

1 Eurodelta-Trends, a multi-model experiment of air quality hindcast 2 in Europe over 1990-2010.

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32 **Abstract**

33 The Eurodelta-Trends multi-model chemistry-transport experiment has been designed to facilitate a
34 better understanding of the evolution of air pollution and its drivers for the period 1990-2010 in
35 Europe. The main objective of the experiment is to assess the efficiency of air pollutant emissions
36 mitigation measures in improving regional scale air quality.

37 The present paper formulates the main scientific questions and policy issues being addressed by the
38 Eurodelta-Trends modelling experiment with an emphasis on how the design and technical features
39 of the modelling experiment answer these questions.

40 The experiment is designed in three tiers with increasing degree of computational demand in order
41 to facilitate the participation of as many modelling teams as possible. The basic experiment consists
42 of simulations for the years 1990, 2000 and 2010. Sensitivity analysis for the same three years using
43 various combinations of (i) anthropogenic emissions, (ii) chemical boundary conditions and (iii)

1 meteorology complements it. The most demanding tier consists two complete time series from 1990
2 to 2010, simulated using either time varying emissions for corresponding years or constant
3 emissions.

4 Eight chemistry-transport models have contributed with calculation results to at least one
5 experiment tier, and three models have – to date - completed the full set of simulations (and 21-year
6 trend calculations have been performed by four models). The modelling results are publicly available
7 for further use by the scientific community.

8 The main expected outcomes are (i) an evaluation of the models performances for the three
9 reference years, (ii) an evaluation of the skill of the models in capturing observed air pollution trends
10 for the 1990-2010 time period, (iii) attribution analyses of the respective role of driving factors
11 (emissions/boundary conditions/meteorology), (iv) a dataset based on a multi-model approach, to
12 provide more robust model results for use in impact studies related to human health, ecosystem and
13 radiative forcing.

14

1 **1 Introduction**

2

3 Air pollution is a crucial environmental concern because of its detrimental impacts on health,
4 ecosystems, the built environment and short term climate forcing. Whereas it was originally
5 regarded as an urban issue, in the late 1970s the large scale acidification of precipitation made it
6 clear that at least part of the problem could only be solved through international cooperation (OECD,
7 1977). This was the background for the establishment of the Convention on Long Range
8 Transboundary Air Pollution (CLRTAP) in 1979. The main vehicles of the LRTAP Convention are the
9 Protocols that aim to reduce the emission of various compounds (sulphur in 1985, nitrogen oxides in
10 1988, volatile organic compounds in 1991, heavy metals and persistent organic pollutants in 1998,
11 and the multi-pollutant multi-effect Gothenburg Protocol to abate acidification, eutrophication and
12 ground level ozone in 1999 and subsequent revision in 2012). The design of such mitigation
13 strategies was largely supported by the development of models (chemistry-transport and integrated
14 assessment tools) and monitoring networks.

15 After several decades of international cooperation, it is timely to take stock of the evidence available
16 to assess the efficiency of the LRTAP Convention and the corresponding emission ceilings protocols.
17 The Executive Body of the Convention has therefore requested an assessment of the evolution of air
18 pollution and subsequent effects from its two scientific and technical bodies (i) the European
19 Monitoring and Evaluation Programme (EMEP) and (ii) the Working Group on Effects (WGE). As a
20 result, the Task Force on Measurement and Modelling (TFMM) of EMEP published an assessment of
21 air pollution trends (Colette et al., 2016), whereas the WGE published an assessment of
22 corresponding effects on health and ecosystems (De Wit et al., 2015), and an overall assessment
23 report encompassing all the activities undertaken under the Convention was also released (Maas and
24 Grennfelt 2016).

25 The effects of emissions on the concentrations is rather complex due to (i) the non-linearity of
26 atmospheric chemistry, (ii) the presence of inflow of air pollution due to the intercontinental
27 transport of air pollutants, and (iii) the meteorological variability. This is where Chemistry-Transport
28 Models (CTMs) come into play with the multi-model air quality trend experiment introduced in the
29 present paper.

30 The LRTAP convention relies in part on the results of the EMEP/MS-CW chemistry-transport model
31 (Simpson et al., 2012a). Since the beginning of the 2000s, the Joint Research Centre of the European
32 Commission initiated a number of multi-model assessments to provide a benchmark for the
33 EMEP/MS-CW model through its comparison with the modelling tools being used by the States-
34 Parties to the Convention as part of the Eurodelta project (Bessagnet et al., 2016;van Loon et al.,
35 2007;Thunis et al., 2008). The Eurodelta-Trends (EDT) exercise builds upon this tradition, focusing on
36 the specific context of air quality trends modelling. Its main goal is to assess to what extent observed
37 air pollution trends could be related to emission mitigation, although this overarching question can
38 only be addressed after having assessed the confidence we can have in the models, in particular in
39 their capacity to reproduce the trends.

40 Over the recent past, several multi-model projects covering a time period of one year or less were
41 undertaken such as the earlier phases of Eurodelta cited above but also the various phases of the
42 AQMEII project (Galmarini et al., 2012;Galmarini et al., 2017;Rao et al., 2011;Im et al., 2015).
43 However only a few attempts have been made to address the issue of the long-term evolution of
44 European-scale air quality by means of modelling studies. The first attempts were using only one
45 model as in (Vautard et al. (2006);Jonson et al. (2006);Wilson et al. (2012)). A first ensemble was

1 proposed through the European Project CityZen which relied on 6-models (Colette et al., 2011).
2 While these studies were limited to about 10-yr time periods, a 20-yr hindcast study was presented
3 in (Banzhaf et al., 2015), relying however again on a single model. It is therefore timely to engage in a
4 multi-model hindcast of air quality over two decades.

5 The purpose of the present paper is to define the science and policy questions that are addressed by
6 the EDT exercise, and introduce the experimental setup designed to answer these questions. The
7 models participating in the experiment will also be presented as well as the project database of
8 model results.

9 **2 Experimental design**

10

11 The main policy focus being addressed in EDT analysis is the assessment of the role of European air
12 pollutant emission reductions in improving air quality over the past two decades. Subsequent
13 questions include assessing (1) the role of changes in global air pollution as well as (2) the role of
14 inter-annual meteorological variability. Before addressing such issues, it will be essential to quantify
15 the CTMs' capability in (1) reproducing observed air pollutant concentrations (processes determining
16 air quality: chemistry, physics, transport processes, emissions, meteorology), and (2) capturing the
17 long term evolution of air quality.

