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1 Evaluation of Meso-NH and WRF/CHEM simulated gas and 2 aerosol chemistry over Europe based on hourly observations

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13

14 Abstract

15 Gas and aerosol chemistry of 10 km -resolution mesoscale models Meso-NH and WRF/CHEM were evaluated on three
16 cases over Europe. These one-day duration cases were selected from Freney et al. (2011) and occurred on contrasted
17 meteorological conditions and at different seasons: a cyclonic circulation with a well-marked frontal zone on winter, an
18 anti-cyclonic situation with local storm precipitations on summer and a cold front in the northwest of Europe associated
19 to a convergence of air masses over eastern Europe and conflicting air masses over Spain and France on autumn. To
20 assess the performance of the two models, surface hourly databases from observation stations over Europe were used,
21 together with airborne measurements. For both models, the meteorological fields were in good agreement with the
22 measurements for the three days. Winds presented the largest normalized mean bias integrated over all European
23 stations for both models. Daily gas chemistry was reproduced with normalized mean biases between -14 and 11 %, a
24 level of accuracy that is acceptable for policy support. The two models performances were degraded during night-time
25 quite likely due to the constant primary species emissions. The PM_{2.5} bulk mass concentration was overestimated by
26 Meso-NH over Europe and slightly underestimated by WRF/CHEM. The absence of wet deposition in the models partly
27 explains the local discrepancies with the observations. More locally, the systematic low mixing ratio of volatile organic
28 compounds in the gas phase simulated by WRF/CHEM at three stations was correlated with the underestimation of OM
29 (Organic Matter) mass in the aerosol phase. Moreover, this mass of OM was mainly composed of anthropogenic POA
30 (Primary Organic Aerosols) in WRF/CHEM, suggesting a missing source for SOA (Secondary Organic Aerosols) mass
31 in WRF/CHEM aerosol parameterization. The contribution of OM was well simulated by Meso-NH, with a higher
32 contribution for the summer case. For Meso-NH, SOA made the major contribution to the OM mass. The simulation of
33 the mass of SO₄²⁻ in particles by both models was often overestimated and correlated with an underestimation of the
34 SO₂ mixing ratio. The simulated masses of NO₃⁻ and NH₄⁺ in particles were always higher for Meso-NH than for
35 WRF/CHEM, which was linked to a difference in NO_x mixing ratio between the models. Finally, computations of
36 model performance criterion and model performance goals show that both models can be considered acceptable for
37 standard modelling applications. In particular, Meso-NH model, using a gaseous chemical mechanism designed to
38 compute the organic precursors of aerosols, shows comparable simulated amounts of SOA to observations at local sites.

39

1 **1 Introduction**

2 Air pollutants have an effect on human health (Pope et al., 2004; Rueckerl et al., 2011), ecosystems and regional climate
3 (Monks et al., 2009). Greenhouse gases impact the climate primarily through shortwave and longwave radiation (Myhre
4 et al., 2013), while aerosols, in addition, affect the climate through cloud-aerosol interactions (O'Donnell et al., 2011;
5 Rap et al., 2013; Boucher et al., 2013).

6 Aerosols are composed of solid and liquid particles of varying chemical complexity, size, and phase. New particles are
7 added into the atmosphere by direct emissions and nucleation (secondary particles). Primary particles originate from
8 anthropogenic sources such as fossil fuel combustion and natural sources (fires, desert dust, sea salt, etc). Secondary
9 particles are formed through nucleation and condensation of the gas phase or by in-cloud processes (Ervens et al.,
10 2011). Organic aerosols are a key issue for models as their formation processes and evolution are poorly known.
11 Depending on meteorological conditions and aerosol properties, aerosol particles act as cloud condensation nuclei with
12 the potential to impact the precipitation pattern (Poschl et al., 2005; Duseck et al., 2006; Rosenfeld et al., 2008). The
13 coupling between aerosols and cloud remains an important but poorly understood issue. Unlike well-mixed greenhouse
14 gases, short-lived aerosols exhibit a strong regionality in climate forcing and air quality impacts (Monks et al., 2009).
15 Chemistry-transport models (CTMs) are essential to capture the regional forcing and impacts of aerosols.

