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

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## Abstract

Gas 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 normalized mean bias integrated over all European stations for both models. Daily gas chemistry was reproduced with normalized 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<sub>2.5</sub> 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 POA (Primary Organic Aerosols) in WRF/CHEM, suggesting a missing source for SOA (Secondary Organic Aerosols) mass in WRF/CHEM aerosol parameterization. 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<sub>4</sub><sup>2-</sup> in particles by both models was often overestimated and correlated with an underestimation of the SO<sub>2</sub> mixing ratio. The simulated masses of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in particles were always higher for Meso-NH than for WRF/CHEM, which was linked to a difference in NO<sub>x</sub> 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.



## 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 radiation (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 complexity, size, and phase. 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 (fires, 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 conditions and aerosol properties, aerosol particles act as cloud condensation nuclei with the potential to impact the precipitation pattern (Poschl et al., 2005; Duseck 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 et al., 2008; Zhang et al., 2013; Kukkonen et al., 2012; Baklanov et al., 2014) and the improved knowledge of atmospheric processing. While the progress in simulating air quality are 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 on-line 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 concentrations 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. (2013) 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 parameterizations 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

1 sur Site pour Contraindre les Modèles de Pollution Atmosphérique et de Transport d'Emissions") campaign over  
2 southern Europe. Results showed good agreement between observed and simulated aerosol compounds. However,  
3 nitrate and ammonium were underestimated, probably due to an underestimation of relative humidity. Pollution levels  
4 were also controlled by continental advection of aerosols. Aouizerats et al. (2011) used Meso-NH to simulate a two-day  
5 period in the context of the CAPITOUL ("Canopy and Aerosol Particle Interactions in the Toulouse Urban Layer") field  
6 experiment in order to reproduce the spatial distribution of specific particle pollutants produced at regional and local  
7 scale. Their simulation using three nested domains (10 km, 2.5 km and 500 m horizontal resolution) showed that urban  
8 meteorology could locally affect the pollutant concentrations by up to a factor of 5. Bègue et al. (2012) studied the  
9 evolution of dust optical properties during a major dust event, originating from northern Africa and advected over  
10 northwestern Europe. The impact was found to be large over the Netherlands, with a maximum of aerosol optical  
11 thickness close to 1.

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

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

24

## 25 **2 Model descriptions**

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

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

### 38 **2.1 Gas-phase mechanism**

39 The gas phase chemistry in Meso-NH was described by Suhre et al., (1998) and Tulet et al., (2003). The ReLACS2  
40 scheme (Regional Lumped Atmospheric Chemical Scheme 2) used in this study is based upon a reduction of the  
41 original CACM mechanism (Caltech Atmospheric Chemistry Mechanism; Griffin et al., 2002; Griffin et al., 2005).  
42 ReLACS2 is derived from a reduction by reactivity weighting towards the hydroxyl radical OH<sup>•</sup> 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; Geinger 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. Both ReLACS2 and RACM allow O<sub>3</sub> chemistry to be simulated for a range of conditions found in remote areas to polluted urban sites, from the surface to the upper troposphere. In addition, ReLACS2 was designed to simulate the formation of SOA precursors. For both models, the photolytic rates are calculated using the TUV (Tropospheric Ultraviolet and Visible; <https://www2.acd.ucar.edu/modeling/tropospheric-ultraviolet-and-visible-tuv-radiation-model>) radiation model. The version 5.0 (Madronich and Flocke; 1999) is used for Meso-NH and the version 4.1 (Madronich et al., 1998; McKenzie et al., 2007) for WRF/CHEM.

## 2.2 Aerosol scheme

Emissions, nucleation, coagulation, condensation, dry deposition, sedimentation, and advective and diffusive transport are considered as processes modifying the aerosol population in both models. The evolutions of aerosol size distribution by aerosol microphysics (nucleation, coagulation, sedimentation and dry deposition) and of aerosol chemical composition are predicted respectively by the ORILAM-SOA (Organic Inorganic Lognormal Aerosols Model; Tulet et al., 2005; Tulet et al., 2006) model in Meso-NH and by the MADE-VBS (Modal Aerosol Dynamics Model for Europe; Ackermann et al., 1998, Volatility Basis Set; Donahue et al., 2006) model in WRF/CHEM. Assuming that aerosols are internally mixed (each size class corresponds to a single chemical composition), the particle size distribution of the aerosol is represented by three overlapping lognormal modes (Aitken, accumulation and coarse modes) in these two schemes.

The gas to particle conversion for the inorganic species is handled by the EQSAM model (Equilibrium Simplified Aerosols Model; Metzger et al., 2002) in Meso-NH and by MADE, a modified version of MARS (Binkowski and Shankar, 2005), in WRF/CHEM, both including sulphuric and nitric acid, ammonia and water vapour.

To simulate SOA formation, two different approaches are used in Meso-NH and WRF/CHEM. In Meso-NH, the partitioning of the low volatility organic species between the gas and aerosol phases is based on the thermodynamic equilibrium scheme MPMPO (Model to Predict the Multiphase Partitioning of Organics; Griffin et al., 2003; Griffin et al., 2005). It considers the aerosol phase divided into a liquid organic phase and an aqueous phase. Hydrophobic compounds condense on the organic liquid phase through Raoult's law and hydrophilic compounds condense mainly on the aqueous phase according to the Henry's law amount of liquid water and the pH computed initially by EQSAM (Pun et al., 2002). Finally, 10 groups of SOA (8 from anthropogenic sources and 2 from biogenic sources) are considered in the MPMPO model.

To simulate SOA formation in WRF/CHEM, a new parameterization is used: the VBS set (Volatility Basis Set; Donahue et al., 2006), recently coupled by Ahmadov et al. (2012) to the aerosol module MADE and to the gas phase chemical mechanism RACM in the WRF/CHEM model. In this parameterization, the oxidation of the VOCs by the hydroxyl radicals OH, O<sub>3</sub> or nitrate radicals leads to the production of organic species, which partition between the gas and the particle phases according to their volatility. The volatility spectrum is divided into four bins with saturation vapour concentrations ranging from 1 to 1000  $\mu\text{g m}^{-3}$ . For each bin and each SOA precursor, an SOA yield based on smog chamber studies is defined. Yields are different for two regimes, high and low NO<sub>x</sub> conditions. Moreover, the photochemical aging of the first generation VOC oxidation products is taken into account by shifting mass from high volatility bins to lower ones when further oxidation processes occur.

## **2.3 Dry deposition**

In Meso-NH, the dry deposition of chemical species is treated according to the resistance concept of Wesely (1989) in the SURFEX model, which treats all surface processes (Masson et al., 2013). The deposition velocity is computed by determining the net flux of the species and its concentration at the surface. It depends strongly on surface conditions from the ISBA (Interaction Sol-Biosphère-Atmosphère) scheme for nature surface (Noilhan and Planton, 1989), from the TEB (Town Energy Budget) scheme for town surface (Masson, 2000) and from the sea surface scheme based on the Charnock (1955) formula. The deposition depends on the turbulence in the boundary layer and on the molecular diffusion, which lead gases to effective surface deposition. The surface resistance is based on solubility and equilibrium with vegetation-specific processes such as stomatal activities of leaves for nature surface. The mesophyll resistance and the external leaf uptake are considered as a sink for some soluble gases (inorganic and organic) and are determined by their effective Henry's law constants. Dry deposition of aerosol particles is also based upon Wesely's (1989) parameterization according to the surface type.

In WRF/CHEM, the dry deposition of gas phase species is parameterized according to Erisman et al. (1994) also based on Wesely's (1989) scheme. The deposition velocity is proportional to the sum of the aerodynamic resistance between a specified height and the surface, the sub-layer resistance (dependant upon the molecular diffusion) and the surface resistance. The impact of the nature, the town and the sea surfaces on the exchanges between the soil, the surface and the atmosphere is taken into account by the unified Noah Land Surface Model (Chen and Dudhia, 2001), which considers 24 land use categories (including urban and built-up lands, water bodies, snow and ice) and 16 soil categories according to the United States Geological Survey (USGS). However unlike in Meso-NH, the mesophyll resistances for all gases are assumed to be zero, thus increasing the dry deposition velocity. For organic species, the dry deposition velocity is parameterized as proportional to the dry deposition velocity of nitric acid. Since no data are available to constrain the model, the parameter that determines the fraction of nitric acid is set to 0.25 as defined by Ahmadov et al. (2012). For aerosol particles, the Wesely approach is used (Wesely and Hicks, 2000).

In both models, no wet deposition scheme is activated.

## **3 Observation databases and simulation set-up**

### **3.1 Databases**

Surface meteorological fields were compared to the ISD (Integrated Surface Database) database of NOAA's NCDC (National Climatic Data Center) (<http://cdo.ncdc.noaa.gov/pls/plclimprod/cdomain.abbrev2id>), which provides public access to hourly averages of meteorological surface parameters over Europe. Wind direction (WD), wind speed (WS), temperature and dewpoint were used for this study. Only stations below 700 m elevation were selected for the model evaluation in order to avoid bias due to terrain smoothing by the models (Jimenez and Dudhia, 2012).

The MOZAIC (Measurements of OZone, water vapour, carbon monoxide and nitrogen oxides by In-service Airbus airCraft) database provides measurements of meteorological and chemical fields made during a large number of commercial flights all over the world since 1993 (<http://www.iagos.fr/web/>; Marenco et al., 1998). These 20 years of measurements enable climatology and air quality studies to be made for the troposphere. The database includes data collected from the ground to the upper troposphere during take-offs and landings, with high temporal and 3D-spatial resolution. These vertical profiles are used here to evaluate meteorology and chemistry in the vertical direction. During landings and take-offs, data is collected every 4s, which corresponds approximately to 50-100 m in the vertical direction (Solazzo et al., 2013).

1 Surface chemical measurements were provided by AIRBASE (European AIR quality dataBASE)  
2 (<http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8>). AIRBASE provides hourly  
3 mean concentrations of O<sub>3</sub>, SO<sub>2</sub> (sulphur dioxide), NO<sub>x</sub> (nitrogen dioxide) and PM<sub>2.5</sub> bulk mass concentration for a large  
4 number of European stations, each characterized by its own kind of area (urban, rural, suburban) and its type of station  
5 (background, traffic, industrial). Only a subset of stations is used for the model evaluation, with the following criteria:  
6 stations must be below 700 m elevation (Nopmongcol et al., 2012) and must be classified as background rural stations.  
7 The study excludes urban or suburban stations and stations for which pollution is determined predominantly by traffic  
8 or industrial sources. Because of the low horizontal resolution and the static emission profiles used by the two models,  
9 local anthropogenic (point or mobile) sources, for which time-varying daily profiles are crucial, are not represented in  
10 the models.

11 The chemical composition of aerosol particles at ground level is compared with the AMS (Aerodyne Aerosol Mass  
12 Spectrometer) global database (<https://sites.google.com/site/amsglobaldatabase/>). This database provides hourly  
13 measurements of submicron non-refractory aerosol mass concentrations (NR-PM<sub>1</sub>) for sulphate (SO<sub>4</sub><sup>2-</sup>), ammonium  
14 (NH<sub>4</sub><sup>+</sup>), nitrate (NO<sub>3</sub><sup>-</sup>) and OM (Organic Matter) taken throughout Europe and broken down into several kinds of  
15 environment (urban, urban downwind, rural/remote, aircraft). AMS measurements were available at 3 stations  
16 distributed over Europe for the three simulated case studies: the puy de Dôme station, the Melpitz station and the K-  
17 Puszta station.

18 The puy de Dôme (pdD) station is located in the centre of France (45°77N; 2°96E) at 1465 m a.s.l. This site is  
19 surrounded by agricultural land and forest. As discussed in Sect. 4.1, air masses characterized by specific chemical  
20 compositions reach the pdD station according to the season and the origin of the air masses (Freney et al., 2011).

21 Melpitz (51°54 N; 12°93 E, 86 m a.s.l) is a German station located 50 km east of Leipzig. This site is representative of a  
22 larger rural area in Saxony with almost no local anthropogenic sources. Two main wind directions are observed at  
23 Melpitz station. When the wind blows from the southwest, the air mass reaching Melpitz is mostly a modified maritime  
24 air mass and is less polluted than when the wind direction is from the east, bringing Melpitz a dry continental air mass  
25 composed of anthropogenic pollutants (Spindler et al., 2010).

26 K-Puszta (46°96N; 19°58E, 136 m a.s.l) station is located in Hungary; 80 km southeast of Budapest; in a rural,  
27 continental environment. This station is surrounded by forest and is a good site for studying the formation of SOA from  
28 biogenic VOCs (BVOCs). Maenhaut et al., (2007) determined the chemical aerosol characteristics for the period 24  
29 May-29 June 2006, period in which two different flows occurred (an oceanic air mass from the north west and a static  
30 continental air mass). Results showed that the mass concentration of aerosols was higher for the warm period than for  
31 the cold one and that OM made the greatest contribution by far to the PM<sub>2.5</sub> and PM<sub>10</sub> mass concentration.

### 32 **3.2 Simulation set-up**

33 An intensive campaign (Freney et al., 2011) took place at the pdD station (45°77N, 2°96E) in autumn 2008, winter 2009  
34 and summer 2010, and documented chemical and microphysical properties of atmospheric aerosol particles. From these  
35 measurements, Freney et al., (2011) showed, firstly, that for each season, the aerosol was characterized by its own  
36 chemical composition and, secondly, that the origin of the air mass influenced the mass concentration of aerosols. For  
37 this reason, it was interesting to simulate three cases extracted from this database, which cover three situations, one for  
38 3 March 2009, one for 26 June 2010 and one for 18 September 2008. The air masses reaching the pdD station for these  
39 days are representative of an oceanic, a continental and a Mediterranean environment respectively.

40 The computational domain extended over Europe. It has 360 x 360 grid points with a horizontal resolution of 10 km.  
41 The vertical grid was common to both models and had 50 levels, stretched to allow better definition in the boundary

layer. The first level was at 30 m and the last one was at about 19 km. Each simulation lasted for 96 h, of which 72 h concerned spin-up. The time step was 50 s.