18 The time period covered by the experiment is 1990-2010. The year 1990 has been chosen as the
19 beginning of the period because that year serves as reference for the Gothenburg protocol. The end
20 of the period is 2010 because of the availability of underlying forcing data (emissions, boundary
21 conditions and meteorology) required for model calculations at the time the work was initiated.

22 The EDT model experiment is divided into three tiers, targeting various science and policy questions.
23 The tiers also differ in terms of computational demand that allowed involving as many modelling
24 groups as possible. The tiers of experiments are summarized in Table 2. They differ in terms of the
25 number of modelled years to be addressed in the 1990-2010 period and in terms of forcing data used
26 in model calculations for the anthropogenic emissions, the chemical boundary conditions, and the
27 meteorological year. Most of the experiments consist of variations in one or two of these three
28 factors in order to disentangle the role of each forcing. The role of chemical boundary conditions
29 constitutes one notable exception since two sources of forcing are used: either a global CTM
30 simulation or an observation-based climatology (further details are provided on boundary conditions
31 in Section 7).

32 The first simulation of the EDT experiment is a reference for the year 2010 using the meteorology
33 (M), the chemical boundary conditions (B) and the emissions (E) for that year, named as M10B10E10,
34 with two digits corresponding to the last two digits of the year. They are complemented with
35 simulations for the years 1990 and 2000 (using corresponding meteorology, boundary conditions and
36 emissions: M90B90E90 and M00B00E00 respectively) to form tier 1A. Tier 1A will allow testing the
37 accuracy of all CTMs in simulating pollution changes for the near past (1990, 2000 and 2010), at a
38 lower computational cost than running the full 21-yr period.

39 Tier 1B is dedicated to the first two sensitivity experiments, for which the meteorology and the
40 boundary conditions are those of the year 2010, but the emissions correspond to 1990 and 2000
41 (M10B10E90 and M10B10E00). They will allow assessment of the individual impact of emission
42 changes alone (E10 versus E90 and E10 versus E00) by comparison with Tier 1A (specifically
43 M10B10E10).

1 In Tier 2A, two more sensitivity simulations are performed for the meteorological year 2010, using
2 emissions and boundary conditions of 1990 and 2000 (M10B90E90 and M10B00E00, respectively). By
3 comparison with Tier 1B, they will allow the assessment of the impact of global chemical background
4 changes on European air quality between the years 1990 and 2010, and also for the sub-periods
5 1990-2000 and 2000-2010 (B10 versus B90 and B10 versus B00).

6 Tier 2B is an alternate set of reference simulations for 1990, 2000 and 2010, in which boundary
7 conditions provided by a global model (C) instead of the observation-based boundaries (B) are used
8 (M90C90E90, M00C00E00, M10C10E10). It will allow assessment of the uncertainty related to the
9 large scale chemical forcing by comparison with Tier 1A.

10 Tier 2C is a complement to Tier 2A using the meteorology of 2000 and two combinations of 1990 and
11 2000 boundary conditions and emissions (M00B90E90, M00B00E90). These additional simulations
12 are required to perform the attribution analysis for the concentration changes between 1990 and
13 2000, whereas the simulations required for the attribution of driving factors between 1990 and 2010
14 and between 2000 and 2010 are dealt with in tiers 1A, 1B, and 2A.

15 Tier 3A consists in 21-year simulations covering 1990-2010, using meteorology, boundary conditions
16 and emissions for the respective years (MyyByyEyy, with yy being the 2-digits year between 1990 and
17 2010). It will be used to assess the capability of the models to capture observed trends in air quality
18 by means of comparisons with available measurements. Fewer modelling teams delivered results for
19 this higher tier of experiments, therefore model uncertainty will be put in perspective with the
20 spread of the whole ensemble in modelling Tier 1A (1990, 2000, 2010).

21 Tier 3B is the last sensitivity experiment in which 21-year simulations are performed using the 2010
22 emissions for the complete period (MyyByyE10). By comparison with Tier 3A, it will allow the
23 determination of the role of inter-annual meteorology and chemical boundary condition changes
24 versus the role of European emission changes.

25 Thus, the complete series of model runs included for each air quality model is 5 annual simulations
26 for Tier 1, 7 more simulations for Tier 2, and 39 (2x21 minus one overlap for 2010, and two annual
27 simulations belonging to Tier 1A: M90B90E90 and M00B00E00) more simulated years for Tier 3.

28 Figure 1 provides the schematics of the various combinations of simulations required to perform the
29 attribution analysis for any period of time between the three reference years (1990, 2000 and 2010).
30 The simulations labelled in black are covered by the above simulation plan. They are needed for the
31 assessment of the relative role of emission, meteorology and boundary condition changes.

32 The main limitations of the simulation plan are (i) that the three selected meteorological years may
33 be not representative, or atypical, for the full period, and (ii) the lack of interaction by considering 2^2
34 combinations instead of the 2^3 combinations required to cover the whole space of factors (Stein and
35 Alpert, 1993). In the forthcoming attribution study these limitations will be explored by (i) comparing
36 trend (tier 3A) and sensitivity (tier 1&2) tiers, and (ii) including additional simulations for the 2^3
37 possible combinations from one of the models (Chimere).

38 **3 Participating models**

39

40 Eight European modelling teams submitted their calculation results to the EDT database for at least
41 one tier of experiment (see the experiment design in Section 2) using state-of-art air quality models:
42 Chimere (Menut et al., 2013), CMAQ (Byun and Schere, 2006), EMEP/MSC-W (Simpson et al., 2012),
43 LOTOS-EUROS (Schaap et al., 2008; Manders et al., 2017), MATCH (Robertson et al., 1999) MiNNI

1 (Mircea et al., 2016), Polyphemus(Mallet et al., 2007), and WRF-Chem (Grell et al., 2005;Mar et al.,
2 2016). The main specifications of the eight participating models are summarized in Table 1 (note that
3 they can differ from the public release of the various models according to the elements provided in
4 the table).