16 For the last two decades, numerical CTMs have experienced significant improvements thanks to: the increase in high
17 performance computing resources (Colette et al., 2014), the “online” coupling between meteorological and chemical
18 fields (Zhang et al., 2008; Zhang et al., 2013; Kukkonen et al., 2012; Baklanov et al., 2014) and the improved
19 knowledge of atmospheric processing. While the progress in simulating air quality are notable, many challenges remain
20 for CTMs. the AQMEII (Air Quality Model Evaluation International Initiative) is a joint effort between North America
21 and Europe to establish common methodologies for model evaluation with a focus on ozone and aerosols (Rao et al.,
22 2011). Vautard et al. (2007) concluded that the majority of CTMs used for AQMEII captured the observed gas phase
23 mean values and daily variability fairly well, except for city centres. The skill of aerosol simulations, however, is
24 generally lower. Nopmongcol et al. (2012) highlighted the role of emissions and dilution in the performance of their air
25 quality model. In their comprehensive evaluation of the on-line coupled CTM COSMO-ART, Knote et al. (2011) noted
26 that ozone and NO_x were well reproduced; PM_{2.5} and PM₁₀ were, on average, underestimated. Several processes needed
27 to be improved in the model, such as wet scavenging, SOA formation, distribution and concentrations in primary
28 emissions of aerosol particles. Several authors have also mentioned lateral boundary conditions for aerosols as a source
29 of uncertainties. Aksoyoglu et al. (2011) concluded that the offline-coupled CAMx (Comprehensive Air quality model
30 with extensions) model reproduced the relative composition of aerosols very well over Switzerland but underestimated
31 the absolute concentration by 20%. Tuccella et al. (2012) validated the online-coupled WRF/CHEM (Weather Research
32 and Forecast – Chemistry) model against ground-based measurements over Europe. The model reproduced daily PM_{2.5}
33 aerosol mass with a slight negative bias but underpredicted particulate sulphate by a factor of 2 and overpredicted
34 ammonium and nitrate by about a factor of 2. Missing processes in the aqueous-phase could explain the differences
35 (Ervens et al., 2011). Zhang et al. (2013) compared the offline-coupled WRF/Polyphemus with the online-coupled
36 model WRF/CHEM-MADRID over Western Europe. No model was shown to be superior in terms of aerosol
37 representation. Although the online WRF/CHEM-MADRID accounted for interactions between the meteorology and
38 the chemistry, the model comparison showed that the simulation of atmospheric pollutant was mainly sensitive to the
39 vertical structure, emissions and parameterizations for dry/wet depositions. Online biogenic emissions significantly
40 improved the simulated temporal variations and magnitudes for most variables and for both models. Meteorological
41 conditions also contribute to aerosol composition. Tulet et al. (2005) used the online-coupled Meso-NH (Mesoscale
42 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 CAPITOU ("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_{2.5}
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).
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[•] developed by Crassier

1 et al., (2000). It includes 82 prognostic gaseous chemical species and 363 reactions enabling the formation of SOA
2 precursors to be addressed (Tulet et al., 2006), compared with 189 prognostic species and 361 reactions in CACM.
3 The gas phase chemical mechanism used in WRF/CHEM in the present study is RACM (Regional Atmospheric
4 Chemistry Mechanism; Stockwell et al., 1997; Geinger et al., 2003). This mechanism includes 84 species and 252
5 reactions. It includes 16 aggregated anthropogenic species (alkanes, alkenes, toluene, xylene, cresol) and 3 aggregated
6 biogenic species (isoprene, monoterpenes and sesquiterpenes) representing VOCs.
7 Both ReLACS2 and RACM allow O₃ chemistry to be simulated for a range of conditions found in remote areas to
8 polluted urban sites, from the surface to the upper troposphere. In addition, ReLACS2 was designed to simulate the
9 formation of SOA precursors. For both models, the photolytic rates are calculated using the TUV (Tropospheric
10 Ultraviolet and Visible; <https://www2.acd.ucar.edu/modeling/tropospheric-ultraviolet-and-visible-tuv-radiation-model>)
11 radiation model. The version 5.0 (Madronich and Flocke; 1999) is used for Meso-NH and the version 4.1 (Madronich et
12 al., 1998; McKenzie et al., 2007) for WRF/CHEM.

13 **2.2 Aerosol scheme**

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

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

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

34 To simulate SOA formation in WRF/CHEM, a new parameterization is used: the VBS set (Volatility Basis Set; Donahue
35 et al., 2006), recently coupled by Ahmadov et al. (2012) to the aerosol module MADE and to the gas phase chemical
36 mechanism RACM in the WRF/CHEM model. In this parameterization, the oxidation of the VOCs by the hydroxyl
37 radicals OH, O₃ or nitrate radicals leads to the production of organic species, which partition between the gas and the
38 particle phases according to their volatility. The volatility spectrum is divided into four bins with saturation vapour
39 concentrations ranging from 1 to 1000 $\mu\text{g m}^{-3}$. For each bin and each SOA precursor, an SOA yield based on smog
40 chamber studies is defined. Yields are different for two regimes, high and low NO_x conditions. Moreover, the
41 photochemical aging of the first generation VOC oxidation products is taken into account by shifting mass from high
42 volatility bins to lower ones when further oxidation processes occur.