The initial and boundary conditions for meteorology were provided by the ECMWF (European Centre for Medium-Range Weather Forecasts) analysis with a horizontal resolution of 0.5° and were forced every 6 h. The initial and boundary conditions for gaseous chemical species including inorganic nitrogen species, CO, SO<sub>2</sub>, NH<sub>3</sub>, NMVOCs (Non-methane VOCs), primary (BC, POA) and secondary (inorganics, SOA) aerosol species were taken from MOZART-4 (Model for Ozone and Related chemical Tracers, version 4) (Emmons et al., 2010) driven by meteorology from NCEP centres (National Centers for Environment Prediction). Boundary chemical fields were forced every 6h.

For Meso-NH, surface emissions of atmospheric compounds were taken from the ECCAD database (Emissions of atmospheric Compounds & Compilation of Ancillary Data, [http://eccad.sedoo.fr/eccad\\_extract\\_interface/JSF/page\\_login\\_jsf](http://eccad.sedoo.fr/eccad_extract_interface/JSF/page_login_jsf)), which compiles the main emissions datasets available for the community. The MACCity emissions dataset (MACC/CityZEN EU projects) (Van der Werf et al., 2006; Lamarque et al., 2010; Granier et al. 2011; Diehl et al. 2012) provided “off-line” monthly anthropogenic emissions for CO, NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, NMVOCs such as alkanes, alkenes, alcohols, aldehydes, ketones and aromatics, lumped into 21 species, and for primary aerosol species, with a grid resolution of 0.5°. The MEGANv2 model (Model of Emissions of Gases and Aerosols from Nature, version 2) (Guenther et al., 2006) gave “off-line” monthly net emission of gases and aerosols from vegetation into the atmosphere at 0.5° resolution (for NO<sub>x</sub> and VOCs). The monthly GFED3 (Global Fire Emissions Database, version 3; Van der Werf et al., 2010) database was used to represent biomass burning emissions (CO, NMVOCs, BC and POA) with a spatial resolution of 0.5°.

Emissions in WRF/CHEM also came from MACCity for anthropogenic species. Biogenic emissions were different: the MEGAN model was used in “on-line” configuration because this option was already available in WRF/CHEM. In MEGAN, emissions were estimated using a 1km x 1 km resolution map of vegetation defined for the year 2003. For biomass burning emissions, the daily FINN inventory (Fire INventory from NCAR) (Wiedinmyer et al., 2011) model was used in WRF/CHEM with a 1 km<sup>2</sup> spatial resolution.

For Meso-NH and WRF/CHEM, NO<sub>x</sub> emissions were assumed to be 70% NO and 30% NO<sub>2</sub>. In the chemical mechanisms CACM and RACM, in order to limit the number of model species, some similar organic compounds were grouped together based on the principle of reactivity weighting. Aggregation factors, computed by Middleton et al. (1990), were used for VOCs. For aerosol species, in WRF/CHEM, a conversion factor of 1.6 proposed by Bessagnet et al. (2008) was used to convert the emissions of organic carbon (OC) into particulate organic matter (POA). Then, 20% of BC and POA emissions were allocated to the Aitken mode of the aerosol distribution and 80% to the accumulation mode according to Ackermann et al. (1998). In Meso-NH, 5% of OC and BC were allocated to the Aitken mode and 95% to the accumulation mode, as, according to the actual knowledge, organic aerosol mass is assumed to be distributed mainly in the accumulation mode (Kanakidou et al., 2005). For both models, parameters for the three lognormal modes (total particle number concentrations, mean radius and standard deviation) are initialized from Asmi et al. (2011).

## 4 Evaluation of models at regional scale

### 4.1 Three contrasted meteorological situations over Europe

In general, both models showed similar meteorological patterns (Fig. 1). On 3 March 2009, a cyclonic circulation with a well-marked frontal zone was simulated west of 0°E associated with heavy precipitation behind the cold front (10-20 mm over United Kingdom). Clear-sky conditions and strong southerly winds developed ahead of the cold front with

1 wind speeds exceeding  $20 \text{ m.s}^{-1}$  over France. A second south-north rain band was simulated with weaker precipitation  
2 except over eastern Spain (up to 30 mm). On 26 June 2010, an anticyclone ( $55^\circ\text{N}$ ;  $10^\circ\text{E}$ ) prevents precipitation over  
3 northern Europe with strong northerly flow over northeastern Europe. Continental southern Europe experienced local  
4 storm precipitations (up to 20 mm) over mountain ranges. A second anticyclone centred on Russia induced a southerly  
5 flow over the extreme east of Europe and a second rain band with strong precipitation (up to 30 mm). On 18 September  
6 2008, three precipitation zones were simulated. The first was associated with a cold front in the northwest of the  
7 domain. The second was induced by convergent air masses over eastern Europe, and the third resulted from conflicting  
8 air masses over Spain and France. Southern France, northern Spain and northern Italy experienced strong precipitation.  
9 Both models simulated the contrasting meteorology and the precipitation over Europe for the three episodes described  
10 in Freney et al. (2011).

## 11 **4.2 Surface Meteorological fields**

12 NOAA surface data, coming from European meteorological centres, were compared with results from both models.  
13 For each selected surface meteorological station, the daily bias between observations and the two models was computed  
14 for WD10 (10m-Horizontal Wind Direction) ( $^\circ\text{N}$ ), WS10 (10m-Horizontal Wind Speed) ( $\text{m.s}^{-1}$ ), T2m (Air temperature  
15 at 2 m) (K) and Td2m (dewpoint at 2 m) (K) for the three chosen dates of 18 September 2008, 3 March 2009 and 26  
16 June 2010 (Fig. 2 for 18 September 2008; Fig. S1 and Fig. S2 in the supplement for the other two days).  
17 The corresponding daily NMB (Normalized Mean Bias), expressed as a percentage, is reported in Table 2 and defined  
18 as:

$$19 \quad NMB = 100 \frac{\sum_{i=1}^N (Mod_i - Obs_i)}{\sum_{i=1}^N Obs_i} \quad (1)$$

20 Results in Table 2 show that there were no significant differences between the models for the simulated WD10, as  
21 shown by the weak NMB (-3 to 1%). For 3 March 2009, the well-marked frontal zone (Fig. 1) was correlated with a  
22 zone of weak biases for WD10 (between  $-20^\circ\text{N}$  and  $20^\circ\text{N}$ ) from northern France and the United Kingdom to  
23 Scandinavia (Fig. S1). The NMB of WS10 for Meso-NH was low (-2 to 2%) while, for WRF/CHEM, it showed an  
24 overestimation (15 to 20%) (Table 2). This behaviour is visible on Fig. 2, Fig. S1 and Fig. S2 showing a higher bias on  
25 WS10 for WRF/CHEM than for Meso-NH. In general, except for coastal zone, the Meso-NH bias for WS10 is about  $\pm 2 \text{ m.s}^{-1}$ ,  
26 which is consistent with Aouizerats et al. (2011). For WRF/CHEM, previous studies had already pointed out a  
27 systematic overestimation of surface wind speed (Zhang et al., 2010; Tuccella et al., 2012; Wyszogrodzki et al., 2013).  
28 The NMB of T2m and Td2m was close to 0% for both models (Table 2). The large underestimation of T2m for  
29 WRF/CHEM for the 26 June 2010 (Fig. S2) is consistent with results from Tuccella et al. (2012) showing a cold bias  
30 during the spring-summer period over Europe with WRF/CHEM.

31 Determining the origin of these biases in the two models would require an extensive study of the current uncertainties in  
32 surface-atmosphere coupling, which is beyond the scope of this paper. Simulated near-surface variables depend on  
33 surface conditions, including soil moisture and temperature (Sutton et al. 2006), land surface characteristics (land use,  
34 land cover, vegetation), and the coupling between these surface parameters within the land-surface model (LSM) and  
35 boundary layer parameterizations (Liu et al. 2006; Trier et al., 2008; Misenis et al., 2010; Noilhan et al., 2011). The  
36 parameterization of cloud microphysics and radiation may represent additional sources of biases for temperature. In a  
37 recent study, Zhang et al. (2013a) emphasized the role of the radiative impact of aerosols on shortwave radiation; they  
38 improved the simulation of surface temperature in WRF/CHEM. A recent study from Jimenez and Dudhia (2012)  
39 proposed a new parameterization to account for a subgrid-scale orography effect on surface winds for models at low  
40 resolution.

### 4.3 Vertical distribution of meteorological variables

The simulated vertical distribution of meteorological variables was compared with the high resolution vertical profiles of T (temperature), Td (dewpoint), WD (wind direction) and WS (wind speed) measured during take-off and landing of MOZAIC-IAGOS aircraft above Frankfurt (Fig. 3 for 18 September 2008 and Fig. S3 for 3 March 2009). There was no MOZAIC flight from Frankfurt on 26 June 2010. As Meso-NH and WRF/CHEM grids are horizontally and vertically close, the comparison between models was not spatially biased.

Fig. 3 shows that both models capture the vertical distribution of T and Td between the surface and 300 hPa on 18 September 2008. Below 850 hPa, both models tend to overestimate T and Td. Models simulate stratiform clouds between 500 hPa and 300 hPa as observed during flights where T was equal to Td. In the planetary boundary layer for 18 September 2008, there is a positive bias of T but the observed temperature inversion is well reproduced by the models early in the morning and in the evening. Measurements show a sudden increase in T at 850 hPa, which is less visible for both models. Observed Td decreases strongly at 850 hPa early in the morning, meaning a dry air mass limit. On 3 March 2009 (Fig. S3), the temperature vertical profile is well reproduced except at the surface where there is an underestimation at 10:00 UTC. Larger discrepancies are obtained with both models for Td. Fig. 1 shows that Frankfurt was at the edge of a rain band on 3 March 2009, which could explain the difference between local measurements and low-resolution model outputs. The models reproduce the northeasterly-easterly flow at the surface on 18 September 2008 and the southwesterly flow on 3 March 2009.

Overall, these four meteorological parameters were relatively well represented over Europe by Meso-NH and by WRF/CHEM. This constitutes a good basis for the following part of the study, concerning reactive gases and aerosol particles over Europe.

### 4.4 Gaseous pollutants at the surface

Simulated results for O<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub> are compared with the AIRBASE surface data. The number of rural and background stations for statistics varied between 111 and 259. As emissions have no diurnal variation, the bias of O<sub>3</sub>, NO<sub>x</sub> and SO<sub>2</sub> was calculated for daytime only, between 08:00 and 16:00 UTC (Fig. 4, Fig. S4 and Fig. S5 for 18 September 2008, 3 March 2009 and 26 June 2010 respectively, and Table 2). Fig. 5 shows the diurnal cycle of the median bias for these three chemical species.

Meso-NH tended to overestimate O<sub>3</sub> for the three days (Fig. 4, Fig. 5, Fig. S4 and Fig. S5) with a highest NMB (+11%) on 18 September 2008 (Table 2). Bias of NO<sub>x</sub> was slightly negative (-3% <NMB< -1%, Table 2). Looking at the diurnal cycle of the bias for NO<sub>x</sub>, Meso-NH showed poorer performance during night-time, with mean differences up to 6 µg.m<sup>-3</sup> for 50% of stations (Fig. 5). During daytime, the largest underestimations for NO<sub>x</sub> were found in industrial areas of the Po Valley (Italy), the Ruhr, the Netherlands and Austria, especially on 18 September 2008 (Fig. 4). In the Po Valley, for the three study cases (Fig. 4, Fig. S4 and Fig. S5) a positive bias of up to 20 µg.m<sup>-3</sup> of O<sub>3</sub> was associated with a strong negative bias of NO<sub>x</sub> (-5 µg.m<sup>-3</sup> for 50% of stations).

WRF/CHEM underestimated O<sub>3</sub> on 3 March 2009 and 26 June 2010 (-14% <NMB< -3%, Table 2) but gave good predictions of ozone during the third episode (NMB =1%, Table 2). Daytime NO<sub>x</sub> concentrations were reproduced to within -1 to 2% (Table 2), the worst prediction being made during night-time, with a systematically strong overestimation of up to 10 µg.m<sup>-3</sup> for 50% of stations (Fig. 5). Fig. 4, Fig. S4 and Fig S5 show patterns of maximum biases for ozone and NO<sub>x</sub> over the Po Valley and northern Europe for the WRF/CHEM model that are similar to those of Meso-NH.



Daytime values of NMB ranged between -2% and 0% for SO<sub>2</sub> in both models (Table 2). As shown in Fig. 5, WRF/CHEM tends to underestimate SO<sub>2</sub> for the three cases, with maximum bias during daytime (up to -1.5 µg.m<sup>-3</sup> for 50% of stations). Except in the Po Valley, during daytime, the level of accuracy of both models for the prediction of ozone, NO<sub>x</sub> and SO<sub>2</sub> at the surface is acceptable for regulatory application (AQD, 2008). The maximum bias for both models over the Po Valley is suspected to be due to the absence of diurnal variation of anthropogenic emissions (underestimation for NO<sub>x</sub> and overestimation for ozone). In the same way, both models showed a positive bias for NO<sub>x</sub> during night-time due to this absence of diurnal variation. This effect was also visible on the diurnal cycle of SO<sub>2</sub> bias for both models. However, for ozone bias, the link with the diurnal cycle of anthropogenic emissions is indirect due to the non-linearity of ozone chemistry. For instance, Tao et al. (2004) changed the temporal emission profiles (uniform vs. time-varying) of anthropogenic species and found no impact on daytime ozone concentrations. In the same idea, Menut et al. (2012) increased the NO<sub>2</sub> concentrations by 10-20% in their model using new hourly fluxes representative of traffic emissions with a moderate impact on ozone (0-7% decrease) and the most important changes occurred during night-time. Among potential sources of errors in the models, the chemical lateral boundary conditions are essential to capture the variability of pollution inflow and provide background concentrations, to which local production is added (Tang et al., 2007, Pfister et al., 2011). Global models provide chemical top and lateral boundary conditions for regional models but introduce uncertainties that are inherent in the model or due to the coupling process. This point is discussed in the next part. Obviously, differences between the two models could be related to differences in chemical reaction schemes coupling to differences between thermodynamical modules for gas to particles conversion, dynamics of the planetary boundary layer (venting of pollutants) and dry deposition parameterizations.