5 The representation of physical and chemical processes differs in the models. The vertical distribution
6 of model layers (including altitude of the top layer and derivation of surface concentrations at 3m
7 height in the case of EMEP, LOTOS-EUROS and MATCH) is not prescribed either. However, as further
8 explained in the article, the other features of the model setup are largely constrained by the
9 experiment input data such as forcing meteorology, boundary conditions, emissions and by the
10 experiment characteristics such as horizontal domain and resolution. Only one of the participating
11 models included online coupled chemistry/meteorology (WRF-Chem), while all the other models are
12 offline CTMs.

13 **4 Modelling domain**

14

15 The modelling domain is displayed in Figure 2. The domain follows a regular latitude-longitude
16 projection (plate carrée projection) with increments of 0.25° and 0.4° in latitude and longitude,
17 respectively, which is about 25 km x 25 km at European latitudes. The total coverage extends from
18 17W to 39.8E and from 32N to 70N. All the participating models use the same modelling domain,
19 with only one exception: CMAQB uses a Lambert Conformal Conic projection map with 25 km
20 resolution and delivered their results on the common grid. The south-easternmost part of the
21 domain is not included in the CMAQB modelling domain.

22 **5 Meteorology**

23

24 The horizontal resolution of available global meteorological reanalyses over the 1990-2010 period is
25 considered too coarse to drive regional scale CTMs. Therefore, dynamically downscaled regional
26 climate model simulations using boundary condition from the ERA-Interim global reanalyses (Dee et
27 al., 2011) were used to force the CTMs involved in EDT. Most CTMs used the same meteorological
28 driver, with a couple of exceptions.

29 One of the meteorological drivers was produced using the Weather Research and Forecast Model
30 (WRF version 3.3.1) (Skamarock et al., 2008) at 0.44 degrees of resolution. In the framework of the
31 EuroCordex climate downscaling programme (Jacob et al., 2013) an evaluation of the regional
32 climate models downscaled with reanalysed boundary conditions (ERA-Interim reanalyses instead of
33 free climate runs) was reported by (Kotlarski et al., 2014). One of the WRF realisations in the
34 EuroCordex ensemble was subsequently further optimized as described in (Stegehuis et al., 2015), so
35 that we could identify an optimal WRF setup for our purpose (row #7 of Table S1 in their
36 supplementary material). The model was re-run using grid nudging towards the ERA-Interim
37 reanalyses (above the planetary boundary layer) in order to improve temporal correlations compared
38 to the regular free-running Cordex hindcast simulations. This WRF simulation was interpolated on the
39 25km resolution EDT grid and used to drive Chimere, EMEP, Polyphemus, and Minni. In EMEP model,
40 the interpolation of the meteorological fields from 0.4x0.4° to EDT grid was performed online. For
41 WRF-Chem, an online model that simulates meteorology and chemistry simultaneously (“online”),
42 the meteorology from the WRF-Eurocordex runs (Stegehuis et al 2015) was used as initial and lateral
43 boundary conditions and for applying four-dimensional data assimilation (FDDA), with coefficients as
44 described in Mar et al. 2016.The CMAQ model, which runs on a Lambert Conformal Conic projection,
45 could not use the meteorological data provided on the EuroCordex grid, so that WRF was re-run in a

1 Lambert Conformal projection at 25 km horizontal resolution using identical WRF setup and version
2 (3.3.1).

3 The CTMs LOTOS-EUROS and MATCH have been meteorologically forced by ERA-Interim series
4 further downscaled with respectively RACMO2 (van Meijgaard, 2012) and HIRLAM (Dahlgren et al.,
5 2016). RACMO2 used here was part of the EuroCordex studies documented in (Jacob et al., 2013) and
6 (Kotlarski et al., 2014) and excludes nudging towards Era-Interim. The HIRLAM EURO4M reanalysis
7 uses data assimilation in 3 dimensions for upper air and optimal interpolation for surface fields. An
8 initial analysis is conducted every 6 hours with subsequent forecasts saved on 3-hourly temporal
9 resolution. ERA-Interim is forced to the lateral boundaries. The HIRLAM reanalysis was interpolated
10 from the original 0.2 horizontal resolution on a rotated lat-lon grid (ca. 22km) to the EDT grid. The
11 main features of the mesoscale meteorological models are synthesized in Table 4.

12 **6 Emissions**

13

14 **6.1 Annual totals of anthropogenic emissions**

15

16 National annual emissions, distributed by SNAP (Selected Nomenclature for reporting of Air
17 Pollutants) sectors, were estimated with the GAINS (Greenhouse gases and Air pollution INteractions
18 and Synergies) model (Amann et al., 2011). The calculation was performed for 1990, 1995, 2000,
19 2005, and 2010 for SO₂, NO_x, NMVOC, CO, NH₃, and PM including PM₁₀, PM_{2.5}, BC, and OC. To derive
20 emissions for intermediate years, sectorial results for five-year periods were linearly interpolated.

21 The key activity data originates from Eurostat¹ and International Energy Agency (IEA, 2012) for
22 energy use and from Eurostat, UN Food and Agriculture Organization (FAO)², International Fertilizer
23 Association (IFA) for agriculture. For the transport sector, additionally the results of the COPERT
24 model for the EU-28 countries were used (Ntziachristos et al., 2009); this data includes detailed
25 transport sources, fuel distribution, mileage, and level of penetration of control measures. The
26 emission calculation considers impact of existing national and international source specific emission
27 limits and air quality legislation, e.g., several European Union Directives: Large Combustion Plants,
28 Industrial Emissions, National Emission Ceilings Solvent Directive, as well as the UNECE Gothenburg
29 Protocol (UNECE, 1999; Reis et al., 2012). Finally, the results of consultations with national experts,
30 carried within the work on the review of the National Emission Ceiling Directive (Amann et al., 2012)
31 were considered. This emission dataset was completed in April 2014 and is referred to as
32 ECLIPSE_V5; it is part of a global emission set established during the EU funded FP7 project ECLIPSE.
33 More detailed description of the data and applied emission calculation methodology is given in
34 (Amann et al., 2012) and (Klimont et al., 2016b; Klimont et al., 2016a). The respective scenario is
35 available in the freely accessible on-line version of the GAINS model³ where more detailed outputs
36 and all data inputs can be found.

