone concentration is reduced from 20.8 to 16.9 $\mu g\ m^{-3}$.

The free running model performs well in forecast mode and the agreement between in-situ and modeled ozone concentration is good. The average temporal correlation coefficient ranges from 0.62 for the first day forecast to 0.61 for the third day forecast. Based on these results, assimilated fields were used to initialize the forecast. As for the re-analysis a better comparison between model and observation was observed. The mean correlation coefficient increased by 0.07 and the averaged RMSE decreased by 0.65 $\mu g\ m^{-3}$. However, the addition of an inheritance scheme to import additional information from the data assimilation to the forecast did not significantly improve the concentration fields.

1. Introduction

The chemical composition of the atmosphere is significantly affected by the emissions of trace gases and aerosol particles due to the increased human activities since the industrial revolution. These changes play a key role for air quality on a local to global scale and for climate change. Nowadays, many uncertainties related to the emission, formation, deposition and transport still remain. This paper deals with tropospheric ozone, which is an important species for tropospheric chemistry. Tropospheric ozone results from the oxidation of atmospheric nitrogen oxides and volatile organic compounds. The role of ozone in the troposphere is twofold, as it has both a global impact on climate change as a greenhouse gas and an impact on air quality in the boundary layer. Ozone smog events have adverse effects on vegetation (Felzer et al., 2007), materials and human health (West et al., 2007). In 2004, the World Health Organization concluded that exposure to high ozone levels is clearly linked with increased premature mortality. Therefore, the need to analyze and forecast the air quality in Europe has become an obligation under the EU framework Directive on air quality.

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In recent years, modeling systems on national and international scale have been set up to analyze and forecast air pollution levels (Kukkonen et al., 2011). The ability of the regional air quality model LOTOS-EUROS (Schaap et al., 2008) to reanalyze and forecast tropospheric pollution levels has been demonstrated in various studies. The model has been used for the assessment of particulate air pollution such as PM10 (Denby et al., 2008), secondary inorganic components (Barbu et al., 2009; Schaap et al., 2004), or primary carbonaceous components (Schaap et al., 2008). LOTOS-EUROS has taken part in international model comparisons addressing ozone (van Loon et al., 2007). Evaluation of the modeling system has pointed out that the variability of the ozone concentrations is well reproduced. However, the model skill and uncertainties are not homogeneous over Europe. Northern and Central Europe are better represented than Southern Europe and maximum values due to high ozone episodes are less well captured.

The synergistic use of in-situ measurements with the regional air quality model might hold the answer to this problem. Data assimilation allows to combine model results with observational data to provide an optimal estimate of the spatial and temporal distribution of pollutants. Various studies have presented the added value of data assimilation for re-analysis and forecasting regional air quality (Elbern et al., 2007; Wu et al., 2008; Carmichael et al., 2008). Nowadays, the performances of such a system for re-analysis purposes have been extensively demonstrated while the added-value for forecasts skill remains uncertain. In this study, we investigate the added-value of the assimilation for the re-analysis and forecasting of ozone surface concentrations over Europe of ozone surface observations using LOTOS-EUROS.

2. Methodology

2.1. Description of the in-situ measurement data for assimilation and validation

The ozone in-situ measurements used for assimilation and validation in this study were gathered from the public air quality database of the European Environment Agency (EEA), AIRBASE (2007). The measurement sites are categorized according to their location (rural, suburban and urban) and type (background, industrial and traffic). The spatial resolution of the LOTOS-EUROS model is coarse compared to city/street scales and cannot resolve enhanced concentrations near localized sources. Therefore, only stations flagged as rural were considered.

For 2007, 424 stations were available, for validation purposes the available measurements were split in two sets: two thirds of the stations were used in the assimilation system while the remaining third was used for validation. In keeping with the MACC requirements, randomness and homogenous spatial coverage with respect to the station typology were the principles used for the creation of the two datasets. Fig. 1 provides an overview of the spatial distribution of the ground-based measurement locations throughout Europe. It is noteworthy to point out that the station density is greater over Central Europe and the Benelux whereas very few data were available over Southern and Eastern Europe or Scandinavia.