#### 4.5 Vertical distribution of ozone and carbon monoxide

The simulated vertical distributions of O<sub>3</sub> and CO were assessed using measurements from the MOZAIC database (Fig. 6 for 18 September 2008 and Fig. S6 for 3 March 2009; no data for 26 June 2010).

CO is a significant trace gas in the troposphere, which strongly influences the concentrations of oxidants such as hydroxyl radical (OH.) and O<sub>3</sub>. CO is mainly emitted by anthropogenic activities and has a lifetime of a few months in the boundary layer (BL) and the free troposphere (FT) (Seinfeld and Pandis, 1998).

Above 9-10 km altitude, O<sub>3</sub> and CO mixing ratios and their daily variability are controlled by stratosphere-troposphere exchanges, venting by convective clouds, chemical reactions and long-range transport. The vertical profiles in Fig. 6 show that both models fail to reproduce a well-marked stratospheric dry ozone-enriched intrusion. Both models underestimate O<sub>3</sub> with negative biases of up to 35 ppbv on 18 September 2008 (Fig. 6 at 0700 UTC) and 200 ppbv on 3 March 2009 (Fig. S6). Stratospheric air corresponds to poor CO mixing ratios. CO is overestimated at these altitudes with a large positive bias of 50 ppbv on 18 September 2008 (Fig. 6 at 0700 and 1000 UTC) and 3 March 2009 (Fig. S6 at 0400 UTC).

Below 500 m, the two models reproduced CO mixing ratios poorly, with large biases between -90 and 90 ppbv. These large discrepancies can possibly be attributed to two principal processes:

- The influence of local sources near the airport, the intensity and temporal variations of which are not captured by the models,
- The BL dynamics and the associated vertical venting of the pollutants. Interestingly, the worse biases are obtained during night-time, when the surface BL is expected to be low, favouring the accumulation of pollutants. The models performed better during daytime (1000 UTC on Fig. 6 and Fig. S6) as the depth of the BL increased.

1 Solazzo et al. (2013), which studied the performance of regional-scale air quality models in reproducing the vertical  
2 distribution of pollutants over the North American and European continents for 2006, also found such strong biases for  
3 CO in the BL.  
4 O<sub>3</sub> in the BL was generally well simulated with biases between -10 and 10 ppbv. The best scores were obtained for 3  
5 March 2009 (Fig. S6).  
6 Above the BL and below 9 km of altitude, biases for O<sub>3</sub> and CO were variable depending on the altitude and on the time  
7 of day. Both models exhibited similar vertical distributions of the biases. The vertical distribution of biases between O<sub>3</sub>  
8 and CO observations and the MOZART model interpolated on the Meso-NH vertical and horizontal grid at 0600 and  
9 1200 UTC (Fig. 6) clearly shows that the vertical profiles of CO and O<sub>3</sub> for both models are strongly influenced by the  
10 MOZART fields. The difference between CO biases for MOZART and the two models is the smallest (< 10 ppbv) in the  
11 entire vertical column above the BL. Below 500 m, simulated CO departs from the MOZART fields. The largest  
12 differences between the regional models and MOZART are found below 500 m for CO. Below 500 m, the MOZART  
13 model gives a strong overestimation of CO compared to the measurements (30-100 ppbv). The boundary conditions  
14 impact the simulated surface concentrations of CO but the biases are reduced in the regional models in comparison with  
15 MOZART bias because of local meteorology and chemical processes. The difference of CO bias between Meso-NH and  
16 WRF/CHEM at the surface and up to 3 km is certainly due to the difference in WD and in WS up to this altitude (Fig.  
17 3).  
18 The vertical profiles of O<sub>3</sub> biases for the two models also show the impact of MOZART fields. The bias of ozone  
19 MOZART fields is strongest in the upper troposphere and in the BL; this bias is reduced for Meso-NH and for  
20 WRF/CHEM. The difference between ozone biases for MOZART and both models is the weakest (5 ppbv) in the  
21 middle troposphere (2500-5000 m). This difference is stronger (20 ppbv) below 1000-1500 m. As for CO, the biases are  
22 reduced in the regional models in comparison with MOZART bias because of local meteorology and chemical  
23 processes. This is consistent with Solazzo et al., (2013) who found that model performance for ozone in the BL was  
24 generally good and mainly influenced by lateral boundary conditions and surface processes such as emissions, transport  
25 and photochemistry. Appel et al. (2012) underlined the influence of the forcing model for surface ozone over North  
26 America by improving the CMAQ model simulations using the global GEOS-Chem model instead of GEMS (Global  
27 and regional Earth-system Monitoring using Satellite and in-situ data) data (Schere et al., 2012) for the boundary  
28 conditions. However, if the surface ozone is mainly influenced by local photochemistry instead of the pollution inflow,  
29 the surface ozone mixing ratio can have low sensitivity large-scale chemical conditions (Szopa et al., 2009). For vertical  
30 profiles, Tang et al. (2007) found that the mean O<sub>3</sub> vertical profile below 3 km was insensitive to changes in chemical  
31 boundary conditions, whereas CO concentrations were sensitive throughout the troposphere.

#### 32 **4.6 PM<sub>2.5</sub> bulk aerosol at the surface**

33 Simulated daily mean PM<sub>2.5</sub> bulk aerosol mass concentrations over Europe are compared with AIRBASE stations  
34 measurements for the three chosen dates in Fig. 7. For both models, PM<sub>2.5</sub> mass concentration was computed by  
35 integrating Aitken and accumulation lognormal modes up to 2.5 micrometres in diameter. It included primary aerosol  
36 mass (BC and POA for organic), secondary inorganic (NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup>) and organic (SOA) aerosol components.  
37 The corresponding daily NMB (in %) was computed (Table 2) but it should be borne in mind that PM<sub>2.5</sub> measurements  
38 were available for only a small number of rural and background stations (6 and 12 respectively).  
39 Table 2 and Fig. 7 indicate that Meso-NH overestimates the PM<sub>2.5</sub> mass concentration with positive NMB between 6  
40 and 14% for the three dates. WRF/CHEM slightly underestimates the PM<sub>2.5</sub> mass concentration with a NMB between -3  
41 and 0% (Fig. 7 and Table 2). The same computation considering all stations (urban and suburban stations, traffic and

1 industrial sources), which represented more than 50 stations of AIRBASE (not shown here), changed the results  
2 considerably, with a large underestimation for WRF/CHEM (NMB between -17% and -1%) and a weaker positive  
3 NMB for Meso-NH (-5% to 8%).

4 The high observed values for the Po Valley in Italy (range from 20 to 40  $\mu\text{g.m}^{-3}$ ) were well captured by both models for  
5 the three cases despite an overestimation (ranging from 30 to 60  $\mu\text{g.m}^{-3}$ ) by Meso-NH and an underestimation (ranging  
6 from 5 to 25  $\mu\text{g.m}^{-3}$ ) by WRF/CHEM (Fig. 7). Over stations in the Netherlands,  $\text{PM}_{2.5}$  mass concentrations were  
7 relatively low. This was well reproduced by WRF/CHEM whereas Meso-NH overestimated the observations by 15 to  
8 20  $\mu\text{g.m}^{-3}$  (Fig. 7).

9 Several recent studies have shown that air quality models tend to underestimate  $\text{PM}_{2.5}$  mass concentrations over Europe.  
10 Appel et al. (2012) studied the performance of the CMAQ model over Europe and North America for the entire year of  
11 2006. They found a global underestimation of the  $\text{PM}_{2.5}$  mass concentration, mostly in winter (NMB=-55% and MB=-  
12 12.9  $\mu\text{g.m}^{-3}$ ), with better performance over France, Spain and Portugal. However, in their study, all the stations of the  
13 AIRBASE database were taken into account, which certainly translated, for WRF/CHEM, into a lack of anthropogenic  
14 sources or a too strong dry deposition. Tagaris et al., (2013) found an underestimation of  $\text{PM}_{2.5}$  mass concentration over  
15 most European countries during winter and summer 2006. Their paper discusses the chemical aerosol speciation, which  
16 indicates an insufficient contribution of OC to  $\text{PM}_{2.5}$  mass concentration in their model. By simulating gas and aerosol  
17 with WRF/CHEM over Europe, Tuccella et al (2012) showed an underestimation with a mean bias of  $\text{PM}_{2.5}$  mass (-  
18 7.3%) linked to a strong underestimation of the carbonaceous fraction by RADM2 mechanism.

19 The Meso-NH model overestimated the observed  $\text{PM}_{2.5}$  mass concentration. This could be due to a missing sink, to an  
20 underestimation of dry deposition, or to an overestimation of aerosol sources, as well as a too low BL height. The  
21 following section provides a detailed analysis of the chemical composition of the simulated aerosols and discusses the  
22 aerosol sources.

23

## 24 **5 Evaluation and discussion of sites**

25 Both models present significant biases on daily  $\text{PM}_{2.5}$  mass concentration whatever the situation. To explain these  
26 biases, it is necessary to look at the detail of the aerosol chemical composition by comparison between simulated and  
27 observed hourly mean mass concentration for each aerosol constituent. AMS measurements are available at three local  
28 sites: puy de Dôme, Melpitz and K-Pusztá for the three selected dates. In this section, the local mixing ratios of aerosol  
29 gaseous precursors are studied before analysing the aerosol hourly mean mass composition for the three local sites. For  
30 the pdD station, in order to avoid errors due to topography smoothing, all simulated results are taken at the real altitude  
31 of the station: 1465 meters.

### 32 **5.1 Gas precursors of aerosols**

33 Fig. 8 suggests that simulated  $\text{NH}_3$ ,  $\text{NO}_x$  and VOC mixing ratios have a behaviour partly linked to the season with  
34 higher values for 3 March 2009 and lower values for 26 June 2010 during daytime at the surface stations (Melpitz and  
35 K-Pusztá). At these stations, VOC,  $\text{NH}_3$ ,  $\text{NO}_x$  and  $\text{SO}_2$  show diurnal variations with higher mixing ratios at night and  
36 minimum values during the day, although this is visible to a lesser extent for 3 March 2009. These diurnal evolutions  
37 are due to the combination of the smallest BL height during the night and the constant temporal profiles of  
38 anthropogenic emissions in both models. The chemical species emitted accumulate during the night, when the BL  
39 height is small and no photochemistry takes place. The association of rain and cold temperatures simulated for 3 March  
40 2009 limits photochemistry, which partly explains the contrast with the two summer cases where the weather was  
41 warmer (Fig. 1). Another reason could be connected with the fact that the BL height is smaller in winter than in summer,

1 implying a weaker contrast between night-time and daytime mixing ratios. For K-Pusztta station, some data are available  
 2 on the EMEP database (Tørseth et al., 2012) for nitrogen dioxide, ammonia and sulphur dioxide as measurements on  
 3 filter packs corresponding to 24 hours averaged values between 07:00 on the day of interest to 07:00 on the next day.  
 4 However, because of the error already discussed on night-time simulated mixing ratios, the following discussion focuses  
 5 on simulated daytime values. For 3 March 2009, observed ammonia mixing ratio was 2.5 ppbv showing an  
 6 overestimation by both models, and especially by WRF/CHEM. However, both models simulated mixing ratio for  
 7 ammonia close to observations for 18 September 2008 and for 26 June 2010 (observed mixing ratio: 2.5 ppbv and 1.8  
 8 ppbv respectively). For  $\text{NO}_x$ , the comparison was done with  $\text{NO}_2$  as no  $\text{NO}$  observations are available but, as  $\text{NO}_2$   
 9 dominates  $\text{NO}_x$  mixing ratio, mixing ratio of  $\text{NO}_2$  can be seen as a lower limit for  $\text{NO}_x$ . Except for WRF/CHEM for the  
 10 3 March 2009, simulated  $\text{NO}_x$  mixing ratio for both models are of the same order of magnitude than observed  $\text{NO}_2$   
 11 mixing ratio (observed mixing ratios: 5.4 ppbv for 3 March 2009, 2.6 ppbv for 26 June 2010 and 3.5 ppbv for 18  
 12 September 2008). For  $\text{SO}_2$ , Meso-NH overestimated the observed values for the three episodes whereas WRF/CHEM  
 13 tended to slightly underestimate it (observed mixing ratios: 0.8 ppbv for 3 March 2009, 0.7 ppbv for 26 June 2010 and  
 14 0.25 ppbv for 18 September 2008).

15 At the mountainous pdD station, night-time observations, with mixing ratios lower than or similar to those found during  
 16 the day, are representative of FT air masses (Freney et al., 2011). During the day, depending on the season and the  
 17 meteorological conditions, the station lies in the FT (winter cases) or in the BL (summer) or in a transition area between  
 18 BL and FT (spring and autumn) (Freney et al., 2011). Results for the three cases show the same trend, with the station  
 19 probably located in a transition area between BL and FT during the day for both summer cases, which show no diurnal  
 20 cycle. Some EMEP data are available for pdD on the EMEP database for sulphur dioxide as hourly observations. As  
 21 these data are few dispersed, they were not added on Fig. 8 for clarity. For 3 March 2009, the observed mixing ratio was  
 22  $0.11 \pm 0.03$  ppbv; for 26 June 2010, it was  $0.12 \pm 0.04$  ppbv and for 18 September 2008, it was  $0.17 \pm 0.06$  ppbv. In  
 23 comparison to these values, Meso-NH overestimated observed  $\text{SO}_2$  mixing ratio whereas WRF/CHEM slightly  
 24 underestimated it. This trend is similar to K-Pusztta.