37

38 **6.2 Spatial distribution of anthropogenic emissions**

39

¹ <http://ec.europa.eu/eurostat> [last access date 14 June 2017]

² <http://www.fao.org/statistics/en/> [last access date 14 June 2017]

³ <http://magcat.iiasa.ac.at> [last access date 14 June 2017]; select 'Europe' in order to access respective data and results

1 The emissions were provided by INERIS for the EDT modelling domain using the spatial regridding
2 methodology introduced in (Terrenoire et al., 2015;Bessagnet et al., 2016) which consists of:

- 3 • Europe-wide road and shipping proxies for SNAP sectors 7 and 8 (road transport and other
4 mobile sources and machinery);
- 5 • A proxy based on the population density for residential emissions (SNAP 2: non-industrial
6 combustion plants), note that emissions are not linearly proportional to the population
7 density, a fit tested with the bottom-up inventory for France is used;
- 8 • For industrial emissions (SNAP 1, 3, and 4: Combustion in energy and transformation
9 industries; Combustion in manufacturing industry; Production processes) we use the flux and
10 location from the EPRTR inventory⁴ . When the total emissions exceed the flux reported in
11 EPRTR, we used a default pattern applying the CEIP spatial distribution, available by SNAP
12 sectors (“emissions as used in EMEP models”⁵). The only exception is for particulate matter
13 emissions for which a spatial distribution was not available for 1990; for that year a
14 combination of officially reported emissions was produced by order of priority: SNAP, NRF01,
15 NRF02 and NRF09 (NFR standing for “Nomenclature for Reporting” following the 2001, 2002,
16 or 2009 guidelines).
- 17 • Bottom-up emission inventories for all SNAP for France & United Kingdom (such information
18 was not available elsewhere);
- 19 • TNO-MACC inventory for NH₃ emissions (largely dominated by SNAP10: agricultural
20 emissions);
- 21 • Default CEIP spatial distribution at a 50km resolution for the other sectors (SNAP5, 6, 9:
22 Extraction and distribution of fossil fuels and geothermal energy, Solvents and other product
23 use, Waste treatment and disposal).

24 In the applied method, only the spatial distribution of industrial emissions is supposed to have
25 changed in time over the past decades. For the residential and road sector, it was considered that
26 the recent techniques involving consistent and high-resolution proxies over Europe provide a more
27 realistic view of emissions than the 50km resolution emission data from the 1990s and early 2000s.

28 **6.3 Biogenic and natural emissions**

29

30 There were no specific constraints imposed to biogenic emissions (including soil NO emission) which
31 are represented by most CTMs using an online module. Forest fires were ignored and each modelling
32 team could decide whether they would include lightning as well as natural and dust emissions from
33 road resuspension of dust emissions (see also the synthesis in Table 1).

34

35 **7 Chemical Boundary Conditions**

36

37 Two sources of lateral and top chemical boundary conditions are used by the regional CTMs: a
38 climatology of observational data, and global model results. Both have pros and cons. Global models
39 carry biases but include a wider array of chemical species. The trend in observations matches in-situ
40 data by nature, but only at one point over the domain. For the EDT experiment the consensus in the
41 experiment design was in favour of observation-based boundary conditions for most experiments

⁴ <http://prtr.ec.europa.eu> [last access date 14 June 2017]

⁵ <http://www.ceip.at/> [last access date 14 June 2017]

1 (Tier 1A, 1B, 2A, 2C, 3A, 3B) but also include a sensitivity study based on modelled boundary
2 conditions (Tier 2B).

3 Note that a possible impact of changing chemistry composition on large scale circulation was
4 integrated in the forcing meteorological fields through the data assimilation of the ERA-Interim
5 reanalysis. This factor was not considered important to isolate for the 2-decade timescale of the
6 experiment.

7 Note also that both sources are provided on the basis of monthly averages so that sporadic advection
8 of large intercontinental pollution plumes or dust events will not be captured, although their impact
9 on monthly means is taken into account.

10 **7.1 Observations-based boundary conditions**

11

12 The boundary conditions (BCs) are a simplified version of those used in the standard EMEP/MSC-W
13 model (Simpson et al., 2012a). The values are based upon climatological data (except from those for
14 natural particles). The most important gaseous boundary condition compounds are O₃, CO and CH₄.
15 For ozone, the 3D climatology based on observational vertical profiles constructed by (Logan, 1998)
16 are used in conjunction with a temporal (monthly) variation over the past 20 years. These
17 climatological values are modified each month to ensure that their variability matches the observed
18 variability of concentrations in the clean westerly Atlantic air masses as measured at Mace Head on
19 the coast of Ireland. The 'Mace Head correction' has been derived for each year from ozone data
20 from Mace Head, sorted using sector-analysis (based on trajectories obtained from MSC-W⁶).
21 Monthly mean values of the ozone associated with easterly sectors have been calculated for
22 respective years/months, as described in (Simpson et al., 2012a).

23 For methane, uniform boundary conditions around the European domain are set to: 1780 ppb in
24 1990, 1820 ppb in 2000, and 1870 ppb in 2010 according to Mace-Head observations. For the
25 intermediary years, an interpolation is applied.

26 For sulphate (SO₄²⁻) and nitrate (NO₃⁻) aerosols, the trends for 1990-2010 are derived from the trend
27 in EPA emissions for North America of SO₂ and NO_x (Hicks et al., 2002b)⁷. For ammonium (NH₄⁺), the
28 trends are derived as 2/3*SO₄²⁻ + 1/3*NO_x. The rationale for SO₂ lies in the demonstration of the
29 close correspondence between national emissions and concentration trend in (Hicks et al., 2002a).

30 Monthly (3-dimensional) boundary conditions for sea salt and windblown mineral dust are
31 constructed based on a global run performed with the EMEP/MSC-W model for 2012. The
32 description of EMEP parameterization for sea spray and windblown dust can be found in (Simpson et
33 al., 2012b). The accuracy of the model results for sea salt and mineral dust is regularly evaluated with
34 available observations over Europe and documented in EMEP reports⁶. Model evaluation for mineral
35 dust is limited due to the scarcity of dust in-situ measurements, therefore also AOD/extinction
36 measurements from satellite, Aeronet and Earlinet have recently been used for model evaluation
37 within AeroCom⁸.

38 The uncertainty of these observation-based boundary conditions trends is important and needs to be
39 addressed in the forthcoming analyses of the experiment results, also including a comparison with
40 the model-based boundary conditions.