1 **2.3 Dry deposition**

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

13 In WRF/CHEM, the dry deposition of gas phase species is parameterized according to Erisman et al. (1994) also based
14 on Wesely's (1989) scheme. The deposition velocity is proportional to the sum of the aerodynamic resistance between a
15 specified height and the surface, the sub-layer resistance (dependant upon the molecular diffusion) and the surface
16 resistance. The impact of the nature, the town and the sea surfaces on the exchanges between the soil, the surface and
17 the atmosphere is taken into account by the unified Noah Land Surface Model (Chen and Dudhia, 2001), which
18 considers 24 land use categories (including urban and built-up lands, water bodies, snow and ice) and 16 soil categories
19 according to the United States Geological Survey (USGS). However unlike in Meso-NH, the mesophyll resistances for
20 all gases are assumed to be zero, thus increasing the dry deposition velocity. For organic species, the dry deposition
21 velocity is parameterized as proportional to the dry deposition velocity of nitric acid. Since no data are available to
22 constrain the model, the parameter that determines the fraction of nitric acid is set to 0.25 as defined by Ahmadov et al.
23 (2012). For aerosol particles, the Wesely approach is used (Wesely and Hicks, 2000).

24 In both models, no wet deposition scheme is activated.

25

26 **3 Observation databases and simulation set-up**

27 **3.1 Databases**

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

33 The MOZAIC (Measurements of OZone, water vapour, carbon monoxide and nitrogen oxides by In-service Airbus
34 airCraft) database provides measurements of meteorological and chemical fields made during a large number of
35 commercial flights all over the world since 1993 (<http://www.iagos.fr/web/>; Marengo et al., 1998). These 20 years of
36 measurements enable climatology and air quality studies to be made for the troposphere. The database includes data
37 collected from the ground to the upper troposphere during take-offs and landings, with high temporal and 3D-spatial
38 resolution. These vertical profiles are used here to evaluate meteorology and chemistry in the vertical direction. During
39 landings and take-offs, data is collected every 4s, which corresponds approximately to 50-100 m in the vertical direction
40 (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 O₃, SO₂ (sulphur dioxide), NO_x (nitrogen dioxide) and PM_{2.5} 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-PM₁) for sulphate (SO₄²⁻), ammonium
14 (NH₄⁺), nitrate (NO₃⁻) 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_{2.5} and PM₁₀ 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

1 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
2 concerned spin-up. The time step was 50 s.

3 The initial and boundary conditions for meteorology were provided by the ECMWF (European Centre for Medium-
4 Range Weather Forecasts) analysis with a horizontal resolution of 0.5° and were forced every 6 h. The initial and
5 boundary conditions for gaseous chemical species including inorganic nitrogen species, CO, SO₂, NH₃, NMVOCs
6 (Non-methane VOCs), primary (BC, POA) and secondary (inorganics, SOA) aerosol species were taken from
7 MOZART-4 (Model for Ozone and Related chemical Tracers, version 4) (Emmons et al., 2010) driven by meteorology
8 from NCEP centres (National Centers for Environment Prediction). Boundary chemical fields were forced every 6h.

9 For Meso-NH, surface emissions of atmospheric compounds were taken from the ECCAD database (Emissions of
10 atmospheric Compounds & Compilation of Ancillary Data, [http://eccad.sedoo.fr/eccad_extract_interface/JSF/
11 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
12 dataset (MACC/CityZEN EU projects) (Van der Werf et al., 2006; Lamarque et al., 2010; Granier et al. 2011; Diehl et
13 al. 2012) provided “off-line” monthly anthropogenic emissions for CO, NO_x, SO₂, NH₃, NMVOCs such as alkanes,
14 alkenes, alcohols, aldehydes, ketones and aromatics, lumped into 21 species, and for primary aerosol species, with a
15 grid resolution of 0.5°. The MEGANv2 model (Model of Emissions of Gases and Aerosols from Nature, version 2)
16 (Guenther et al., 2006) gave “off-line” monthly net emission of gases and aerosols from vegetation into the atmosphere
17 at 0.5° resolution (for NO_x and VOCs). The monthly GFED3 (Global Fire Emissions Database, version 3; Van der Werf
18 et al., 2010) database was used to represent biomass burning emissions (CO, NMVOCs, BC and POA) with a spatial
19 resolution of 0.5°.