25 The  $\text{NH}_3$  mixing ratio typically ranges between 0.1 and 10 ppb over continents (Seinfeld and Pandis, 1998).  $\text{NH}_3$   
 26 mixing ratios higher than 10 ppbv are reported close to urban centres, or in recent plumes impacted by burning biomass  
 27 (Yokelson et al., 2003) or an industrial accident (Nowak et al., 2010). The main sinks of  $\text{NH}_3$  are dry and wet deposition  
 28 and its transfer in the particulate phase as  $\text{NH}_4^+$ .  $\text{NH}_3$  simulated by Meso-NH at the Melpitz station reached 55 ppbv at  
 29 0700 UTC on 18 September 2008 and WRF/CHEM simulated more than 30 ppbv for 3 March 2009 at 0800 and 1800  
 30 UTC (Fig. 8). These high mixing ratios of simulated  $\text{NH}_3$  can probably be attributed to the combination of high  
 31 emission rates and the absence of a sink by precipitation, as both models simulate precipitation for 3 March 2009 and  
 32 for 18 September 2008.

33 The largest differences in  $\text{NH}_3$  mixing ratio between the two models are simulated on 3 March 2009 at all stations, with  
 34 higher values simulated by WRF/CHEM. Simulated  $\text{NO}_x$  mixing ratios are close except for 3 March 2009, when values  
 35 simulated by WRF/CHEM are higher, as already shown on Fig. 5. VOCs simulated by Meso-NH are almost twice those  
 36 given by WRF/CHEM at the three stations and for the three dates. The maximum of VOCs is simulated for the Melpitz  
 37 (26 ppbv) and K-Pusztta (24 ppbv) stations on 18 September 2008 and for Melpitz (30 ppbv) on 26 June 2010 (Fig. 8).  
 38  $\text{SO}_2$  simulated by Meso-NH is three or four times higher than simulated by WRF/CHEM most of the time. The  
 39 behaviour of  $\text{NO}_x$  and  $\text{SO}_2$  is consistent with the conclusions drawn over Europe as seen above (cf. Sect. 4.4; Fig. 5).  
 40 The Comparison of the diurnal cycles of boundary layer height at the tree sites for the three dates show quite similar  
 41 results for both models. This means that the observed differences between  $\text{NO}_x$  and  $\text{SO}_2$  cannot be explained by  
 42 difference in venting of pollutants but more probably by differences between chemical mechanisms and

thermodynamical modules for gas to particles conversion. Indeed, discrepancies between  $\text{NO}_x$  and  $\text{SO}_2$  are highest when boundary layer height is small (during night-time at plain sites and all along the day at K-Pusztta for the 3 March 2009), so, when chemical species are more concentrated and chemical reactivity is higher.

## 5.2 Chemical composition of aerosol particles

The model performance in term of aerosol composition is evaluated with the AMS global database. This database provides the mass concentration of NR- $\text{PM}_1$  for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and OM, mentioned as ORG on Figs. 9 and 10. OM includes POM and SOM (for Primary and Secondary OM respectively). Here, OM measured by AMS is compared to the sum of anthropogenic primary organic aerosol (APOA) and secondary organic aerosol (SOA) simulated by Meso-NH and WRF/CHEM. Simulated and observed mass concentrations are compared as hourly mean to highlight how models are able to represent the detailed processes driving the aerosol composition and its temporal evolution. For each station, this temporal evolution of the aerosol mass composition on 18 September 2008 and on 3 March 2009 is presented in Fig. 9 and Fig. 10 respectively (cf. supplementary materials for 26 June 2010, Fig. S7). The daily mean mass concentration for each aerosol component is also indicated as well as their daily mean mass fraction.

In order to assess the ability of the models to reproduce aerosol chemical composition, statistical parameters such as Root Mean Square Error (RMSE), mean fractional bias (MFB) and mean fractional error (MFE), as discussed by Boylan and Russell (2006), are computed for each chemical component of aerosols and for the three case study cases (Tables 3, 4 and 5). When the performance criterion ( $|\text{MFB}| < 60\%$  and  $\text{MFE} < 75\%$ ) is satisfied, the level of accuracy is considered to be acceptable for standard modelling applications. When the performance goals ( $|\text{MFB}| < 30\%$  and  $\text{MFE} < 50\%$ ) are met, the level of accuracy is considered to be close to the best a model can be expected to achieve.

Models provide aerosol mass compositions for Aitken and accumulation modes for an aerodynamic diameter less than  $2.5 \mu\text{m}$  or  $\text{PM}_{2.5}$ . As AMS-observed NR- $\text{PM}_1$  mass concentration is compared to the simulated  $\text{PM}_{2.5}$  in the following, the models are expected to give an overestimation of aerosol mass concentration. The difference between  $\text{PM}_1$  and  $\text{PM}_{2.5}$  mass concentrations can be high (previous studies report  $\text{PM}_1/\text{PM}_{2.5}$  mass ratios between 55 and 75%) and can vary according to the meteorological conditions and the emission rates of chemical compounds (Aksoyoglu et al, 2011; Spindler et al., 2013). However, the proportions of the predominant chemical components in NR- $\text{PM}_1$  and simulated  $\text{PM}_{2.5}$  are expected to be of the same order.

### 5.2.1 At puy de Dôme

Observations (Fig. 9 and Fig. S7) suggest that, during the summer cases (26 June 2010 and 18 September 2008), the total aerosol mass was mostly composed of OM ( $15.69 \mu\text{g.m}^{-3}$  (53.05%) and  $2.39 \mu\text{g.m}^{-3}$  (35.09%) respectively). Meso-NH shows consistent results, with the highest contribution of simulated OM on 26 June 2010 ( $11.56 \mu\text{g.m}^{-3}$  (66.49%)) and a significant contribution on 18 September 2008 ( $5.13 \mu\text{g.m}^{-3}$  (43.77%)). SOA mass makes the highest contribution to OM for the three study cases. WRF/CHEM simulates the highest masses of OM during the two summer cases ( $0.69 \mu\text{g.m}^{-3}$  for 18 September 2008 and  $1.57 \mu\text{g.m}^{-3}$  for 26 June 2010) associated with the highest SOA mass fraction in OM. At the pdD station, the mass of OM simulated by WRF/CHEM is weaker by a factor of 10 than those simulated by Meso-NH.

On 3 March 2009 (Fig. 10), both models and measurements show a minimum of the mass concentration of  $\text{NH}_4^+$  ( $1.36 \mu\text{g.m}^{-3}$ ,  $0.93 \mu\text{g.m}^{-3}$  and  $1.28 \mu\text{g.m}^{-3}$  for Meso-NH, WRF/CHEM and observations, respectively). This day corresponds to the presence of an oceanic air mass (Fig. 1). Both models simulate a minimum of the mass concentration of  $\text{NO}_3^-$  ( $0.6 \mu\text{g.m}^{-3}$  and  $0.57 \mu\text{g.m}^{-3}$  respectively for Meso-NH and WRF/CHEM) for 18 September 2008, during the presence of a Mediterranean air mass. The observed mass concentration of  $\text{NO}_3^-$  is the weakest for this situation ( $0.93 \mu\text{g.m}^{-3}$ ) but still about twice that simulated by the models. Whatever the case, WRF/CHEM systematically underestimates the mass

1 concentration of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  at pdD by a factor of 2-4. Meso-NH underestimates the mass concentration of  $\text{NH}_4^+$   
2 and  $\text{NO}_3^-$  on 18 September 2008 and 26 June 2010 (Fig. 9 and Fig. S7). Meso-NH overestimates slightly  $\text{NH}_4^+$  and  $\text{NO}_3^-$   
3 on 3 March 2009.  
4 Concerning  $\text{SO}_4^{2-}$ , Meso-NH and WRF/CHEM simulate a comparable mass concentration at pdD for the three days.  
5 The highest mass concentrations and the highest difference between models for  $\text{SO}_4^{2-}$  are simulated for 18 September  
6 2008 ( $4.3 \mu\text{g.m}^{-3}$  and  $3.03 \mu\text{g.m}^{-3}$  respectively for Meso-NH and WRF/CHEM). The observed mass concentration is  
7 about 2, roughly 4 times weaker than the simulated ones for 18 September 2008 and 3 March 2009. In contrast, for 26  
8 June 2010, the observed mass concentration of  $\text{SO}_4^{2-}$  is 2-3 higher than the simulated ones.  
9 The performance criterion, computed for Meso-NH, is satisfied for  $\text{NH}_4^+$  on 18 September 2008 (Table 4), for  $\text{NH}_4^+$  and  
10 OM on 26 June 2010 (Table 5), and for  $\text{NH}_4^+$  on 3 March 2009 (Table 3). Concerning WRF/CHEM, the performance  
11 goal is only attained for  $\text{NH}_4^+$  on 3 March 2009.

## 12 **5.2.2 At Melpitz**

13 Fig. 9 and Fig. 10 indicate that Meso-NH (respectively WRF/CHEM) systematically overestimates (respectively  
14 underestimates) the observed OM at Melpitz. Measurements (Fig. 9, Fig. 10) show a more significant contribution of  
15 OM to the aerosol mass concentration for 18 September 2008 ( $3.42 \mu\text{g.m}^{-3}$  (47.71%)) than for 3 March 2009 ( $1.83$   
16  $\mu\text{g.m}^{-3}$  (13.58%)). Meso-NH reproduces the observed predominant contribution of OM to the aerosol mass  
17 concentration for 18 September 2008 ( $5.74 \mu\text{g.m}^{-3}$  (53.32%)) and a lower value for 3 March 2009 ( $3.34 \mu\text{g.m}^{-3}$   
18 (16.23%)). The speciation of OM provided by Meso-NH indicates a significant mass contribution of SOA for both 18  
19 September (88%) and 3 March 2009 (80%). Unlike the observations and Meso-NH, WRF/CHEM does not show a  
20 weaker mass of OM on 3 March 2009 than on 18 September 2008 ( $1.09 \mu\text{g.m}^{-3}$  (10.76%) versus  $1.03 \mu\text{g.m}^{-3}$  (20.56%)  
21 respectively). For WRF/CHEM, speciation for 3 March 2009 shows a higher contribution of POA to the OM mass  
22 concentration (89%) than on 18 September 2008 (72%).

23 For 3 March 2009, measurements show  $\text{NH}_4^+$  ( $6.60 \mu\text{g.m}^{-3}$  (49.02%)) and  $\text{SO}_4^{2-}$  ( $2.94 \mu\text{g.m}^{-3}$  (21.86%)) to be the main  
24 components of the aerosol. For this date, Meso-NH gives more than half of the total simulated aerosol mass  
25 concentration in the form of  $\text{NO}_3^-$  ( $10.94 \mu\text{g.m}^{-3}$  (53.14%)). Like the observations, WRF/CHEM shows a strong  
26 contribution of  $\text{SO}_4^{2-}$  ( $4.72 \mu\text{g.m}^{-3}$  (46.27%)) and  $\text{NH}_4^+$  ( $2.36 \mu\text{g.m}^{-3}$  (23.15%)).

27 On 18 September, the proportion of each inorganic aerosol component is quite similar ( $\sim 1 \mu\text{g.m}^{-3}$ ) for the observations  
28 and both models. While  $\text{NH}_4^+$  (44%) dominates the inorganic fraction in observations,  $\text{SO}_4^{2-}$  dominates the inorganic  
29 fraction simulated by WRF/CHEM (45%) and Meso-NH (38%).

30 On 3 March 2009, the two models meet the performance criterion for  $\text{SO}_4^{2-}$  and OM (Table 3). It is also met by Meso-  
31 NH for  $\text{NH}_4^+$  on 3 March 2009 (Table 3). On 18 September 2008, the performance criterion is only fulfilled by  
32 WRF/CHEM for  $\text{NO}_3^-$  and Meso-NH for OM. (Table 4). For this date, the two models overestimate the observed  $\text{SO}_4^{2-}$   
33 (Fig. 9).

## 34 **5.2.3 At K-Pusztta**

35 Observations at the K-Pusztta station are available only for 18 September 2008 (Fig. 9). As for the other two stations of  
36 pdD and Melpitz, the two models overestimate the observed  $\text{SO}_4^{2-}$ .

37 As found at the other two sites, observations show a high contribution of OM to the aerosol mass ( $3.09 \mu\text{g.m}^{-3}$   
38 (35.51%)), comparable to the other sites. Meso-NH simulates the same tendency with a significant contribution of OM  
39 ( $7.86 \mu\text{g.m}^{-3}$  (60.42%)), stronger than at the pdD and at Melpitz stations, for 18 September 2008. SOA makes the main  
40 contribution to OM (79%). For WRF/CHEM, OM mass also makes a higher contribution than the other two sites.

1 Unlike Meso-NH, WRF/CHEM simulates a high mass fraction (81%) of POA in OM. For this particular day, Meso-NH  
 2 overestimates the relative contribution of OM while WRF-CHEM underestimates it.  
 3 In the inorganic part of aerosols, WRF/CHEM and Meso-NH simulate a dominant contribution of  $\text{SO}_4^{2-}$  while  
 4 measurements show a dominant contribution of  $\text{NO}_3^-$  ( $2.36 \mu\text{g.m}^{-3}$ ). Moreover, the observed mass concentration of  $\text{NO}_3^-$   
 5 found at K-Pusztas is higher than at the other two sites on the same date. Both models underestimate  $\text{NO}_3^-$  (Fig. 9).  
 6 The model performance criterion is met for all species and for both models only at K-Pusztas for 18 September 2008  
 7 (Table 4). On this day, the Meso-NH performance goal is met for  $\text{NH}_4^+$  and  $\text{NO}_3^-$  at this station. Concerning  
 8 WRF/CHEM, the performance goal is reached for  $\text{NH}_4^+$  and for OM.