⁶ <http://www.emep.int> [last access date 14 June 2017]

⁷ <https://www.epa.gov/air-trends> [last access date 14 June 2017]

⁸ <http://aerocom.met.no> [last access date 14 June 2017]

1 7.2 Global model based boundary conditions

2

3 A global model simulation from the Climate-Chemistry Model Initiative (CCMI) is also used in EDT.
4 CCMI undertakes a global atmospheric chemistry reanalysis over the 1960-2010 time period (Eyring,
5 2014) based on the MACCity emissions (Granier et al., 2011). The CAM4-chem (Tilmes et al., 2016)
6 member of the CCMI ensemble was made available at monthly temporal resolution for use in EDT.

7 The model uses a full tropospheric and stratospheric chemistry scheme (Lamarque et al., 2012) based
8 on MOZART (Model for Ozone and Related chemical Tracers) version 4 (Emmons et al., 2010). CAM4-
9 chem considers 56 vertical levels from the surface to about 40 km with 1.9° x 2.5° horizontal
10 resolution. The simulation used in this analysis was performed in nudging the model to
11 meteorological fields from the MERRA GEOS-5 (Modern Era Retrospective Analysis for Research and
12 Application Goddard Earth Observing System Data Assimilation System Version 5) reanalysis provided
13 by the Global Modelling and Assimilation Office (GMAO).

14 Evaluation of this global reanalysis is ongoing, but the preliminary results are encouraging as
15 illustrated in Figure 3 which shows the modelled and observed ozone trend at the Mace Head
16 station.

17 8 Output format and database status

18

19 The model simulations were delivered in a common NetCDF format, so that each of the files contains
20 gridded fields of one pollutant for a whole year. The air pollutant concentrations from only the
21 lowest model level (or corrected to 3m height for EMEP, LOTOS-EUROS and MATCH) are delivered to
22 the project database, but the participants are encouraged to store 3D data if their storage capacities
23 allow such an archiving.

24 The requested variables are:

- 25 • Hourly concentrations of O₃ (O3_HL) and NO₂ (NO2_HL);
- 26 • Daily concentrations of aerosols: nitrate (NO₃⁻), sulphate (SO₄²⁻) and ammonia (NH₄⁺), sea-
27 salt, dust, total primary PM, anthropogenic and biogenic secondary organic aerosols, and
28 total PM, both for the fraction below 2.5µm (PM_{2.5}), and the fraction below 10µm (PM₁₀);
- 29 • Daily concentrations of reactive gases: NH₃, SO₂, an indicator of alpha-pinene that shall
30 depend on the chemical mechanism of each model, isoprene, HNO₃, H₂O₂, HCHO, PAN, total
31 VOC, biogenic VOC;
- 32 • Daily emission rate of biogenic species: isoprene, and an indicator of alpha-pinene that shall
33 depend on the chemical mechanism of each model;
- 34 • Monthly dry and wet deposition of total oxidized sulphur (SO_x), oxidised nitrogen (NO_x) and
35 reduced nitrogen (NH_x);
- 36 • Hourly meteorological fields: temperature at 2m, wind speed, PBL, rain.

37 Additional diagnostics were subsequently computed and delivered on the common database, the list
38 of indicators and their definitions is available in Table 5.

1 The status of models' delivery of results for each of the experiment tiers at the time of submission of
2 the present article is summarized in Table 3. The access to the database is open for research use
3 through the AeroCom server (see also the section on data availability)⁹.

4 **9 Sample Results**

5

6 A few illustrations of the results of the Eurodelta-Trend multi-model air quality hindcast are provided
7 in Figure 4 and Figure 5 with ensemble-median and ensemble-spread concentration maps of ozone
8 and particulate matter in 1990 and 2010, obtained from the eight models which delivered output to
9 Tier 1A. For ozone, we show the summertime (June-July-August) average of the daily maxima of 8-hr
10 mean ozone. For particulate matter, the annual mean PM₁₀ is presented.

11 It is the ambition of the whole Eurodelta-Trend experiment to assess how those maps compare with
12 observations, both in terms of spatial variability and temporal trends, and also to further explain the
13 rationale for the changes observed between 1990 and 2010. Such analyses require however
14 substantial work that is left out of the present article devoted to the presentation of the experiment.

15 It is worth highlighting however that substantial decreases of both ozone peaks and particulate
16 pollution are modelled in the Eurodelta-Trends ensemble between 1990 and 2010. We present here
17 the decrease on the basis of 1990 and 2010 snapshots for the whole 8 model ensemble that
18 contributed to the experiment. But it would require to be further documented in terms of trends by
19 comparison with the subset of five models that produced the full set of 21-yr trend simulation in tier
20 3A.

21 For summertime ozone, concentrations exceeding the European target value of 120 µg/m³ are only
22 found in the greater Mediterranean region in 2010, whereas in the early 1990s, such concentrations
23 affected much larger areas of continental Europe. The spread (standard deviation) of the models is
24 much larger in 1990 than 2010, especially over the polluted areas of Europe at that time.

25 Particulate matter concentrations also decreased substantially. The largest spread in the eight-
26 member ensemble is found over sea and desert areas (also because absolute concentrations are high
27 over North Africa), where the differences between the models changes significantly between 1990
28 and 2010. This raises important questions regarding the uncertainties of the models for natural
29 sources and the role of inter-annual meteorological variability on aerosol concentrations.

30 **10 Summary and Outlook**

31

32 The Eurodelta-Trend modelling experiment (EDT) will allow a better understanding of the evolution
33 of regional scale air quality over Europe over the 1990-2010 period. This is facilitated by the
34 thoroughly designed modelling plan. Eight modelling teams have participated in the EDT experiment,
35 though with a variable degree of involvement. The base runs of Tier 1A, completed with eight
36 participating models, offer a great opportunity to assess the capability of these state-of-the-art
37 chemistry-transport models to reproduce the observed changes in the concentrations of the main
38 pollutants, including ozone, particulate matter and its individual components, as well as in
39 precipitation chemistry. This analysis will then be complemented by an assessment of the capability
40 in reproducing the actual trends over the 21yr in the 1990-2010 period for the models participating
41 in the more demanding tier 3A experiment. If this evaluation phase concludes that the skill of these

⁹ <https://wiki.met.no/aerocom/user-server> [last access date 14 June 2017]

1 models in capturing air quality evolution is satisfactory, we would then rely on the results of the
2 trend (or decadal changes) calculations and the sensitivity experiments and recommend that they
3 can be used when addressing science and policy questions underlying the evolution of air quality in
4 Europe over the past couple of decades.