20 Emissions in WRF/CHEM also came from MACCity for anthropogenic species. Biogenic emissions were different: the
21 MEGAN model was used in “on-line” configuration because this option was already available in WRF/CHEM. In
22 MEGAN, emissions were estimated using a 1km x 1 km resolution map of vegetation defined for the year 2003. For
23 biomass burning emissions, the daily FINN inventory (Fire INventory from NCAR) (Wiedinmyer et al., 2011) model
24 was used in WRF/CHEM with a 1 km² spatial resolution.

25 For Meso-NH and WRF/CHEM, NO_x emissions were assumed to be 70% NO and 30% NO₂. In the chemical
26 mechanisms CACM and RACM, in order to limit the number of model species, some similar organic compounds were
27 grouped together based on the principle of reactivity weighting. Aggregation factors, computed by Middleton et al.
28 (1990), were used for VOCs. For aerosol species, in WRF/CHEM, a conversion factor of 1.6 proposed by Bessagnet et
29 al. (2008) was used to convert the emissions of organic carbon (OC) into particulate organic matter (POA). Then, 20%
30 of BC and POA emissions were allocated to the Aitken mode of the aerosol distribution and 80% to the accumulation
31 mode according to Ackermann et al. (1998). In Meso-NH, 5% of OC and BC were allocated to the Aitken mode and
32 95% to the accumulation mode, as, according to the actual knowledge, organic aerosol mass is assumed to be
33 distributed mainly in the accumulation mode (Kanakidou et al., 2005). For both models, parameters for the three
34 lognormal modes (total particle number concentrations, mean radius and standard deviation) are initialized from Asmi
35 et al. (2011).

36

37 **4 Evaluation of models at regional scale**

38 **4.1 Three contrasted meteorological situations over Europe**

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

1 wind speeds exceeding 20 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°N ; 10°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) (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°N and 20°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.

1 **4.3 Vertical distribution of meteorological variables**

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

7 Fig. 3 shows that both models capture the vertical distribution of T and Td between the surface and 300 hPa on 18
8 September 2008. Below 850 hPa, both models tend to overestimate T and Td. Models simulate stratiform clouds
9 between 500 hPa and 300 hPa as observed during flights where T was equal to Td. In the planetary boundary layer for
10 18 September 2008, there is a positive bias of T but the observed temperature inversion is well reproduced by the
11 models early in the morning and in the evening. Measurements show a sudden increase in T at 850 hPa, which is less
12 visible for both models. Observed Td decreases strongly at 850 hPa early in the morning, meaning a dry air mass limit.
13 On 3 March 2009 (Fig. S3), the temperature vertical profile is well reproduced except at the surface where there is an
14 underestimation at 10:00 UTC. Larger discrepancies are obtained with both models for Td. Fig. 1 shows that Frankfurt
15 was at the edge of a rain band on 3 March 2009, which could explain the difference between local measurements and
16 low-resolution model outputs. The models reproduce the northeasterly-easterly flow at the surface on 18 September
17 2008 and the southwesterly flow on 3 March 2009.

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

21 **4.4 Gaseous pollutants at the surface**

22 Simulated results for O₃, NO_x and SO₂ are compared with the AIRBASE surface data. The number of rural and
23 background stations for statistics varied between 111 and 259. As emissions have no diurnal variation, the bias of O₃,
24 NO_x and SO₂ was calculated for daytime only, between 08:00 and 16:00 UTC (Fig. 4, Fig. S4 and Fig. S5 for 18
25 September 2008, 3 March 2009 and 26 June 2010 respectively, and Table 2). Fig. 5 shows the diurnal cycle of the
26 median bias for these three chemical species.

27 Meso-NH tended to overestimate O₃ for the three days (Fig. 4, Fig. 5, Fig. S4 and Fig. S5) with a highest NMB (+11%)
28 on 18 September 2008 (Table 2). Bias of NO_x was slightly negative (-3% <NMB< -1%, Table 2). Looking at the diurnal
29 cycle of the bias for NO_x, Meso-NH showed poorer performance during night-time, with mean differences up to 6 µg.m⁻³
30 for 50% of stations (Fig. 5). During daytime, the largest underestimations for NO_x were found in industrial areas of the
31 Po Valley (Italy), the Ruhr, the Netherlands and Austria, especially on 18 September 2008 (Fig. 4). In the Po Valley, for
32 the three study cases (Fig. 4, Fig. S4 and Fig. S5) a positive bias of up to 20 µg.m⁻³ of O₃ was associated with a strong
33 negative bias of NO_x (-5 µg.m⁻³ for 50% of stations).