### 9 **5.3 Discussion**

10 Whereas the chemical sinks and sources are specific to each chemical compound, physical processes applied to the total  
 11 aerosol mass conserve the mass fraction of each aerosol compound.  
 12 The sudden decrease in measured aerosol mass concentration observed at pdD at 10:00 and 18:00 UTC on 18  
 13 September 2008 (Fig. 9) and at 11:00 on 26 June 2010 (Fig. S7) is certainly linked to wet scavenging of aerosol  
 14 particles by rain. Both models show rainy weather at pdD on 18 September 2008 and stormy weather with convective  
 15 cells is visible over the Massif Central Mountains on 26 June 2010 (Fig. 1 and Sect. 4.1). The effect of wet scavenging  
 16 on measured aerosol mass concentration is also observed at 10:00 UTC on 3 March 2009 (Fig. 10) and at 12:00 UTC on  
 17 18 September 2008 (Fig. 9). The weather simulated by both models is rainy at Melpitz on 3 March 2009 and on 18  
 18 September 2008. As wet scavenging is not activated in either model, the effect of this process is not visible. For Meso-  
 19 NH, as the mass fraction of each compound in the aerosol is generally well represented, this missing sink process  
 20 probably explains the overestimation of aerosol mass concentration. For instance, Chung and Seinfeld (2002) showed  
 21 that about 80% of SOA could be scavenged by cloud water and rainwater.  
 22 Variations of the total aerosol mass concentration at pdD can be also associated with the transition between FT and BL  
 23 and with the long distance transport of polluted air mass up to the pdD station. For instance, on 3 March 2009, both  
 24 observed and simulated mass concentrations of all compounds are weak and comparable (Fig. 10), indicating that the  
 25 pdD station is located inside the FT as expected for a winter air mass (Freney et al., 2011). In contrast, on 26 June 2010,  
 26 the observed mass concentrations are high for all compounds (Fig. S7), indicating that the pdD station was in the BL as  
 27 is typical in summer (Freney et al., 2011). Even though simulated local meteorological conditions show that the altitude  
 28 of the pdD station in the models is within the BL, simulated mass concentrations for this day show an underestimation  
 29 for all compounds in both models (Fig. S7). This can be attributed to, for example, a more pronounced influence of  
 30 Clermont-Ferrand city pollution in observations than in simulations. It can be noted that Meso-NH simulates a large  
 31 mass contribution of SOA on 26 June 2010, in agreement with the high mass contribution of OM observed (Fig. S7).  
 32 Observations (Fig. 9, Fig. S7) suggest that, in the summer cases (26 June 2010 and 18 September 2008), the aerosol is  
 33 mostly composed of OM at all sites. Freney et al. (2011) attributed the organic aerosol mass at the pdD station to  
 34 biogenic sources by analysing the correlation of low volatility oxygenated organic aerosol particles with fragments of  
 35 mass spectral markers of wood burning. For all cases, Meso-NH simulates a significant contribution of SOA to OM  
 36 mass concentration (>80%) whereas, for WRF/CHEM, OM is predominantly composed of POA (>70% for 3 March  
 37 2009 and for 18 September 2008 at Melpitz and K-Pusztas). However, for WRF/CHEM, at the pdD station for summer  
 38 cases, SOA dominates the OM mass concentration (57% for 18 September 2008 and 79% for 26 June 2010). These  
 39 results stress that the pdD station is farther from polluted sources than Melpitz and K-Pusztas, which are located in plains  
 40 and that, as expected, the SOA production is highest for summer cases in both models. OM mass concentration  
 41 simulated by WRF/CHEM is systematically underestimated by a factor varying between 2 and 10 in comparison with

1 Meso-NH and observations. This systematic underestimation of OM by WRF/CHEM is associated with VOC mixing  
 2 ratios that are systematically lower than those in Meso-NH (Fig. 8). This underestimation of OM is the strongest for 26  
 3 June 2010 at pdD, where the difference of VOC mixing ratios between WRF/CHEM and Meso-NH during daytime is  
 4 the highest. As both models simulate close values of POA at all stations and in all cases, the lowest OM mass  
 5 concentration simulated by WRF/CHEM comes from its lower ability to convert VOCs into SOA across the aging  
 6 constant of organic condensable vapours (OCVs) according to the oxidizing with the hydroxyl radicals used into the  
 7 VBS parameterization (cf. Sect. 2.2). Moreover, the low VOC mixing ratios in WRF/CHEM can be explained by a too  
 8 high dry deposition of VOCs due to a failure to consider mesophyll resistances and the use of a factor of 0.25 (cf. Sect.  
 9 2.3). An important difference between both models is also their chemical mechanism: RACM was designed to compute  
 10 the classical regional atmospheric chemistry whereas CACM treated in addition to this classical chemistry the formation  
 11 of gaseous organic precursors of SOA. This difference likely contributed to the low VOC mixing ratios associated to  
 12 low amounts of SOA simulated by WRF/CHEM. While anthropogenic emissions came from the same inventory for  
 13 both models, the difference of VOC mixing ratios is strengthened by the use of different biogenic and biomass burning  
 14 emissions. In particular, Meso-NH used, for biogenic emissions, a static inventory computed from the MEGAN model  
 15 while WRF/CHEM computed biogenic emissions from MEGAN model coupled online with its surface module.  
 16 For inorganic aerosol compounds, the differences between the two models and between models and observations are  
 17 very contrasted. For the pdD station, when the contribution of inorganic species mass to the total inorganic mass is  
 18 considered (not shown), observations and simulated inorganic mass contributions are very close on 26 June 2010. On 18  
 19 September 2008 and 3 March 2009, both models overestimate the inorganic mass contribution of sulphate. For 18  
 20 September, the inorganic mass contributions simulated by both models are in agreement whereas, for 3 March 2009, the  
 21 nitrate inorganic mass contribution and the nitrate mass concentration simulated by Meso-NH is higher than those  
 22 simulated by WRF/CHEM. On 3 March 2009 during the day, local weather conditions show a temperature up to 4 K  
 23 higher simulated by Meso-NH than by WRF/CHEM (not shown). In addition,  $\text{NH}_3$  and  $\text{NO}_x$  simulated by WRF/CHEM  
 24 are higher than simulated by Meso-NH on 3 March 2009 at all stations (Fig. 8). These differences in temperature and  
 25 gas-phase aerosol precursors on 3 March 2009 at pdD are likely reasons for the higher nitrate inorganic mass  
 26 contribution simulated by Meso-NH than by WRF/CHEM.  
 27 At the two plain stations (Melpitz and K-Puszt), both models underestimate  $\text{NH}_4^+$  mass concentrations. Simulated  
 28  $\text{NO}_3^-$  and  $\text{NH}_4^+$  mass concentrations are higher for Meso-NH than for WRF/CHEM (Fig. 9 and Fig. 10) except on 18  
 29 September 2008 at K-Puszt, where the  $\text{NH}_4^+$  mass concentration simulated by WRF/CHEM is slightly higher than that  
 30 simulated by Meso-NH. This general behaviour of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  mass concentrations is linked to a systematic  
 31 underestimation of simulated  $\text{NO}_x$  by Meso-NH compared to WRF/CHEM (Fig. 5, Fig. 8), which is maximum on 3  
 32 March 2009. This underestimation of simulated  $\text{NO}_x$  by Meso-NH may come from a more efficient transfer of oxidation  
 33 products of  $\text{NO}_x$  as nitric acid from the gas to the aerosol phase. Both models overestimate the  $\text{SO}_4^{2-}$  mass  
 34 concentration, except for Meso-NH at Melpitz on 3 March 2009 - linked to its high simulated  $\text{NO}_3^-$  mass concentration.  
 35  $\text{SO}_2$  simulated by WRF/CHEM is underestimated in comparison with observations (cf. Sect. 4.4 and Sect. 5.1). In both  
 36 models,  $\text{SO}_4^{2-}$  is formed by oxidation of  $\text{SO}_2$  in the gas phase. Therefore, the underestimation of  $\text{SO}_2$  in the gas phase by  
 37 WRF/CHEM, which is linked to its overestimation of aerosol  $\text{SO}_4^{2-}$ , seems to indicate a faster oxidation in WRF/CHEM  
 38 than in Meso-NH. On 18 September 2008 at K-Puszt and on 3 March 2009 at Melpitz, the  $\text{SO}_4^{2-}$  mass concentration  
 39 simulated by WRF/CHEM is higher by a factor of about two than that simulated by Meso-NH. However, as noted  
 40 before, the relative behaviour of  $\text{NH}_4^+$  mass concentration for these two cases is opposite: Meso-NH simulates more  
 41  $\text{NH}_4^+$  than WRF/CHEM at Melpitz on 3 March 2009 and less at K-Puszt on 18 September 2008. This behaviour can be  
 42 explained by the very high  $\text{NO}_3^-$  mass concentration simulated by Meso-NH at Melpitz on 3 March 2009, leading to



1 additional transfer of  $\text{NH}_3$  to the particulate phase. After ammonium sulphate was formed, the formation of  $\text{NH}_4\text{NO}_3$   
2 and subsequently of  $\text{NH}_4^+$  and  $\text{NO}_3^-$  ions in the aerosol phase is favoured by high relative humidity and low  
3 temperature, which was the case on 3 March 2009 at Melpitz as shown by the local simulated meteorological condition.  
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## 5 **6 Summary and Conclusion**

6 In this paper, our goal was to evaluate the ability of the Meso-NH and WRF/CHEM models to simulate three one-day  
7 cases over Europe. Meteorological fields (wind direction, wind speed, temperature, dewpoint and precipitation),  
8 gaseous species concentrations ( $\text{O}_3$ ,  $\text{NO}_x$ ,  $\text{SO}_2$ ) and aerosol particle compositions (inorganic and OM) have been  
9 compared model to model as well as to available measurements. These one-day duration cases came from Freney et al.  
10 (2011) and were chosen in order to simulate different seasons and air mass characteristics.

11 Simulated surface fields were compared to several surface hourly databases from stations (NOAA's NCDC for  
12 meteorology, AIRBASE for gaseous species and AMS global database for aerosol composition) over Europe. Simulated  
13 vertical profiles were evaluated above Frankfurt against airborne measurements of meteorological parameters and  
14 gaseous species from the MOZAIC database. The differences in simulated and observed CO and ozone at Frankfurt are  
15 mostly due to the initial fields provided by the MOZART CTM.

16 The two models reproduced the contrasted meteorological conditions for the three selected days. The comparison with  
17 vertical profiles of meteorological variables at Frankfurt and with observations at the surface was satisfactory.

18 The results concerning gaseous species at the surface are in good agreement with observations, mostly during the  
19 daytime for  $\text{NO}_x$  and  $\text{SO}_2$ . Globally,  $\text{O}_3$  presents the same diurnal evolution of bias between observed and simulated  
20 concentrations for both models with a quasi-constant gap of between 10 and 20  $\mu\text{g}\cdot\text{m}^{-3}$ . In the vertical direction,  $\text{O}_3$  and  
21 CO are well represented in the FT. The maximum bias appears near the surface for CO, with local emissions not  
22 correctly included in the models, and near the tropopause for CO and  $\text{O}_3$ , due to stratospheric intrusion not being well  
23 simulated by either model. The results suggest that the photochemistry is comparable for both models. Differences are  
24 probably due to simulated dynamics.

25 The simulated aerosol chemical composition is encouraging, with several model performance criteria met.

26 At the three local stations (pdD, Melpitz and K-Puszt), the systematic underestimation of simulated VOCs by  
27 WRF/CHEM is correlated with an underestimation of OM mass concentration in the aerosol phase. Moreover, OM  
28 simulated by WRF/CHEM presents a major contribution of POA, suggesting a missing source for SOA in WRF/CHEM  
29 parameterization. The contribution of OM is well simulated by Meso-NH in both proportion and quantity, with a higher  
30 contribution for summer cases. For Meso-NH, SOA make the major contribution to OM. Mass concentration of  $\text{SO}_4^{2-}$   
31 simulated by both models is often overestimated, a fact that is certainly associated with biases of  $\text{SO}_2$  mixing ratio.  
32 Simulated  $\text{NO}_3^-$  and  $\text{NH}_4^+$  mass concentrations are almost always higher for Meso-NH than for WRF/CHEM, in  
33 connection with differences in  $\text{NO}_x$  mixing ratio between the models. Finally, computations of model performance  
34 criterion (met if  $(|\text{MFB}| < 60\% \text{ and } \text{MFE} < 75\%)$  and model performance goals (met if  $(|\text{MFB}| < 30\% \text{ and } \text{MFE} < 50\%)$ )  
35 show that both models can be considered acceptable for standard modelling applications.

36 It should be kept in mind that the use of different biogenic and biomass burning emission inventories associated with  
37 the use of different gaseous chemical mechanisms in Meso-NH and WRF/CHEM lead to differences between the  
38 simulated amounts of gas phase precursors of aerosols, and thus aerosol amounts. In particular, Meso-NH model, using  
39 a gaseous chemical mechanism designed to compute the organic precursors of aerosols, shows comparable simulated  
40 amounts of SOA to observations at local sites.  
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**References**

Ackermann, I.J., Hass, H., Memmesheimer, M., Ebel, A., Binkowski, F.S., Shankar, U., 1998. Modal aerosols dynamics model for Europe: Development and first applications. *Atmos. Environ.* 32, 2981–2999.

Ahmadov, R., McKeen, S.A., Robinson, A.L., Bahreini, R., Middlebrook, A.M., de Gouw, J.A., Meagher, J., Hsie, E.-Y., Edgerton, E., Shaw, S., et al., 2012. A volatility basis set model for summertime secondary organic aerosols over the eastern United States in 2006. *J. Geophys. Res.* 117, D06301.

Aksoyoglu, S., Keller, J., Barmapadimos, I., Oderbolz, D., Lanz, V.A., Prevot, A.S.H., Baltensperger, U., 2011. Aerosols modelling in Europe with a focus on Switzerland during summer and winter episodes. *Atmos. Chem. Phys.* 11, 7355–7373.

Aouizerats, B., Tulet, P., Pigeon, G., Masson, V., Gomes, L., 2011. High resolution modelling of aerosol dispersion regimes during the CAPITOUL field experiment: from regional to local scale interactions. *Atmos. Chem. Phys.* 11, 7547–7560.

Appel, K.W., Chemel, C., Roselle, S.J., Francis, X.V., Hu, R.-M., Sokhi, R.S., Rao, S.T., Galmarini, S., 2012. Examination of the Community Multiscale Air Quality (CMAQ) model performance over the North American and European domains. *Atmos. Environ.* 53, 142–155.

