



## Evaluation of Meso-NH and WRF/CHEM simulated gas and aerosol chemistry over Europe based on hourly observations



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### ABSTRACT

Gas chemistry and aerosol chemistry of 10 km-resolution mesoscale models Meso-NH and WRF/CHEM were evaluated on three cases over Europe. These one-day duration cases were selected from Freney et al. (2011) and occurred on contrasted meteorological conditions and at different seasons: a cyclonic circulation with a well-marked frontal zone on winter, an anti-cyclonic situation with local storm precipitations on summer and a cold front in the northwest of Europe associated to a convergence of air masses over eastern Europe and conflicting air masses over Spain and France on autumn. To assess the performance of the two models, surface hourly databases from observation stations over Europe were used, together with airborne measurements. For both models, the meteorological fields were in good agreement with the measurements for the three days. Winds presented the largest normalised mean bias integrated over all European stations for both models. Daily gas chemistry was reproduced with normalised mean biases between  $-14$  and  $11\%$ , a level of accuracy that is acceptable for policy support. The two models' performances were degraded during night-time quite likely due to the constant primary species emissions. The  $PM_{2.5}$  bulk mass concentration was overestimated by Meso-NH over Europe and slightly underestimated by WRF/CHEM. The absence of wet deposition in the models partly explains the local discrepancies with the observations. More locally, the systematic low mixing ratio of volatile organic compounds in the gas phase simulated by WRF/CHEM at three stations was correlated with the underestimation of OM (organic matter) mass in the aerosol phase. Moreover, this mass of OM was mainly composed of anthropogenic POAs (primary organic aerosols) in WRF/CHEM, suggesting a missing source for SOAs (secondary organic aerosols) mass in WRF/CHEM aerosol parameterisation. The contribution of OM was well simulated by Meso-NH, with a higher contribution for the summer case. For Meso-NH, SOA made the major contribution to the OM mass. The simulation of the mass of  $SO_4^{2-}$  in particles by both models was often overestimated and correlated with an underestimation of the  $SO_2$  mixing ratio. The simulated masses of  $NO_3^-$  and  $NH_4^+$  in particles were always higher for Meso-NH than for WRF/CHEM, which was linked to a difference in  $NO_x$  mixing ratio between the models. Finally, computations of model performance criterion and model performance goals show that both models can be considered acceptable for standard modelling applications. In particular, Meso-NH model, using a gaseous chemical mechanism designed to compute the organic precursors of aerosols, shows comparable simulated amounts of SOA to observations at local sites.

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### 1. Introduction

Air pollutants have an effect on human health (Pope et al., 2004; Rueckerl et al., 2011), ecosystems and regional climate (Monks et al., 2009). Greenhouse gases impact the climate primarily through shortwave and longwave radiations (Myhre et al., 2013), while aerosols,

in addition, affect the climate through cloud–aerosol interactions (O'Donnell et al., 2011; Rap et al., 2013; Boucher et al., 2013).

Aerosols are composed of solid and liquid particles of varying chemical complexities, sizes, and phases. New particles are added into the atmosphere by direct emissions and nucleation (secondary particles). Primary particles originate from anthropogenic sources such as fossil fuel combustion and natural sources (fire, desert dust, sea salt, etc). Secondary particles are formed through nucleation and condensation of the gas phase or by in-cloud processes (Ervens et al., 2011). Organic aerosols are a key issue for models as their formation processes and evolution are poorly known. Depending on meteorological

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conditions and aerosol properties, aerosol particles act as cloud condensation nuclei with the potential to impact the precipitation pattern (Poschl, 2005; Dusek et al., 2006; Rosenfeld et al., 2008). The coupling between aerosols and cloud remains an important but poorly understood issue. Unlike well-mixed greenhouse gases, short-lived aerosols exhibit a strong regionality in climate forcing and air quality impacts (Monks et al., 2009). Chemistry-transport models (CTMs) are essential to capture the regional forcing and impacts of aerosols.