34 WRF/CHEM underestimated O₃ on 3 March 2009 and 26 June 2010 (-14% <NMB< -3%, Table 2) but gave good
35 predictions of ozone during the third episode (NMB =1%, Table 2). Daytime NO_x concentrations were reproduced to
36 within -1 to 2% (Table 2), the worst prediction being made during night-time, with a systematically strong
37 overestimation of up to 10 µg.m⁻³ for 50% of stations (Fig. 5). Fig. 4, Fig. S4 and Fig S5 show patterns of maximum
38 biases for ozone and NO_x over the Po Valley and northern Europe for the WRF/CHEM model that are similar to those
39 of Meso-NH.

1 Daytime values of NMB ranged between -2% and 0% for SO₂ in both models (Table 2). As shown in Fig. 5,
2 WRF/CHEM tends to underestimate SO₂ for the three cases, with maximum bias during daytime (up to -1.5 µg.m⁻³ for
3 50% of stations).
4 Except in the Po Valley, during daytime, the level of accuracy of both models for the prediction of ozone, NO_x and SO₂
5 at the surface is acceptable for regulatory application (AQD, 2008). The maximum bias for both models over the Po
6 Valley is suspected to be due to the absence of diurnal variation of anthropogenic emissions (underestimation for NO_x
7 and overestimation for ozone). In the same way, both models showed a positive bias for NO_x during night-time due to
8 this absence of diurnal variation. This effect was also visible on the diurnal cycle of SO₂ bias for both models. However,
9 for ozone bias, the link with the diurnal cycle of anthropogenic emissions is indirect due to the non-linearity of ozone
10 chemistry. For instance, Tao et al. (2004) changed the temporal emission profiles (uniform vs. time-varying) of
11 anthropogenic species and found no impact on daytime ozone concentrations. In the same idea, Menut et al. (2012)
12 increased the NO₂ concentrations by 10-20% in their model using new hourly fluxes representative of traffic emissions
13 with a moderate impact on ozone (0-7% decrease) and the most important changes occurred during night-time. Among
14 potential sources of errors in the models, the chemical lateral boundary conditions are essential to capture the variability
15 of pollution inflow and provide background concentrations, to which local production is added (Tang et al., 2007,
16 Pfister et al., 2011). Global models provide chemical top and lateral boundary conditions for regional models but
17 introduce uncertainties that are inherent in the model or due to the coupling process. This point is discussed in the next
18 part. Obviously, differences between the two models could be related to differences in chemical reaction schemes
19 coupling to differences between thermodynamical modules for gas to particles conversion, dynamics of the planetary
20 boundary layer (venting of pollutants) and dry deposition parameterizations.

21 **4.5 Vertical distribution of ozone and carbon monoxide**

22 The simulated vertical distributions of O₃ and CO were assessed using measurements from the MOZAIC database (Fig.
23 6 for 18 September 2008 and Fig. S6 for 3 March 2009; no data for 26 June 2010).

24 CO is a significant trace gas in the troposphere, which strongly influences the concentrations of oxidants such as
25 hydroxyl radical (OH.) and O₃. CO is mainly emitted by anthropogenic activities and has a lifetime of a few months in
26 the boundary layer (BL) and the free troposphere (FT) (Seinfeld and Pandis, 1998).

27 Above 9-10 km altitude, O₃ and CO mixing ratios and their daily variability are controlled by stratosphere-troposphere
28 exchanges, venting by convective clouds, chemical reactions and long-range transport. The vertical profiles in Fig. 6
29 show that both models fail to reproduce a well-marked stratospheric dry ozone-enriched intrusion. Both models
30 underestimate O₃ with negative biases of up to 35 ppbv on 18 September 2008 (Fig. 6 at 0700 UTC) and 200 ppbv on 3
31 March 2009 (Fig. S6). Stratospheric air corresponds to poor CO mixing ratios. CO is overestimated at these altitudes
32 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
33 at 0400 UTC).