AQD, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe. *Official Journal* No. 152.

Asmi, A., Wiedensohler, A., Laj, P., Fjaeraa, A.-M., Sellegri, K., Birmili, W., Weingartner, E., Baltensperger, U., Zdimal, V., Zikova, N., Putaud, J.-P., Marinoni, A., Tunved, P., Hansson, H.-C., Fiebig, M., Kivekäs, N., Lihavainen, H., Asmi, E., Ulevicius, V., Aalto, P.P., Swietlicki, E., Kristensson, A., Mihalopoulos, N., Kalivitis, N., Kalapov, I., Kiss, G., de Leeuw, G., Henzing, B., Harrison, R.M., Beddows, D., O'Dowd, C., Jennings, S.G., Flentje, H., Weinhold, K., Meinhardt, F., Ries, L., Kulmala, M., 2011. Number size distributions and seasonality of submicron particles in Europe 2008–2009. *Atmos. Chem. Phys.* 11, 5505–5538. doi:10.5194/acp-11-5505-2011.

Baklanov, A., Schlutzen, K., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael, G., Douros, J., Flemming, J., Forkel, R., et al., 2014. Online coupled regional meteorology chemistry models in Europe: current status and prospects. *Atmos. Chem. Phys.* 14, 317–398.

Bechtold, P., Bazile, E., Guichard, F., Mascart, P., Richard, E., 2001. A mass-flow convection scheme for regional and global models. *Q. J. R. Meteorol. Soc.* 127, 869–886.

1 Bègue, N., Tulet, P., Chaboureau, J.-P., Roberts, G., Gomes, L., Mallet, M., 2012. Long-range transport of Saharan dust  
2 over northwestern Europe during EUCAARI 2008 campaign: Evolution of dust optical properties by scavenging. *J.*  
3 *Geophys. Res.-Atmos.* 117, D17201.

4 Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Lioussé, C., Moukhtar, S., Pun, B., Seigneur, C.,  
5 Schulz, M., 2008. Regional modeling of carbonaceous aerosols over Europe - focus on secondary organic aerosols.  
6 *Journal of Atmospheric Chemistry* 61, 175–202.

7 Binkowski, F.S., Shankar, U., 1995. The Regional Particulate Matter Model: 1. Model description and preliminary  
8 results. *J. Geophys. Res.-Atmos.* 100, 26191–26209.

9 Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H.,  
10 Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens B., Zhang, X.Y., 2013. Clouds and Aerosols. In: *Climate*  
11 *Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the*  
12 *Intergovernmental Panel on Climate Change* [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung,  
13 A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and  
14 New York, NY, USA.

15 Bougeault, P., Lacarrere, P., 1989. Parameterization of Orography-Induced Turbulence in a Mesobeta-Scale Model.  
16 *Mon. Weather Rev.* 117, 1872–1890.

17 Boylan, J.W., Russell, A.G., 2006. PM and light extinction model performance metrics, goals, and criteria for three-  
18 dimensional air quality models. *Atmos. Environ.* 40, 4946–4959.

19 Carlson, T.N., Boland, F.E., 1978. Analyses of urban-rural canopy using a surface heat flux/temperature model. *J. Appl.*  
20 *Meteor.* 17, 998–1013.

21 Charnock, H., 1955. Wind stress on a water surface, *Q. J. R. Meteorol. Soc.* 81, 639–640, 1955.

22 Chen, F., Dudhia, J., 2001. Coupling an advanced land surface-hydrology model with the Penn State-NCAR MM5  
23 modeling system. Part I: Model implementation and sensitivity. *Mon. Weather Rev.* 129, 569–585.

24 Chou, M.D., Suarez, M.J., Ho, C.H., Yan, M.M.H., Lee, K.T., 1998. Parameterizations for cloud overlapping and  
25 shortwave single-scattering properties for use in general circulation and cloud ensemble models. *J. Clim.* 11, 202–214.

26 Chung, S.H., Seinfeld, J.H., 2002. Global distribution and climate forcing of carbonaceous aerosols. *J. Geophys. Res.-*  
27 *Atmos.* 107, 4407.

28 Cohard, J.M., Pinty, J.P., Bedos, C., 1998. Extending Twomey's analytical estimate of nucleated cloud droplet  
29 concentrations from CCN spectra. *J. Atmos. Sci.* 55, 3348–3357.

30 Colette, A., Bessagnet, B., Meleux, F., Terrenoire, E., Rouil, L., 2014. Frontiers in air quality modelling. *Geosci. Model*  
31 *Dev.* 7, 203–210.

32 Crassier, V., Suhre, K., Tulet, P., Rosset, R., 2000. Development of a reduced chemical scheme for use in mesoscale  
33 meteorological models. *Atmos. Environ.* 34, 2633–2644.

34 Cuxart, J., Bougeault, P., Redelsperger, J.L., 2000. A turbulence scheme allowing for mesoscale and large-eddy  
35 simulations. *Q. J. R. Meteorol. Soc.* 126, 1–30.

36 Diehl, T., Heil, A., Chin, M., Pan, X., Streets, D., Schultz, M., Kinne, S., 2012. Anthropogenic, biomass burning, and  
37 volcanic emissions of black carbon, organic carbon, and SO<sub>2</sub> from 1980 to 2010 for hindcast model experiments.  
38 *Atmos. Chem. Phys. Discuss.* 12, 24895–24954.

39 Donahue, N.M., Robinson, A.L., Stanier, C.O., Pandis, S.N., 2006. Coupled partitioning, dilution, and chemical aging  
40 of semivolatile organics. *Environ. Sci. Technol.* 40, 2635–2643.

1 Dusek, U., Frank, G.P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung,  
2 D., et al., 2006. Size matters more than chemistry for cloud-nucleating ability of aerosol particles. *Science* 312, 1375–  
3 1378.

4 Emmons, L.K., Walters, S., Hess, P.G., Lamarque, J.-F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison,  
5 D., Laepple, T., et al., 2010. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4  
6 (MOZART-4). *Geosci. Model Dev.* 3, 43–67.

7 Erisman, J., Vanpul, A., Wyers, P., 1994. Parametrization of surface-resistance for the quantification of atmospheric  
8 deposition of acidifying pollutants and ozone. *Atmos. Environ.* 28, 2595–2607.

9 Ervens, B., Turpin, B.J., Weber, R.J., 2011. Secondary organic aerosol formation in cloud droplets and aqueous particles  
10 (aqSOA): a review of laboratory, field and model studies. *Atmos. Chem. Phys.* 11, 11069–11102.

11 Fouquart, Y., Bonnel, B., 1980. Computations of solar heating of the earth's atmosphere: A new parametrization. *Beitr.*  
12 *Phys. Atmosph.* 53, 35–62.

13 Freney, E.J., Sellegri, K., Canonaco, F., Boulon, J., Hervo, M., Weigel, R., Pichon, J.M., Colomb, A., Prevot, A.S.H.,  
14 Laj, P., 2011. Seasonal variations in aerosols particle composition at the puy-de-Dome research station in France.  
15 *Atmos. Chem. Phys.* 11, 13047–13059.

16 Geiger, H., Barnes, I., Bejan, I., Benter, T., Spittler, M., 2003. The tropospheric degradation of isoprene: an updated  
17 module for the regional atmospheric chemistry mechanism. *Atmos. Environ.* 37, 1503–1519.

18 Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Gon, H.D. van der, Frost, G.J., Heil, A., Kaiser, J.W., Kinne, S.,  
19 Klimont, Z., et al., 2011. Evolution of anthropogenic and biomass burning emissions of air pollutants at global and  
20 regional scales during the 1980–2010 period. *Climatic Change* 109, 163–190.

21 Grell, G.A., Devenyi, D., 2002. A generalized approach to parameterizing convection combining ensemble and data  
22 assimilation techniques. *Geophys. Res. Lett.* 29, 1693.

23 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C. Eder, B., 2005. Fully coupled  
24 “online” chemistry within the WRF model. *Atmos. Environ.* 39,, 6957–6975, doi:10.1016/j.atmosenv.2005.04.027.

25 Griffin, R.J., Dabdub, D., Seinfeld, J.H., 2002. Secondary organic aerosol: 1. Atmospheric chemical mechanism for  
26 production of molecular constituents. *J. Geophys. Res.* 107(D17), 4332, doi:10.1029/2001JD000541.

27 Griffin, R.J., Nguyen, K., Dabdub, D., Seinfeld, J.H., 2003. A coupled hydrophobic-hydrophilic model for predicting  
28 secondary organic aerosols formation. *J. Atmos. Chem.* 44, 171–190.

29 Griffin, R.J., Dabdub, D., Seinfeld, J.H., 2005. Development and initial evaluation of a dynamic species-resolved model  
30 for gas phase chemistry and size-resolved gas/particle partitioning associated with secondary organic aerosols  
31 formation. *J. Geophys. Res.-Atmos.* 110, D05304.

32 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P.I., Geron, C., 2006. Estimates of global terrestrial isoprene  
33 emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmos. Chem. Phys.* 6, 3181–  
34 3210.

35 Jimenez, P.A., Dudhia, J., 2012. Improving the representation of resolved and unresolved topographic effects on surface  
36 wind in the WRF model. *J. Appl. Meteorol. Climatol.* 51, 300–316.

37 Hong, S.-Y., Noh, Y., Dudhia, J., 2006. A new vertical diffusion package with an explicit treatment of entrainment  
38 processes. *Monthly Weather Review* 134, 2318–2341.

39 Kain, J., Fritsch, J., 1990. A one-dimensional entraining detraining plume model and its application. *J. Atmos. Sci.* 47,  
40 2784–2802.

41 Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B.,  
42 Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter,

1 R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., Wilson, J., 2005. Organic aerosol and global climate  
2 modelling: a review. *Atmos. Chem. Phys.* 5, 1053–1123, doi:10.5194/acp-5-1053-2005.

3 Knote, C., Brunner, D., Vogel, H., Allan, J., Asmi, A., Äijälä, M., Carbone, S., van der Gon, H.D., Jimenez, J.L.,  
4 Kiendler-Scharr, A., et al., 2011. Towards an online-coupled chemistry-climate model: evaluation of trace gases and  
5 aerosols in COSMO-ART. *Geosci. Model Dev.* 4, 1077–1102.

6 Kukkonen, J., Olsson, T., Schultz, D.M., Baklanov, A., Klein, T., Miranda, A.I., Monteiro, A., Hirtl, M., Tarvainen, V.,  
7 Boy, M., et al., 2012. A review of operational, regional-scale, chemical weather forecasting models in Europe. *Atmos.*  
8 *Chem. Phys.* 12, 1–87.

9 Lafore, J.P., Stein, J., Asencio, N., Bougeault, P., Ducrocq, V., Duron, J., Fischer, C., Hereil, P., Mascart, P., Masson, V.,  
10 et al., 1998. The Meso-NH atmospheric simulation system. Part I: adiabatic formulation and control simulations. *Ann.*  
11 *Geophys.-Atmos. Hydrospheres Space Sci.* 16, 90–109.

12 Lamarque, J.-F., Bond, T.C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B.,  
13 et al., 2010. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and  
14 aerosols: methodology and application. *Atmos. Chem. Phys.* 10, 7017–7039.

15 Lee-Taylor, J., Madronich, S., Aumont, B., Baker, A., Camredon, M., Hodzic, A., Tyndall, G.S., Apel, E., Zaveri, R.A.,  
16 2011. Explicit modeling of organic chemistry and secondary organic aerosol partitioning for Mexico City and its  
17 outflow plume. *Atmos. Chem. Phys.* 11, 13219–13241.

18 Lin, S.J., Rood, R.B., 1996. Multidimensional flux-form semi-Lagrangian transport schemes. *Mon. Weather Rev.* 124,  
19 2046–2070.

20 Liu, Y., Li, F.L., 2006. A preliminary approach on the land surface temperature (LST) lapse rate of mountain area using  
21 MODIS data - art. no. 619907. In *Remote Sensing and Space Technology for Multidisciplinary Research*  
22 *and Applications*, Q. Tong, X. Chen, A. Huang, and W. Gao, eds. (Bellingham: Spie-Int Soc Optical Engineering), pp.  
23 19907–19907.

24 Madronich, S., McKenzie, R.L., Bjorn, L.O., Caldwell, M.M., 1998. Changes in biologically active ultraviolet radiation  
25 reaching the Earth's surface. *J. Photochem. Photobiol. B-Biol.* 46, 5–19.

26 Madronich, S., Flocke, S., 1999. The role of solar radiation in atmospheric chemistry, in: *Handbook of Environmental*  
27 *Chemistry*, edited by: Boule, P., Springer-Verlag, Heidelberg, 1–26.

28 Maenhaut, W., Raes, N., Chi, X., Cafmeyer, J., Wang, W., 2008. Chemical composition and mass closure for PM<sub>2.5</sub> and  
29 PM<sub>10</sub> aerosols at K-puszta, Hungary, in summer 2006. *X-Ray Spectrom.* 37, 193–197.

30 Marengo, A., Thouret, V., Nedelec, P., Smit, H., Helten, M., Kley, D., Karcher, F., Simon, P., Law, K., Pyle, J., et al.,  
31 1998. Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, An  
32 overview. *J. Geophys. Res.-Atmos.* 103, 25631–25642.

33 Masson V., 2000. A physically-based scheme for the urban energy budget in atmospheric models. *Bound. Layer Meteor.*  
34 1994, 357–397.

35 Masson, V., Champeaux, J.L., Chauvin, F., Meriguet, C., Lacaze, R., 2003. A global database of land surface parameters  
36 at 1-km resolution in meteorological and climate models. *J. Clim.* 16, 1261–1282.