For the last two decades, numerical CTMs have experienced significant improvements thanks to: the increase in high performance computing resources (Colette et al., 2014), the “online” coupling between meteorological and chemical fields (Zhang, 2008; Zhang et al., 2013a, 2013b; Kukkonen et al., 2012; Baklanov et al., 2014) and the improved knowledge of atmospheric processing. While the progress in simulating air quality is notable, many challenges remain for CTMs. The AQMEII (Air Quality Model Evaluation International Initiative) is a joint effort between North America and Europe to establish common methodologies for model evaluation with a focus on ozone and aerosols (Rao et al., 2011). Vautard et al. (2007) concluded that the majority of CTMs used for AQMEII captured the observed gas phase mean values and daily variability fairly well, except for city centres. The skill of aerosol simulations, however, is generally lower. Nopmongcol et al. (2012) highlighted the role of emissions and dilution in the performance of their air quality model. In their comprehensive evaluation of the online coupled CTM COSMO-ART, Knote et al. (2011) noted that ozone and  $\text{NO}_x$  were well reproduced;  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  were, on average, underestimated. Several processes needed to be improved in the model, such as wet scavenging, SOA formation, distribution and concentration in primary emissions of aerosol particles. Several authors have also mentioned lateral boundary conditions for aerosols as a source of uncertainties. Aksoyoglu et al. (2011) concluded that the offline-coupled CAMx (comprehensive air quality model with extensions) model reproduced the relative composition of aerosols very well over Switzerland but underestimated the absolute concentration by 20%. Tuccella et al. (2012) validated the online-coupled WRF/CHEM (Weather Research and Forecast – Chemistry) model against ground-based measurements over Europe. The model reproduced daily  $\text{PM}_{2.5}$  aerosol mass with a slight negative bias but underpredicted particulate sulphate by a factor of 2 and overpredicted ammonium and nitrate by about a factor of 2. Missing processes in the aqueous-phase could explain the differences (Ervens et al., 2011). Zhang et al. (2013a, 2013b) compared the offline-coupled WRF/Polyphemus with the online-coupled model WRF/CHEM-MADRID over Western Europe. No model was shown to be superior in terms of aerosol representation. Although the online WRF/CHEM-MADRID accounted for interactions between the meteorology and the chemistry, the model comparison showed that the simulation of atmospheric pollutant was mainly sensitive to the vertical structure, emissions and parameterisations for dry/wet depositions. Online biogenic emissions significantly improved the simulated temporal variations and magnitudes for most variables and for both models. Meteorological conditions also contribute to aerosol composition. Tulet et al. (2005) used the online-coupled Meso-NH (Mesoscale Non-Hydrostatic model) model to simulate a coastal summer pollution episode during the ESCOMPTE (Expérience sur Site pour Contraindre les Modèles de Pollution Atmosphérique et de Transport d’Emissions) campaign over southern Europe. Results showed good agreement between observed and simulated aerosol compounds. However, nitrate and ammonium were underestimated, probably due to an underestimation of relative humidity. Pollution levels were also controlled by continental advection of aerosols. Aouizerats et al. (2011) used Meso-NH to simulate a two-day period in the context of the CAPITOU (Canopy and Aerosol Particle Interactions in the Toulouse Urban Layer) field experiment in order to reproduce the spatial distribution of specific particle pollutants produced at regional and local scales. Their simulation using three nested domains (10 km, 2.5 km and 500 m horizontal resolution) showed that urban meteorology could locally affect the pollutant

concentrations by up to a factor of 5. Bègue et al. (2012) studied the evolution of dust optical properties during a major dust event, originating from northern Africa and advected over northwestern Europe. The impact was found to be large over the Netherlands, with a maximum of aerosol optical thickness close to 1.

This work fits in with the current effort to make a careful benchmarking of numerical CTMs against observational data and/or between models. Two online-coupled models, Meso-NH and WRF/CHEM, are evaluated over Europe during three one-day episodes here. The objective is to assess the capacity of the models to reproduce the magnitude of intra-day gas and aerosol fluctuations over Europe for specific episodes. These episodes were chosen from the seminal study of Freney et al. (2011) and occurred during contrasted meteorological conditions over Europe. Models are compared with daily and hourly ground-based observations of meteorological data, gases and aerosols and to vertical profiles of meteorological data and gaseous pollutants.

A first section describes the Meso-NH and WRF/CHEM models (Section 2). A second section concerns the measurement databases and the simulation set-up (Section 3). The evaluation of the meteorology, gases and  $\text{PM}_{2.5}$  aerosol at the regional scale is discussed in Section 4. The last section is dedicated to model performance in terms of gases and aerosols at three particular chosen sites where detailed observations of aerosol chemical composition are available (Section 5).

## 2. Model descriptions

Meso-NH (Lafore et al., 1998; Tulet et al., 2003) and WRF/CHEM (Grell et al., 2005) are two non-hydrostatic, and “on-line” mesoscale atmospheric models (Baklanov et al., 2014). Meso-NH is anelastic and WRF/CHEM is fully compressible. Both models simulate atmospheric phenomena with horizontal resolutions from a few metres (LES) to a few kilometres (synoptic-scale). Meso-NH is developed by the Laboratoire d’Aérodynamique et Météo-France. In the present study, version 4.9.3 of Meso-NH is implemented. It uses terrain-following  $z$  coordinates. WRF/CHEM is developed among the community and the code is controlled by NOAA/ESRL (National Oceanic and Atmospheric Administration/Earth System Research Laboratory) scientists. WRF/CHEM uses terrain-following Eta-coordinates and, in this study, the version 3.4.1 is used.

Several parameterisations have been integrated in these models for convection, cloud microphysics, turbulence, surface processes, gas chemistry, and aerosol composition (<http://mesonh.aero.obs-mip.fr/mesonh410> and [www.wrf-model.org](http://www.wrf-model.org)). The parameterisations used by the two models are reported in Table 1 and dynamical and physical parameterisations are detailed in Appendix A. Chemical options used here are detailed in the two next sections.

### 2.1. Gas-phase mechanism

The gas phase chemistry in Meso-NH was described by Suhre et al. (1998) and Tulet et al. (2003). The ReLACS2 scheme (Regional Lumped Atmospheric Chemical Scheme 2) used in this study is based upon a reduction of the original CACM mechanism (Caltech Atmospheric Chemistry Mechanism; Griffin et al., 2002, 2005). ReLACS2 is derived from a reduction by reactivity weighting towards the hydroxyl radical  $\text{OH}^\bullet$  developed by Crassier et al. (2000). It includes 82 prognostic gaseous chemical species and 363 reactions enabling the formation of SOA precursors to be addressed (Tulet et al., 2006), compared with 189 prognostic species and 361 reactions in CACM.

The gas phase chemical mechanism used in WRF/CHEM in the present study is RACM (Regional Atmospheric Chemistry Mechanism; Stockwell et al., 1997; Geiger et al., 2003). This mechanism includes 84 species and 252 reactions. It includes 16 aggregated anthropogenic species (alkanes, alkenes, toluene, xylene, cresol) and 3 aggregated biogenic species (isoprene, monoterpenes and sesquiterpenes) representing VOCs.

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