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

- 36 – The influence of local sources near the airport, the intensity and temporal variations of which are not captured
37 by the models,
- 38 – The BL dynamics and the associated vertical venting of the pollutants. Interestingly, the worse biases are
39 obtained during night-time, when the surface BL is expected to be low, favouring the accumulation of
40 pollutants. The models performed better during daytime (1000 UTC on Fig. 6 and Fig. S6) as the depth of the
41 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 O₃ 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 O₃ 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 O₃
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 O₃ 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 O₃ 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 O₃ 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_{2.5} bulk aerosol at the surface**

33 Simulated daily mean PM_{2.5} 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_{2.5} 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 (NO₃⁻, SO₄²⁻ and NH₄⁺) and organic (SOA) aerosol components.
37 The corresponding daily NMB (in %) was computed (Table 2) but it should be borne in mind that PM_{2.5} 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_{2.5} mass concentration with positive NMB between 6
40 and 14% for the three dates. WRF/CHEM slightly underestimates the PM_{2.5} 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 NH_3 , 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, NH_3 , NO_x and 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-Pusztas 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 NO_x , the comparison was done with NO_2 as no NO observations are available but, as NO_2
9 dominates NO_x mixing ratio, mixing ratio of NO_2 can be seen as a lower limit for NO_x . Except for WRF/CHEM for the
10 3 March 2009, simulated NO_x mixing ratio for both models are of the same order of magnitude than observed 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 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 ± 0.03 ppbv; for 26 June 2010, it was 0.12 ± 0.04 ppbv and for 18 September 2008, it was 0.17 ± 0.06 ppbv. In
23 comparison to these values, Meso-NH overestimated observed SO_2 mixing ratio whereas WRF/CHEM slightly
24 underestimated it. This trend is similar to K-Pusztas.

25 The NH_3 mixing ratio typically ranges between 0.1 and 10 ppb over continents (Seinfeld and Pandis, 1998). 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 NH_3 are dry and wet deposition
28 and its transfer in the particulate phase as NH_4^+ . 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 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 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 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-Pusztas (24 ppbv) stations on 18 September 2008 and for Melpitz (30 ppbv) on 26 June 2010 (Fig. 8).
38 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 NO_x and 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 NO_x and SO_2 cannot be explained by
42 difference in venting of pollutants but more probably by differences between chemical mechanisms and

1 thermodynamical modules for gas to particles conversion. Indeed, discrepancies between NO_x and SO_2 are highest
2 when boundary layer height is small (during night-time at plain sites and all along the day at K-Pusztta for the 3 March
3 2009), so, when chemical species are more concentrated and chemical reactivity is higher.

4 **5.2 Chemical composition of aerosol particles**

5 The model performance in term of aerosol composition is evaluated with the AMS global database. This database
6 provides the mass concentration of NR- PM_1 for NO_3^- , SO_4^{2+} , NH_4^+ and OM, mentioned as ORG on Figs. 9 and 10. OM
7 includes POM and SOM (for Primary and Secondary OM respectively). Here, OM measured by AMS is compared to
8 the sum of anthropogenic primary organic aerosol (APOA) and secondary organic aerosol (SOA) simulated by Meso-
9 NH and WRF/CHEM. Simulated and observed mass concentrations are compared as hourly mean to highlight how
10 models are able to represent the detailed processes driving the aerosol composition and its temporal evolution. For each
11 station, this temporal evolution of the aerosol mass composition on 18 September 2008 and on 3 March 2009 is
12 presented in Fig. 9 and Fig. 10 respectively (cf. supplementary materials for 26 June 2010, Fig. S7). The daily mean
13 mass concentration for each aerosol component is also indicated as well as their daily mean mass fraction.

14 In order to assess the ability of the models to reproduce aerosol chemical composition, statistical parameters such as
15 Root Mean Square Error (RMSE), mean fractional bias (MFB) and mean fractional error (MFE), as discussed by
16 Boylan and Russell (2006), are computed for each chemical component of aerosols and for the three case study cases
17 (Tables 3, 4 and 5). When the performance criterion ($|\text{MFB}| < 60\%$ and $\text{MFE} < 75\%$) is satisfied, the level of accuracy is
18 considered to be acceptable for standard modelling applications. When the performance goals ($|\text{MFB}| < 30\%$ and MFE
19 $< 50\%$) are met, the level of accuracy is considered to be close to the best a model can be expected to achieve.

20 Models provide aerosol mass compositions for Aitken and accumulation modes for an aerodynamic diameter less than
21 $2.5 \mu\text{m}$ or $\text{PM}_{2.5}$. As AMS-observed NR- PM_1 mass concentration is compared to the simulated $\text{PM}_{2.5}$ in the following,
22 the models are expected to give an overestimation of aerosol mass concentration. The difference between PM_1 and
23 $\text{PM}_{2.5}$ mass concentrations can be high (previous studies report $\text{PM}_1/\text{PM}_{2.5}$ mass ratios between 55 and 75%) and can
24 vary according to the meteorological conditions and the emission rates of chemical compounds (Aksoyoglu et al, 2011;
25 Spindler et al., 2013). However, the proportions of the predominant chemical components in NR- PM_1 and simulated
26 $\text{PM}_{2.5}$ are expected to be of the same order.