37 Masson, V., Le Moigne, P., Martin, E., Faroux, S., Alias, A., Alkama, R., Belamari, S., Barbu, A., Boone, A., Bouysse,  
38 F., Brousseau, P., Brun, E., Calvet, J.-C., Carrer, D., Decharme, B., Delire, C., Donier, S., Essaouini, K., Gibelin, A.-L.,  
39 Giordani, H., Habets, F., Jidane, M., Kerdraon, G., Kourzeneva, E., Lafaysse, M., Lafont, S., Lebeaupin Brossier, C.,  
40 Lemonsu, A., Mahfouf, J.-F., Marguinaud, P., Mokhtari, M., Morin, S., Pigeon, G., Salgado, R., Seity, Y., Taillefer, F.,  
41 Tanguy, G., Tulet, P., Vincendon, B., Vionnet, V., Voldoire, A., 2013. the SURFEXv7.2 land and ocean surface platform  
42 for coupled or offline simulation of earth surface variables and fluxes. *Geosci. Model Dev.* 6, 929–960.

1 McKenzie, R.L., Bjorn, L.O., Bais, A., Ilyas, M., 2003. Changes in biologically active ultraviolet radiation reaching  
2 the Earth's surface. *Photochem. Photobiol. Sci.* 2, 5–15.

3 Metzger, S., Dentener, F., Pandis, S., Lelieveld, J., 2002. Gas/aerosols partitioning: 1. A computationally efficient  
4 model. *J. Geophys. Res.* 107, ACH 16–1.

5 Menut, L., Goussebaile, A., Bessagnet, B., Khvorostiyarov, D., Ung, A., 2012. Impact of realistic hourly emissions  
6 profiles on air pollutants concentrations modelled with CHIMERE. *Atmos. Environ.* 49, 233–244.

7 Metzger, S., Dentener, F., Pandis, S., Lelieveld, J., 2002. Gas/aerosol partitioning: 1. A computationally efficient model.  
8 *J. Geophys. Res.* 107, ACH 16–1.

9 Middleton, P., Stockwell, W., Carter, W., 1990. Aggregation and analysis of volatile organic-compound emissions for  
10 regional modeling. *Atmos. Environ. Part a-General Topics* 24, 1107–1133.

11 Misenis, C., Zhang, Y., 2010. An examination of sensitivity of WRF/Chem predictions to physical parameterizations,  
12 horizontal grid spacing, and nesting options. *Atmos. Res.* 97, 315–334.

13 Mlawer, E.J., Taubman, S.J., Brown, P.D., Iacono, M.J., Clough, S.A., 1997. Radiative transfer for inhomogeneous  
14 atmospheres: RRTM, a validated correlated-k model for the longwave. *J. Geophys. Res.-Atmos.* 102, 16663–16682.

15 Monin, A. S., Obukhov, A. M., 1954. Basic laws of turbulent mixing in the atmosphere near the ground. *Tr. Inst. Teor.*  
16 *Geofiz. Akad. Nauk SSSR* 24, 1963–1987.

17 Monks, P.S., Granier, C., Fuzzi, S., Stohl, A., Williams, M.L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger,  
18 U., Bey, I., et al., 2009. Atmospheric composition change - global and regional air quality. *Atmos. Environ.* 43, 5268–  
19 5350.

20 Morrison, H., Thompson, G., Tatarskii, V., 2009. Impact of cloud microphysics on the development of trailing stratiform  
21 precipitation in a simulated squall line: Comparison of one- and two-moment schemes. *Mon. Weather Rev.* 137, 991–  
22 1007.

23 Myhre, G., Shindell, D., Breon, F.-M., Collins, W., Fuglestad, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D.,  
24 Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhang, H., 2013. Anthropogenic and Natural  
25 Radiative Forcing. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth*  
26 *Assessment Report of the Intergovernmental Panel on Climate Change* [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor,  
27 S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge,  
28 United Kingdom and New York, NY, USA.

29 Noilhan, J., Planton, S., 1989. A Simple Parameterization of Land Surface Processes for Meteorological Models. *Mon.*  
30 *Weather Rev.* 117, 536–549.

31 Noilhan, J., Donier, S., Lacarrere, P., Sarrat, C., Le Moigne, P., 2011. Regional-scale evaluation of a land surface  
32 scheme from atmospheric boundary layer observations. *J. Geophys. Res.-Atmos.* 116, D01104.

33 Nopmongkol, U., Koo, B., Tai, E., Jung, J., Piyachaturawat, P., Emery, C., Yarwood, G., Pirovano, G., Mitsakou, C.,  
34 Kallos, G., 2012. Modeling Europe with CAMx for the Air Quality Model Evaluation International Initiative  
35 (AQMEII). *Atmos. Environ.* 53, 177–185.

36 Nowak, J.B., Neuman, J.A., Bahreini, R., Brock, C.A., Middlebrook, A.M., Wollny, A.G., Holloway, J.S., Peischl, J.,  
37 Ryerson, T.B., Fehsenfeld, F.C., 2010. Airborne observations of ammonia and ammonium nitrate formation over  
38 Houston, Texas. *J. Geophys. Res.-Atmos.* 115, D22304.

39 O'Donnell, D., Tsigaridis, K., Feichter, J., 2011. Estimating the direct and indirect effects of secondary organic aerosols  
40 using ECHAM5-HAM. *Atmos. Chem. Phys.* 11, 8635–8659.

41 Pergaud, J., Masson, V., Malardel, S., Couvreur, F., 2009. A parameterization of dry thermals and shallow cumuli for  
42 mesoscale numerical weather prediction. *Bound.-Layer. Meteor.* 132, 83–106.

1 Pfister, G.G., Parrish, D.D., Worden, H., Emmons, L.K., Edwards, D.P., Wiedinmyer, C., Diskin, G.S., Huey, G.,  
2 Oltmans, S.J., Thouret, V., et al., 2011. Characterizing summertime chemical boundary conditions for air masses  
3 entering the US West Coast. *Atmos. Chem. Phys.* 11, 1769–1790.

4 Pinty, J.-P., Jabouille, P., 1998. A mixed-phase cloud parameterization for use in mesoscale non-hydrostatic model:  
5 simulations of a squall line and of orographic precipitations. *Proc. Conf. of Cloud Physics*, Everett, WA, USA, Amer.  
6 Meteor. soc., Aug. 1999, 217 - 220.

7 Pope, C.A., Hansen, M.L., Long, R.W., Nielsen, K.R., Eatough, N.L., Wilson, W.E., Eatough, D.J., 2004. Ambient  
8 particulate air pollution, heart rate variability, and blood markers of inflammation in a panel of elderly subjects. *Environ*  
9 *Health Perspect* 112, 339–345.

10 Poschl, U., 2005. Atmospheric aerosols: Composition, transformation, climate and health effects. *Angew. Chem.-Int.*  
11 *Edit.* 44, 7520–7540.

12 Pun, B.K., Griffin, R.J., Seigneur, C., Seinfeld, J.H., 2002. Secondary organic aerosols 2. Thermodynamic model for  
13 gas/particle partitioning of molecular constituents. *J. Geophys. Res.-Atmos.* 107, AAC 4–1–AAC 4–15.

14 Rao, S.T., Galmarini, S., Puckett, K., 2011. Air Quality Model Evaluation International Initiative (AQMEII) Advancing  
15 the State of the Science in Regional Photochemical Modeling and Its Applications. *Bull. Amer. Meteorol. Soc.* 92, 23–  
16 30.

17 Rap, A., Scott, C.E., Spracklen, D.V., Bellouin, N., Forster, P.M., Carslaw, K.S., Schmidt, A., Mann, G., 2013. Natural  
18 aerosols direct and indirect radiative effects. *Geophys. Res. Lett.* 40, 3297–3301.

19 Rosenfeld, D., Lohmann, U., Raga, G.B., O'Dowd, C.D., Kulmala, M., Fuzzi, S., Reissell, A., Andreae, M.O., 2008.  
20 Flood or drought: How do aerosols affect precipitation? *Science* 321, 1309–1313.

21 Rueckerl, R., Schneider, A., Breitner, S., Cyrys, J., Peters, A., 2011. Health effects of particulate air pollution: A review  
22 of epidemiological evidence. *Inhal. Toxicol.* 23, 555–592.

23 Seinfeld, J.H., Pandis, S.N., 1998. *Atmospheric chemistry and Physics: From Air Pollution to Climate Change*. J. Wiley,  
24 New York.

25 Schere, K., Flemming, J., Vautard, R., Chemel, C., Colette, A., Hogrefe, C., Bessagnet, B., Meleux, F., Mathur, R.,  
26 Roselle, S., et al., 2012. Trace gas/aerosol boundary concentrations and their impacts on continental-scale AQMEII  
27 modeling domains. *Atmos. Environ.* 53, 38–50.

28 Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M.G., Huang, X.-Y., Wang, W., Powers,  
29 J.G., 2008. A description of the advanced research WRF version 3. NCAR Tech. Note.

30 Solazzo, E., Bianconi, R., Pirovano, G., Moran, M.D., Vautard, R., Hogrefe, C., Appel, K.W., Matthias, V., Grossi, P.,  
31 Bessagnet, B., et al., 2013. Evaluating the capability of regional-scale air quality models to capture the vertical  
32 distribution of pollutants. *Geosci. Model Dev.* 6, 791–818.

33 Spindler, G., Brüggemann, E., Gnauk, T., Grüner, A., Müller, K., Herrmann, H., 2010. A four-year size-segregated  
34 characterization study of particles PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> depending on air mass origin at Melpitz. *Atmos. Environ.* 44,  
35 164–173.

36 Spindler, G., Gruener, A., Mueller, K., Schlimper, S., Herrmann, H., 2013. Long-term size-segregated particle (PM<sub>10</sub>,  
37 PM<sub>2.5</sub>, PM<sub>1</sub>) characterization study at Melpitz - influence of air mass inflow, weather conditions and season. *J. Atmos.*  
38 *Chem.* 70, 165–195.

39 Stockwell, W.R., Kirchner, F., Kuhn, M., Seefeld, S., 1997. A new mechanism for regional atmospheric chemistry  
40 modeling. *J. Geophys. Res.* 102, 25847–25879.

41 Suhre, K., C. Mari, T. Bates, J. Johnson, R. Rosset, Q. Wang, A. Bandy, D. Blake, S. Businger, F. Eisels, B. Huebert, G.  
42 Kok, R. I. Mauldin, A. Prévôt, R. Schillawski, D. Tanner, D. Thornton, 1998, Physico chemical modeling of the first

1 aerosol characterization experiment (ace 1) lagrangian b, 1. a moving column approach, *J. Geophys. Res.*, 103, 16,433–  
2 16,455.

3 Sun, Y., Wang, Z., Dong, H., Yang, T., Li, J., Pan, X., Chen, P., Jayne, J.T., 2012. Characterization of summer organic  
4 and inorganic aerosols in Beijing, China with an Aerosols Chemical Speciation Monitor. *Atmos. Environ.* 51, 250–259.

5 Sutton, C., Hamill, T.M., Warner, T.T., 2006. Will perturbing soil moisture improve warm-season ensemble forecasts? A  
6 proof of concept. *Mon. Weather Rev.* 134, 3174–3189.

7 Szopa, S., Foret, G., Menut, L., and Cozic, A., 2009. Impact of large scale circulation on European summer surface  
8 ozone and consequences for modelling forecast. *Atmos. Environ.* 43, 1189–1195.

9 Tagaris, E., Sotiropoulou, R.E.P., Gounaris, N., Andronopoulos, S., Vlachogiannis, D., 2013. Air quality over Europe:  
10 modelling gaseous and particulate pollutants. *Atmos. Chem. Phys.* 13, 9661–9673.

11 Tang, Y., Carmichael, G.R., Thongboonchoo, N., Chai, T., Horowitz, L.W., Pierce, R.B., Al-Saadi, J.A., Pfister, G.,  
12 Vukovich, J.M., Avery, M.A., et al., 2007. Influence of lateral and top boundary conditions on regional air quality  
13 prediction: A multiscale study coupling regional and global chemical transport models. *J. Geophys. Res.* 112, D10S18.

14 Tao, Z.N., Larson, S.M., Williams, A., Caughey, M., Wuebbles, D.J., 2004. Sensitivity of regional ozone concentrations  
15 to temporal distribution of emissions. *Atmos. Environ.* 38, 6279–6285.

16 Tørseth, K., Aas, W., Breivik, K., Fjæraa, A. M., Fiebig, M., Hjellbrekke, A. G., Lund Myhre, C., Solberg, S., Yttri, K.  
17 E., 2012. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric  
18 composition change during 1972–2009. *Atmos. Chem. Phys.* 12, 5447–5481, doi:10.5194/acp-12-5447-2012.

19 Trier, S.B., Chen, F., Manning, K.W., LeMone, M.A., Davis, C.A., 2008. Sensitivity of the PBL and precipitation in 12-  
20 day simulations of warm-season convection using different land surface models and soil wetness conditions. *Mon.*  
21 *Weather Rev.* 136, 2321–2343.

22 Tuccella, P., Curci, G., Visconti, G., Bessagnet, B., Menut, L., Park, R.J., 2012. Modeling of gas and aerosols with  
23 WRF/Chem over Europe: Evaluation and sensitivity study. *J. Geophys. Res.* 117.

24 Tulet, P., Crassier, V., Solmon, F., Guedalia, D., Rosset, R., 2003. Description of the MESOScale NonHydrostatic  
25 Chemistry model and application to a transboundary pollution episode between northern France and southern France. *J.*  
26 *Geophys. Res.* 108(D1), 4021.

27 Tulet, P., Crassier, V., Cousin, F., Suhre, K., Rosset, R., 2005. ORILAM, a three-moment lognormal aerosol scheme for  
28 mesoscale atmospheric model: Online coupling into the Meso-NH-C model and validation on the Escompte campaign.  
29 *J. Geophys. Res.* 110, D18201.

30 Tulet, P., Grini, A., Griffin, R.J., Petitcol, S., 2006. ORILAM-SOA: A computationally efficient model for predicting  
31 secondary organic aerosols in three-dimensional atmospheric models. *J. Geophys. Res.* 111, D23208.

32 Twomey, D.S., 1959. The nuclei of natural cloud formation part II: The supersaturation in natural clouds and the  
33 variation of cloud droplet concentration. *Geofisica Pura E Applicata* 43, 243–249.