5 The critical policy question lies in the attribution of air quality trends to emission changes, to influx at
6 the boundaries of the European domain, and to interannual meteorological variability (and natural
7 sources of trace species) and will be addressed in a series of upcoming papers. Furthermore, thanks
8 to the multi-model design of the experiment, other scientific questions with regard to the role of
9 specific chemical and physical processes will be investigated in forthcoming studies based on the
10 Eurodelta-Trends results.

11 The model results will also be publicly distributed in order to serve for in depth analyses to scientific
12 communities working on the impacts of air pollution on health, ecosystems or aerosol radiative
13 forcing.

14 **Data availability**

15

16 Technical details allowing forthcoming replication of the experiment are available on the wiki of the
17 EMEP Task Force on Measurement and Modelling¹⁰ and that also provides ESGF links to
18 corresponding input forcing data.

19 The Eurodelta-Trends model results are made available for public use on the AeroCom server¹¹ under
20 the following terms:

- 21 • Data provided on this server may be used solely for research and education purposes;
- 22 • Eurodelta-Trends partners cannot guarantee that the data are correct in all circumstances.
23 Neither do they accept any liability whatsoever for any error or omission in the data, or for
24 any loss or damage arising from its use;
- 25 • Data must not be supplied as a whole or in part to any third party without authorization;
- 26 • Articles, papers, or written scientific works of any form, based in whole or in part on data,
27 images or other products supplied by Eurodelta-Trends will contain an acknowledgment
28 concerning the supplied data reading:
 - 29 ○ “Modelling data used in the present analysis were produced in the framework of the
30 EuroDelta-Trends Project initiated by the Task Force on Measurement and Modelling
31 of the Convention on Long Range Transboundary Air Pollution. EuroDelta-Trends is
32 coordinated by INERIS and involves modelling teams of BSC, CERE, CIEMAT, ENEA,
33 IASS, JRC, MET Norway, TNO, SMHI. The views expressed in this study are those of
34 the authors and do not necessarily represent the views of Eurodelta-Trends
35 modelling teams.”
- 36 • Users of these data must offer co-authorship to the modelling teams for any study submitted
37 for publication until June 2018. The list of modellers is: CHIMERE (A. Colette, F. Couvidat, B.
38 Bessagnet), CMAQ (M.T. Pay), EMEP (S. Tsyro, H Fagerli, P. Wind), ex-JRC (C. Cuvelier),
39 LOTOS-EUROS (A. Manders), MATCH (C. Andersson, R. Bergström), MINNI (M. Mircea, G.
40 Briganti, A. Cappelletti, M. Adani, M. D'Isidoro), POLR (V. Raffort), WRF-Chem (K.A. Mar, N.
41 Otero, N. Ojha). After this date, users must inform the Eurodelta-Trends coordinator

¹⁰ <https://wiki.met.no/emep/emep-experts/tfmmtrendeuodelta> [last access date: 14 June 2017]

¹¹ <https://wiki.met.no/aerocom/user-server> [last access date: 14 June 2017]

1 (augustin.colette@ineris.fr) about the expected use of the data. The coordinator will, in turn,
2 inform a representative from each modelling team.
3

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30 research programmes (<http://www.cresco.enea.it/english>).
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- 36 • The MATCH participation was partly funded by the Swedish Environmental Protection
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- 39 • RACMO2 simulations at KNMI to provide meteorological forcings for LOTOS-EUROS were
40 supported by the Dutch Ministry of Infrastructure and the Environment.

1 Table 1: Main features of the Chemistry-Transport Models involved in the Eurodelta-Trends modelling exercise

MODEL	CHIMERE	CMAQB	EMEP	LOTOS-EUROS	MATCH	MINNI	POLYPHEMUS	WRF-CHEM
version	Modified Chimere2013	V5.0.2	rv4.7	v1.10.005	VSOA April 2016	V4.7	V1.9.1	V3.5.1
operator	INERIS	BSC	MET Norway	TNO	SMHI	ENEA/Arianet S.r.l.	CEREA	IASS
Chemistry/Meteorology coupling	offline	offline	offline	offline	offline	offline	Offline	Online
Name and resolution of the meteorological driver	WRF (common driver after (Stegehuis et al., 2015)). 0.44 deg.	WRF. 25km	WRF (common driver after (Stegehuis et al., 2015)). 0.44 deg.	RACOMO2, 0.22 deg.	HIRLAM EURO4M reanalysis, approx. 22km	WRF (common driver after (Stegehuis et al., 2015)). 0.44 deg.	WRF (common driver after (Stegehuis et al., 2015)). 0.44 deg.	WRF, approx. 25km (common driver used for initial and lateral boundary conditions, and for applying four-dimensional data assimilation (FDDA), with coefficients as described in (Mar et al. 2016).
Vertical layers	9 sigma	15 sigma	20 sigma	5(4 dynamic layers and a surface layer)	39 hybrid eta utilizing the meteorological model layers	16 fixed terrain-following layers	9 Fixed terrain following layers	35 terrain-following
Vertical extent	500 hPa	50 hPa	100 hPa	5000 m	ca 5000 m (4700 – 6000 m)	10000 m	12000m	10 hPa
Depth first layer	20 m	40 m	90 m	25 m	ca 60m	40 m	40m	50 m
Surface concentration	First model level	First model level	Downscaled to 3m using dry deposition velocity and similarity theory	Downscaled to 3m	Downscaled to 3m	First model level	First model level	First model level
Biogenic VOC	MEGAN model v2.1 with high resolution spatial and temporal LAI (Yuan et al., 2011) and recomputed emissions factors	MEGAN model v2.04 (Guenther et al., 2006)	Based upon maps of 115 species from (Koeble and Seufert, 2001), and hourly temperature and light using (Guenther et al.,	Based upon maps of 115 species from (Koeble and Seufert, 2001), and hourly temperature and light (Guenther et al 1991,	(Simpson et al., 2012a), based on hourly temperature and light.	MEGAN v2.04 (Guenther et al., 2006)	MEGAN V2.04 (Guenther et al., 2006)	MEGAN v2.04 (Guenther et al., 2006)