27 **5.2.1 At puy de Dôme**

28 Observations (Fig. 9 and Fig. S7) suggest that, during the summer cases (26 June 2010 and 18 September 2008), the
29 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-
30 NH shows consistent results, with the highest contribution of simulated OM on 26 June 2010 ($11.56 \mu\text{g.m}^{-3}$ (66.49%))
31 and a significant contribution on 18 September 2008 ($5.13 \mu\text{g.m}^{-3}$ (43.77%)). SOA mass makes the highest contribution
32 to OM for the three study cases. WRF/CHEM simulates the highest masses of OM during the two summer cases (0.69
33 $\mu\text{g.m}^{-3}$ 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.
34 At the pdD station, the mass of OM simulated by WRF/CHEM is weaker by a factor of 10 than those simulated by
35 Meso-NH.

36 On 3 March 2009 (Fig. 10), both models and measurements show a minimum of the mass concentration of NH_4^+ (1.36
37 $\mu\text{g.m}^{-3}$, $0.93 \mu\text{g.m}^{-3}$ and $1.28 \mu\text{g.m}^{-3}$ for Meso-NH, WRF/CHEM and observations, respectively). This day corresponds
38 to the presence of an oceanic air mass (Fig. 1). Both models simulate a minimum of the mass concentration of NO_3^- (0.6
39 $\mu\text{g.m}^{-3}$ and $0.57 \mu\text{g.m}^{-3}$ respectively for Meso-NH and WRF/CHEM) for 18 September 2008, during the presence of a
40 Mediterranean air mass. The observed mass concentration of NO_3^- is the weakest for this situation ($0.93 \mu\text{g.m}^{-3}$) but still
41 about twice that simulated by the models. Whatever the case, WRF/CHEM systematically underestimates the mass

1 concentration of NH_4^+ and NO_3^- at pdD by a factor of 2-4. Meso-NH underestimates the mass concentration of NH_4^+
2 and NO_3^- on 18 September 2008 and 26 June 2010 (Fig. 9 and Fig. S7). Meso-NH overestimates slightly NH_4^+ and NO_3^-
3 on 3 March 2009.
4 Concerning 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 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 SO_4^{2-} is 2-3 higher than the simulated ones.
9 The performance criterion, computed for Meso-NH, is satisfied for NH_4^+ on 18 September 2008 (Table 4), for NH_4^+ and
10 OM on 26 June 2010 (Table 5), and for NH_4^+ on 3 March 2009 (Table 3). Concerning WRF/CHEM, the performance
11 goal is only attained for 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 NH_4^+ ($6.60 \mu\text{g.m}^{-3}$ (49.02%)) and 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 NO_3^- ($10.94 \mu\text{g.m}^{-3}$ (53.14%)). Like the observations, WRF/CHEM shows a strong
26 contribution of SO_4^{2-} ($4.72 \mu\text{g.m}^{-3}$ (46.27%)) and 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 NH_4^+ (44%) dominates the inorganic fraction in observations, 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 SO_4^{2-} and OM (Table 3). It is also met by Meso-
31 NH for NH_4^+ on 3 March 2009 (Table 3). On 18 September 2008, the performance criterion is only fulfilled by
32 WRF/CHEM for NO_3^- and Meso-NH for OM. (Table 4). For this date, the two models overestimate the observed SO_4^{2-}
33 (Fig. 9).

34 **5.2.3 At K-Puszt**

35 Observations at the K-Puszt 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 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 SO_4^{2-} while
4 measurements show a dominant contribution of NO_3^- ($2.36 \mu\text{g}\cdot\text{m}^{-3}$). Moreover, the observed mass concentration of NO_3^-
5 found at K-Pusztas is higher than at the other two sites on the same date. Both models underestimate 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 NH_4^+ and NO_3^- at this station. Concerning
8 WRF/CHEM, the performance goal is reached for 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, NH_3 and 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-Pusztza), both models underestimate NH_4^+ mass concentrations. Simulated
28 NO_3^- and 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-Pusztza, where the NH_4^+ mass concentration simulated by WRF/CHEM is slightly higher than that
30 simulated by Meso-NH. This general behaviour of NO_3^- and NH_4^+ mass concentrations is linked to a systematic
31 underestimation of simulated 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 NO_x by Meso-NH may come from a more efficient transfer of oxidation
33 products of NO_x as nitric acid from the gas to the aerosol phase. Both models overestimate the SO_4^{2-} mass
34 concentration, except for Meso-NH at Melpitz on 3 March 2009 - linked to its high simulated NO_3^- mass concentration.
35 SO_2 simulated by WRF/CHEM is underestimated in comparison with observations (cf. Sect. 4.4 and Sect. 5.1). In both
36 models, SO_4^{2-} is formed by oxidation of SO_2 in the gas phase. Therefore, the underestimation of SO_2 in the gas phase by
37 WRF/CHEM, which is linked to its overestimation of aerosol SO_4^{2-} , seems to indicate a faster oxidation in WRF/CHEM
38 than in Meso-NH. On 18 September 2008 at K-Pusztza and on 3 March 2009 at Melpitz, the 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 NH_4^+ mass concentration for these two cases is opposite: Meso-NH simulates more
41 NH_4^+ than WRF/CHEM at Melpitz on 3 March 2009 and less at K-Pusztza on 18 September 2008. This behaviour can be
42 explained by the very high NO_3^- mass concentration simulated by Meso-NH at Melpitz on 3 March 2009, leading to