34 Van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Kasibhatla, P.S., Arellano Jr., A.F., 2006. Interannual  
35 variability in global biomass burning emissions from 1997 to 2004. *Atmos. Chem. Phys.* 6, 3423–3441.

36 Van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Mu, M., Kasibhatla, P.S., Morton, D.C., DeFries, R.S.,  
37 Jin, Y., van Leeuwen, T.T., 2010. Global fire emissions and the contribution of deforestation, savanna, forest,  
38 agricultural, and peat fires (1997–2009). *Atmos. Chem. Phys.* 10, 11707–11735.

39 Vautard, R., Builtjes, P.H.J., Thunis, P., Cuvelier, C., Bedogni, M., Bessagnet, B., Honore, C., Moussiopoulos, N.,  
40 Pirovano, G., Schaap, M., et al., 2007. Evaluation and intercomparison of Ozone and PM10 simulations by several  
41 chemistry transport models over four European cities within the CityDelta project. *Atmos. Environ.* 41, 173–188.



- 1 Wesely, M., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical-models.
- 2 *Atmos. Environ.* 23, 1293–1304.
- 3 Wesely, M.L., Hicks, B.B., 2000. A review of the current status of knowledge on dry deposition. *Atmos. Environ.*
- 4 34(12–14), 2261–2282.
- 5 Wicker, L. J., Skamarock, W. C., 2002. Time-splitting methods for elastic models using forward time schemes. *Mon.*
- 6 *Weather Rev.*, 130, 2088–2097.
- 7 Wiedinmyer, C., Akagi, S.K., Yokelson, R.J., Emmons, L.K., Al-Saadi, J.A., Orlando, J.J., Soja, A.J., 2011. The Fire
- 8 INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. *Geosci.*
- 9 *Model Dev.* 4, 625–641.
- 10 Wyszogrodzki, A.A., Liu, Y., Jacobs, N., Childs, P., Zhang, Y., Roux, G., Warner, T.T., 2013. Analysis of the surface
- 11 temperature and wind forecast errors of the NCAR-AirDat operational CONUS 4-km WRF forecasting system.
- 12 *Meteorol. Atmos. Phys.* 122, 125–143.
- 13 Yokelson, R.J., Christian, T.J., Bertschi, I.T., Hao, W.M., 2003. Evaluation of adsorption effects on measurements of
- 14 ammonia, acetic acid, and methanol. *J. Geophys. Res.* 108, 4649.
- 15 Yokelson, R.J., Bertschi, I.T., Christian, T.J., Hobbs, P.V., Ward, D.E., Hao, W.M., 2003. Trace gas measurements in
- 16 nascent, aged, and cloud-processed smoke from African savanna fires by airborne Fourier transform infrared
- 17 spectroscopy (AFTIR). *J. Geophys. Res.* 108, 8478.
- 18 Zhang, Y., Pan, Y., Wang, K., Fast, J.D., Grell, G.A., 2010. WRF/Chem-MADRID: Incorporation of an aerosols module
- 19 into WRF/Chem and its initial application to the TexAQS2000 episode. *J. Geophys. Res.* 115, D18202.
- 20 Zhang, Y., Sartelet, K., Wu, S.-Y., Seigneur, C., 2013. Application of WRF/Chem-MADRID and WRF/Polyphemus in
- 21 Europe – Part 1: Model description, evaluation of meteorological predictions, and aerosols–meteorology interactions.
- 22 *Atmos. Chem. Phys.* 13, 6807–6843.
- 23 Zhang, Y., Sartelet, K., Zhu, S., Wang, W., Wu, S.-Y., Zhang, X., Wang, K., Tran, P., Seigneur, C., Wang, Z.-F., 2013.
- 24 Application of WRF/Chem-MADRID and WRF/Polyphemus in Europe – Part 2: Evaluation of chemical concentrations
- 25 and sensitivity simulations. *Atmos. Chem. Phys.* 13, 6845–6875.

26

## Figure captions

Fig. 1: Daily averaged accumulated precipitation (in mm) over Europe simulated by Meso-NH (top) and WRF/CHEM (bottom) right to left for 3 March 2009, for 26 June 2010 and for 18 September 2008. Wind direction is represented by black vector in  $\text{m.s}^{-1}$  at 850 hPa at 12:00 UTC. Accumulation precipitation (in mm) is daily averaged, represented by colours and is the sum of accumulated explicit precipitation (rain, snow and graupel) and convective accumulated precipitation.

Fig. 2: Simulated daily biases for 18 September 2008 between models and observations at NOAA stations. Wind direction ( $^{\circ}\text{N}$ ) and wind speed ( $\text{m.s}^{-1}$ ) at 10 m, and temperature ( $^{\circ}\text{C}$ ) and dewpoint ( $^{\circ}\text{C}$ ) at 2 m are represented for Meso-NH and WRF/CHEM respectively top to bottom and right to left.

Fig. 3: Simulated (blue and red lines, for Meso-NH and WRF/CHEM respectively) and observed (black line) skew-T plots for temperature (solid lines), dewpoint (dashed lines), wind speed and wind direction (staffs and attached barbs). Observed vertical profiles were collected during take-off and landing of MOZAIC-IAGOS aircraft at Frankfurt airport on 18 September 2008 at four different times of day.

Fig. 4: Simulated daytime biases of mixing ratio (in  $\mu\text{g.m}^{-3}$ ) for 18 September 2008 between models and observations at “background” and “rural” AIRBASE stations.  $\text{O}_3$ ,  $\text{NO}_x$  and  $\text{SO}_2$  are represented for Meso-NH and WRF/CHEM respectively left to right and top to bottom.

Fig. 5: Diurnal cycle of median bias of mixing ratio of  $\text{O}_3$ ,  $\text{NO}_x$  and  $\text{SO}_2$  between simulated and observed values on the Airbase stations with mention “Background” and “rural”. The left column is for 3 March 2009, the middle one is for 26 June 2010 and the right one is for 18 September 2008. On each plot, the blue and red colours are for Meso-NH and WRF/CHEM respectively. Solid lines represent median, and dashed lines 25<sup>th</sup> and 75<sup>th</sup> percentiles. Black solid line separates negative from positive values.

Fig. 6: Vertical profiles of CO and ozone mixing ratio biases (in ppbv) between models and observations from take-off and landing of IAGOS-MOZAIC aircraft at Frankfurt airport on 18 September 2008 at four different times of day: Meso-NH (CO dark grey,  $\text{O}_3$  gold), WRF-Chem (CO black,  $\text{O}_3$  red). CO and  $\text{O}_3$  mixing ratio biases between MOZART model and observations at 06:00 and 12:00 UTC are superimposed on the 07:00 UTC and 10:00 UTC plots respectively (light grey for CO, light gold for  $\text{O}_3$ ).

Fig. 7: Maps of daily mean  $\text{PM}_{2.5}$  mass concentration (in  $\mu\text{g.m}^{-3}$ ) simulated by Meso-NH (top) and WRF/CHEM (bottom). Daily means from observations are represented by circles at “background” and “rural” AIRBASE stations for 3 March 2009 (left), for 26 June 2010 (middle) and for 18 September 2008 (right).

1 Fig. 8: Temporal evolution of mixing ratio of gas phase aerosol precursors simulated by Meso-NH (solid lines) and  
2 WRF/CHEM (dashed lines). Top to bottom for 3 March 2009, 26 June 2010 and 18 September 2008. Left to right at puy  
3 de Dôme, Melpitz and K-Pusztas stations. Precursors are represented with different colours:  $\text{NH}_3$  (grey),  $\text{SO}_2$  (red),  $\text{NO}_x$   
4 (blue) (left Y axis) and VOCs (green) (right Y axis).

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6 Fig. 9: Diurnal cycle of aerosol chemical composition in mass concentration ( $\mu\text{g.m}^{-3}$ ) of  $\text{NR-PM}_{10}$  on the left of each plot  
7 (top: measured by AMS, middle: simulated by Meso-NH, bottom: simulated by WRF/CHEM) for 18 September 2008 at  
8 puy de Dôme, Melpitz and K-Pusztas stations. On the right of each plot, the corresponding daily averaged fractional  
9 mass composition is also indicated by pie charts. Percentages in parentheses correspond to the mass fraction of the total  
10 aerosol mass for each compound.  $\text{NR-PM}_{10}$  include  $\text{NH}_4^+$  (orange),  $\text{SO}_4^{2-}$  (red),  $\text{NO}_3^-$  (blue) and ORG (green). Meso-NH  
11 and WRF/CHEM differentiate APOA (dark green) and SOA (light green) in OM.

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13 Fig. 10: Diurnal cycle of aerosol chemical composition in mass concentration ( $\mu\text{g.m}^{-3}$ ) of  $\text{NR-PM}_{10}$  on the left of each  
14 plot (top: measured by AMS, middle: simulated by Meso-NH, bottom: simulated by WRF/CHEM) for 3 March 2009 at  
15 puy-de-Dôme, Melpitz and K-Pusztas stations. On the right of each plot, the corresponding daily averaged fractional  
16 mass composition is also indicated by pie charts. Percentages in parentheses correspond to the mass fraction of the total  
17 aerosol mass for each compound.  $\text{NR-PM}_{10}$  include  $\text{NO}_3^-$  (blue),  $\text{SO}_4^{2-}$  (red),  $\text{NH}_4^+$  (orange) and ORG (green). Meso-NH  
18 and WRF/CHEM differentiate APOA (dark green) and SOA (light green) in OM.

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1 Table 1: Meso-NH and WRF/CHEM schemes

Scheme	Meso-NH	WRF/CHEM
Surface layer	SURFEX (Masson et al., 2013) ISBA for nature surface (Noilhan and Planton, 1989)	MM5 similarity (Monin and Obukhov, 1954; Carlson and Boland, 1978)
Land and ocean-surface schemes	TEB for town surface (Masson, 2000)	NOAH LSM (Chen and Dudhia, 2001)
Turbulence	Boundary layer with diagnostic ML (Bougeault and Lacarrere, 1989) Prognostic TKE (Cuxart et al., 2000)	YSU for boundary layer (Hong et al., 2006) Diffusion 2 <sup>nd</sup> order
Deep convection	KAFR (Kain and Fritsch, 1990; Bechtold et al., 2001)	Grell-Devenyi (Grell and Devenyi, 2002)
Shallow convection	EDKF (Pergaud et al., 2009)	
Shortwave radiation	ECMWF (Fouquart and Bonnel, 1980)	Goddard (Chou et al., 1998)
Longwave radiation	RRTM (Mlawer et al., 1997)	RRTMG (Mlawer et al., 1997)
Microphysics	ICE3 (Pinty and Jabouille, 1998)	Morrison double-moment scheme (Morrison et al., 2009)
Advection scheme	4 <sup>th</sup> order scheme centred on space and time for horizontal and vertical velocities PPM for meteorological and scalar variables (Lin and Rood, 1996) Leapfrog scheme for time integration	Horizontal advection for momentum and scalar is 5 <sup>th</sup> order Vertical advection for momentum and scalar is 3 <sup>rd</sup> order Moisture, scalars, TKE, chemical variables: monotonic transport Third-order Runge-Kutta (RK3) time integration scheme (Wicker and Skamarock, 2002)
Gas-phase mechanism	ReLACS2 (Tulet et al., 2006)	RACM (Stockwell et al., 1997)
Aerosols model	ORILAM-SOA (Tulet et al., 2006)	MADE (Ackermann et al., 1998) – VBS for SOA (Ahmadov et al., 2012)

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1 Table 2: Normalized Mean Bias (%) of meteorology, gaseous pollutants and PM<sub>2.5</sub> bulk mass concentration from  
2 Airbase and NOAA datasets for the three study cases on a daily basis or for daytime. For meteorology, the number of  
3 stations used for the statistics is between 1239 and 1397 depending on the parameter and on the day. For gaseous  
4 pollutants and for PM<sub>2.5</sub> mass concentration, the number of stations used is presented, respectively on Fig. 5 and Fig. 7.

	<b>03/03/09</b>		<b>26/06/10</b>		<b>18/09/08</b>	
	Meso-NH	WRF/CHEM	Meso-NH	WRF/CHEM	Meso-NH	WRF/CHEM
	Meteorology (daily)					
WD10	1	-1	-2	-3	-2	-3
WS10	-2	15	-1	16	2	20
T2m	0	0	0	0	0	0
Td2m	0	0	0	0	0	0
	Gaseous pollutants daytime					
O <sub>3</sub>	7	-3	4	-14	11	1
NO <sub>x</sub>	-3	2	-1	-1	-3	-2
SO <sub>2</sub>	0	-1	-1	-1	-1	-2
	Bulk mass (daily)					
PM <sub>2.5</sub>	12	0	14	-1	12	-3

1 Table 3: Root Mean Square Error (RMSE), Mean fractional bias (MFB) and error (MFE) for aerosol components for 3  
 2 March 2009 at puy de Dôme (FR) and Melpitz (DE) stations. In bold when the model performance criteria are met  
 3 ( $|\text{MFB}| < 60\%$  and  $\text{MFE} < 75\%$ ).

	puy de Dôme						Melpitz					
	MNH	WRF	MNH	WRF	MNH	WRF	MNH	WRF	MNH	WRF	MNH	WRF
	RMSE		MFB (%)		MFE (%)		RMSE		MFB (%)		MFE (%)	
$\mu\text{g.m}^{-3}$												
$\text{NH}_4^+$	0.51	0.59	<b>3</b>	<b>-31</b>	<b>37</b>	<b>49</b>	3.42	4.87	<b>-44</b>	-90	<b>46</b>	90
$\text{SO}_4^{2-}$	1.19	1.48	101	112	101	112	1.06	1.92	<b>-21</b>	<b>49</b>	<b>30</b>	<b>49</b>
$\text{NO}_3^-$	1.37	1.16	<b>20</b>	-74	<b>72</b>	84	9.00	0.39	138	<b>1</b>	138	<b>16</b>
ORG	0.93	0.91	<b>33</b>	-118	<b>58</b>	118	1.70	0.83	<b>60</b>	<b>-49</b>	<b>60</b>	<b>49</b>

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