	based on the landuse (Guenther et al., 2006)		1993;Guenther et al., 1994). See (Simpson et al., 1995;Simpson et al., 2012a)	Guenther et al 1993). See (Beltman et al., 2013)				
Forest fires	None	None	None	None	None	None	None	None
Soil-NO	MEGAN model v2.04	MEGAN model v2.04	See in (Simpson et al., 2012a)	Not used here	None	MEGAN v2.04	MEGAN V2.04	MEGAN v2.04
Lightning	None	None	Monthly climatological fields, (Köhler et al., 1995)	None	None	None	None	None
Sea salt	(Monahan, 1986)	Open ocean and surface-zone (Kelly et al., 2010)	(Monahan, 1986) and (Martensson et al., 2003), see (Tsyro et al., 2011)	(Martensson et al., 2003) and (Monahan, 1986), see (Schaap et al., 2009)	Based on parameterization by (Sofiev et al., 2011)	(Zhang K.M. et al., 2005)	(Monahan, 1986)	(Gong et al., 1997), (O'Dowd et al., 1997)
Windblown Dust	(Vautard et al., 2005), not used here	None	See (Simpson et al., 2012a)	(Schaap et al., 2009)	Not used here	(Vautard et al., 2005)	None	None
Dust traffic suspension	None	None	(Denier van der Gon et al., 2010)	None	Not used here	None	None	None
Landuse database	GLOBCOVER (24 classes)	Corine Land Cover 2006 (44 classes)	CCE/SEI for Europe, elsewhere GLC2000	Corine Land Cover 2000 (13 classes)	CCE/SEI for Europe	Corine Land Cover 2006 (22 classes)	Global Land Cover 2000 (24 classes)	24-category USGS landuse
Advection scheme	(van Leer, 1984)	Horizontal: WRF-based scheme, vertical: Piecewise Parabolic Method	(Bott, 1989)	(Walcek, 2000)	Fourth order mass-conserved advection scheme based on (Bott, 1989)	Blackman cubic polynomials (Yamartino, 1993)	Third-order Direct Space Time scheme (Spee, 1998) with Koren-Sweby flux limiter function	Runge-Kutta 3rd order
Vertical diffusion	Kz approach following (Troen and Mahrt, 1986)	ACM2 PBL scheme (Pleim, 2007)	Kz approach following (O'Brien, 1970) and (Jeričević et al., 2010)	Kz approach Yamartino et al (2004)	Implicit mass conservative Kz approach, see (Robertson et al., 1999) Boundary layer parameterisation as detailed in (Robertson	Kz approach following (Lange, 1989)	Kz approach following (Troen and Mahrt, 1986)	Yonsei University PBL scheme (Hong et al., 2004)

					et al., 1999) forms the basis for vertical diffusion and dry deposition			
Dry deposition	Resistance approach (Emberson et al., 2000a; Emberson et al., 2000b)	Resistance approach (Venkatram and Pleim, 1999)	Resistance approach for gases (Venkatram and Pleim, 1999) for aerosols, (Simpson et al., 2012a)	Resistance approach, DEPAC3.11 for gases, (Van Zanten et al., 2010) and (Zhang et al., 2001) for aerosols	Resistance approach depending on aerodynamic resistance, and land use (vegetation). Similar to (Andersson et al., 2007)	Resistance model based on (Wesely, 1989)	Resistance approach for gases (Zhang et al., 2003) and aerosols (Zhang et al., 2001)	(Wesely, 1989) and (Erisman et al., 1994)
Ammonia compensation points	None	None	None, but zero NH ₃ deposition over growing crops	Only for NH ₃ (for stomatal, external leaf surface and soil (= 0))	None	None	None	None
Stomatal resistance	(Emberson et al., 2000a; Emberson et al., 2000b)	(Wesely, 1989)	DO3SE-EMEP: (Emberson et al., 2000a; Emberson et al., 2000b), (Tuovinen et al., 2004; Simpson et al., 2012a)	(Emberson et al., 2000a; Emberson et al., 2000b)	Simple, seasonally varying, diurnal variation of surface resistance for gases with stomatal resistance (similar to (Andersson et al., 2007))	(Wesely, 1989)	(Zhang et al., 2003)	(Wesely, 1989) and (Erisman et al., 1994)
Wet deposition gases	In-cloud and sub-cloud scavenging coefficients	In-cloud and sub-cloud scavenging which depends on Henry's law constants, dissociation constants and cloud water pH (Chang et al., 1987)	In-cloud and sub-cloud scavenging coefficients	sub-cloud scavenging coefficient	In-cloud scavenging of some species based on Henry's law constants. Simple in-cloud and sub-cloud scavenging coefficients for other gases.	In-cloud and sub-cloud scavenging coefficients (EMEP, 2003)	In-cloud (monodispersed raindrops with constant collection efficiency) and below cloud (Sportisse and Dubois, 2002) scavenging coefficients	In-cloud and sub-cloud scavenging coefficients
Wet deposition particles	In-cloud and sub-cloud scavenging	In-cloud and sub-cloud scavenging	In-cloud and sub-cloud scavenging	sub-cloud scavenging coefficient	In-cloud and sub-cloud scavenging. Similar to (Simpson et al., 2012a)	In-cloud and sub-cloud scavenging coefficients	In-cloud (as for gas) and below cloud (Slinn, 1983) scavenging coefficients	In-cloud and sub-cloud scavenging coefficients
Gas phase chemistry	MELCHIOR2	CB-05 with chlorine chemistry extensions	EmChem09 (Simpson)	TNO-CBM-IV	Based on EMEP (Simpson et al., 2012),	SAPRC99	CB-05 (Yarwood, G. et	RADM2 (Stockwell et al., 1990) with