1 additional transfer of NH_3 to the particulate phase. After ammonium sulphate was formed, the formation of NH_4NO_3
2 and subsequently of NH_4^+ and 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.
4

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 (O_3 , NO_x , 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 NO_x and SO_2 . Globally, 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, 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 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 SO_4^{2-}
31 simulated by both models is often overestimated, a fact that is certainly associated with biases of SO_2 mixing ratio.
32 Simulated NO_3^- and NH_4^+ mass concentrations are almost always higher for Meso-NH than for WRF/CHEM, in
33 connection with differences in 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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1 **Figure captions**

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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 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 ($^{\circ}$ /N) and wind speed (m.s^{-1}) at 10 m, and temperature ($^{\circ}$ C) and dewpoint ($^{\circ}$ 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 (staves 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. O_3 , NO_x and 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 O_3 , NO_x and 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 25th and 75th 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, O_3 gold), WRF-Chem (CO black, O_3 red). CO and 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 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-Pusztta stations. Precursors are represented with different colours: NH₃ (grey), SO₂ (red), NO_x
4 (blue) (left Y axis) and VOCs (green) (right Y axis).

5

6 Fig. 9: Diurnal cycle of aerosol chemical composition in mass concentration ($\mu\text{g}\cdot\text{m}^{-3}$) of NR-PM₁ 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-Pusztta 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. NR-PM₁ include NH₄⁺ (orange), SO₄²⁻ (red), NO₃⁻ (blue) and ORG (green). Meso-NH
11 and WRF/CHEM differentiate APOA (dark green) and SOA (light green) in OM.

12

13 Fig. 10: Diurnal cycle of aerosol chemical composition in mass concentration ($\mu\text{g}\cdot\text{m}^{-3}$) of NR-PM₁ 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-Dome, Melpitz and K-Pusztta 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. NR-PM₁ include NO₃⁻ (blue), SO₄²⁻ (red), NH₄⁺ (orange) and ORG (green). Meso-NH
18 and WRF/CHEM differentiate APOA (dark green) and SOA (light green) in OM.

19

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 nd 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 th 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 th order Vertical advection for momentum and scalar is 3 rd 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_{2.5} 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_{2.5} mass concentration, the number of stations used is presented, respectively on Fig. 5 and Fig. 7.

| | 03/03/09 | | 26/06/10 | | 18/09/08 | |
|-------------------|----------------------------|----------|-----------------|----------|-----------------|----------|
| | 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 ₃ | 7 | -3 | 4 | -14 | 11 | 1 |
| NO _x | -3 | 2 | -1 | -1 | -3 | -2 |
| SO ₂ | 0 | -1 | -1 | -1 | -1 | -2 |
| | Bulk mass (daily) | | | | | |
| PM _{2.5} | 12 | 0 | 14 | -1 | 12 | -3 |

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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 |
| $\mu\text{g.m}^{-3}$ | RMSE | | MFB (%) | | MFE (%) | | RMSE | | MFB (%) | | MFE (%) | |
| NH_4^+ | 0.51 | 0.59 | 3 | -31 | 37 | 49 | 3.42 | 4.87 | -44 | -90 | 46 | 90 |
| SO_4^{2-} | 1.19 | 1.48 | 101 | 112 | 101 | 112 | 1.06 | 1.92 | -21 | 49 | 30 | 49 |
| NO_3^- | 1.37 | 1.16 | 20 | -74 | 72 | 84 | 9.00 | 0.39 | 138 | 1 | 138 | 16 |
| ORG | 0.93 | 0.91 | 33 | -118 | 58 | 118 | 1.70 | 0.83 | 60 | -49 | 60 | 49 |

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