The classification of the AIRBASE stations is currently quite subjective and ambiguous. Therefore, a more refined classification which was recently developed was also used to better interpret the results. Henne et al. (2010) studied the station representativeness of 1287 rural or remote background monitoring sites in Western and Central Europe. The distribution and strength of surface sources, but also transport, chemical transformations and surface deposition influence the variability of trace gases concentrations within a given area. To assess station representativeness, the authors looked at the strength of these processes and at surface

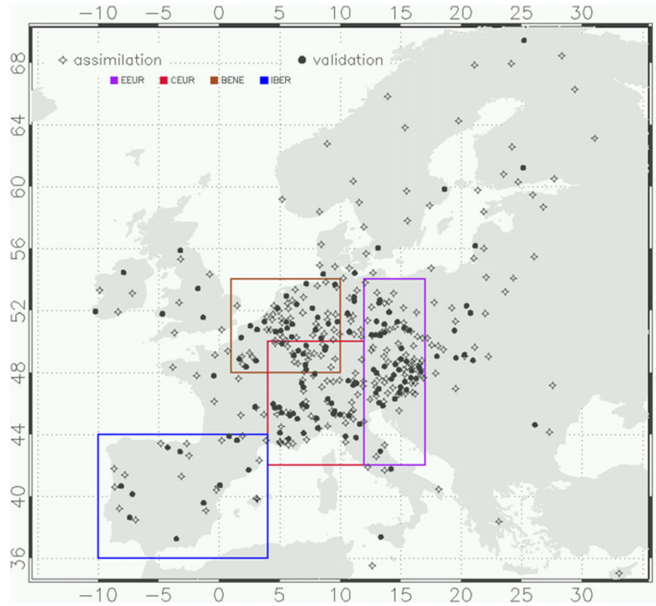

Fig. 1. AIRBASE ozone station providing data for 2007. The red, blue, brown and purple rectangles indicate the 4 sub-regions over Europe which are used to discuss the impact of the data assimilation on the forecast. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

observations of the short lived trace gases i.e. NO_2, O_3 and CO (respectively, nitrogen dioxide, ozone and carbon monoxide) within 10 and 50 km radius around each site. The sites have been categorized according to the following 10 parameters: sum of population within 10 and 50 km radius, mean deposition burdens within 10 and 50 km radius, standard deviation of population density within 10 and 50 km radius, standard deviation of deposition burdens within 10 and 50 km radius, and altitude difference between the site and the mean surface altitude in the selected area. Population data is used as a proxy for emissions and land cover data as a proxy for deposition. Advection towards the sites was also taken into account. This was simulated via backward Lagrangian particle dispersion modeling, which traced the particles backwards in time to determine potential contributions for non-local sources. As a result, eight station classification categories were distinguished: suburban, rural/populated, rural, rural/coastal, valley basin, rural/agricultural, elevated, and remote.

2.2. Modeling set-up

LOTOS-EUROS (Schaap et al., 2008) is a 3D chemistry transport model which simulates the air pollution in the lower troposphere over Europe. Currently, the model is used for operational air quality forecasts over Europe, as contribution to the Monitoring Atmospheric Composition and Climate project (MACC, http://www.gmes-atmosphere.eu) and over the Netherlands, published on the website of the Rijksinstituut voor Volksgezondheid en Milieu (RIVM, http://www.lml.rivm.nl). In this section, we describe the LOTOS-EUROS model (version 1.6) in the applied configuration.

In this study, the master domain which spans from 35° to 70° North and −10° to 60° East is translated by 5° to the west following the domain definition adopted by the MACC regional models. The grid resolution is 0.50° longitude *x* 0.25° latitude. The chemistry is parametrized following the TNO CBM-IV scheme (Schaap et al., 2008) and the aerosol chemistry is accounted for using the ISORROPIA parametrization (Nenes et al., 1998). The transport is represented by advection in three dimensions, vertical diffusion and