		(Yarwood. G. et al., 2005)	et al., 2012a)		with modified isoprene chemistry (Carter, 1996;Langner et al., 1998)	(Carter;Carter, 2000)	al., 2005)	updates made to inorganic rate coefficients as described in supplementary material to (Mar et al. 2016).
Cloud chemistry	Aqueous SO ₂ chemistry and pH dependent SO ₂ chemistry	Aqueous SO ₂ chemistry (Walcek and Taylor, 1986)	Aqueous SO ₂ chemistry, pH dependent	Aqueous SO ₂ chemistry, pH dependent (Banzhaf et al., 2012)	Aqueous SO ₂ chemistry	Aqueous SO ₂ chemistry (Seinfeld and Pandis, 1998)	Aqueous SO ₂ chemistry (Seinfeld and Pandis, 1998)	None
Coarse nitrate	No reaction with Ca even if reaction with Na is taken into account. Coarse nitrate might exists with transfer from smaller particles	None	Two formation rates of coarse NO ₃ from HNO ₃ for relative humidity below/above 90%	(Wichink Kruit et al., 2012)	Yes, transfer of HNO ₃ (g) to aerosol nitrate using rate from (Strand and Hov, 1994)	None	No heterogeneous nitrate formation	None
Ammonium nitrate equilibrium	ISORROPIA v2.1 (Nenes et al., 1999)	ISORROPIAv2.1	MARS (Binkowski and Shankar, 1995)	ISORROPIA v.2	RH & T dependent equilibrium constant (Mozurkewich, 1993)	ISORROPIA v1.7 (Nenes et al., 1998)	ISORROPIA v1.7 (Nenes et al., 1999)	MARS (Binkowski and Shankar, 1995)
SOA formation	H ₂ O (Couvidat et al., 2012) mechanism coupled with the thermodynamic model SOAP (Couvidat and Sartelet, 2015)	SORGAM module (Schell et al., 2001)	VBS-NPAS –(Simpson et al., 2012a)	Not used here	Similar to VBS-NPNA (Bergström et al., 2012)	SORGAM module (Schell et al., 2001)	H ₂ O (Couvidat et al., 2012)	SORGAM module (Schell et al., 2001)
Volatility basis set for aerosols	None	None	(Simpson et al., 2012a;Bergström et al., 2012)	Not used here	Yes, based on Bergström et al. (2012)	None	None	None
Aerosol model	9 bins (10 nm to 10 μm)	AERO5 (Carlton et al., 2010), log-normal approach (3 modes)	Bulk- approach (fine and coarse modes)	Bulk- approach (2 modes)	Bulk approach	AERO3 (Binkowski, 1999); 3 modes: Aitken, accumulation, coarse	5 bins (0.01 to 10μm)	MADE (Ackermann et al., 1998)
Aerosol physics	coagulation/condensation/nucleation	Coagulation/condensation/nucleation	Not used here	Not used here	Not used here	Coagulation/condensation/nucleation	Coagulation/Condensation	Coagulation/condensation/nucleation

	Computation of the wet diameter for each bins as a function of humidity (used for coagulation, condensation, deposition)							
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1

2

- 1 Table 2 : Summary of model experiments (including label), corresponding key scientific questions.
- 2 The simulations are labelled MyyByyEyy where M indicates meteorology, B indicates observation-
- 3 based boundary conditions, C indicates modelled-based boundary condition, E indicates emission,
- 4 and yy is the 2-digits reference of the corresponding year.

Tier	Experiment	Key question (Q) / Action (A)	Label
1A	Meteorology, boundary conditions and emissions of 1990, 2000 and 2010.	Q: What is the uncertainty within the seven CTMs ensemble in 1990, 2000, and 2010? A: Comparison 1A vs. Observations for 1990, 2000 and 2010	M10B10E10
			M00B00E00
			M90B90E90
1B	Meteorology and boundary conditions of 2010. Emissions of 1990 and 2000.	Q: What if no emission change occurred in Europe? A: Comparison 1A vs. 1B	M10B10E00
			M10B10E90
2A	Meteorology of 2010. Emissions and boundary conditions of 1990 and 2000.	Q: What if no emission changed beyond Europe? A: Comparison 2A vs. 1B	M10B00E00
			M10B90E90
2B	Meteorology and emissions of 2010. Modelled boundary conditions of 1990, 2000, 2010	Q: What is the uncertainty related to boundary conditions? A: Comparison 2A vs. 2B	M10C10E10
			M00C00E00
			M90C90E90
2C	Meteorology of 2000, emissions of 1990 and boundary conditions of 2000 and 1990.	Additional simulations for decomposition of factors in the 1990s and 2000s	M00B90E90
			M00B00E90
3A	21-years reference trend from 1990 to 2010	Q: How do the models capture the trend in observations?	MyyByyEyy
		A: Comparison 3A vs. observations	
3B	21-years trend with 2010 emissions	Q: Does meteorological variability contribute to the AQ trend over the past 20 years?	MyyByyE10
		A: Comparison 3A vs. 3B	

5

6

- 1 Table 3: Synthesis of models having delivered (D) data or planning to (P) to the project database for each of the
- 2 experiments.

Tier	Label	LOTOS- MATCH						WRF-	
		CHIMERE	CMAQB	EMEP	EUROS	MINNI	Polyphemus	Chem	
1A	M10B10E10	D	D	D	D	D	D	D	D
	M00B00E00	D	D	D	D	D	D	D	D
	M90B90E90	D	D	D	D	D	D	D	D
1B	M10B10E00	D	D	D	D	D	D	D	D
	M10B10E90	D	D	D	D	D	D	D	D
2A	M10B00E00	D	D	D	D	D	D	D	P
	M10B90E90	D	D	D	D	D	D	D	P
2B	M10C10E10	D		D			D		P
	M00C00E00	D		D			D		P
	M90C90E90	D		D			D		P
2C	M00B90E90	D	D	D	D	D	D	D	P
	M00B00E90	D	D	D	D	D	D	D	P
3A	MyyByyEyy	D		D	D	D	D		P
3B	MyyByyE10	D		D	D	D	D		

3

4

1 Table 4: Meteorological fields used in the EDT project. WRF-0.44 corresponds an optimized and nudged version
 2 of the WRF-IPSL-INERIS Eurocordex member at 0.44 degrees from EuroCordex climate downscaling programme
 3 (Jacob et al., 2013) used by most CTMs in EDT. WRF-25 corresponds to the WRF run in the same condition as
 4 WRF-0.44 in a Lambert Conformal Conic projection used to drive CMAQB. WRF-Chem indicates the
 5 configuration of WRF used within the WRF-Chem online CTM. RACMO2 is the meteorological model used by
 6 LOTOS-EUROS.

Model configuration	WRF-0.44	WRF-25	WRF-Chem	HIRLAM EURO4M	RACMO2
Model version	WRF v3.3.1	WRF v3.3.1	WRF v3.5.1	HIRLAM 3DVAR upper air analysis and OI surface analysis (for details and evaluation see (Dahlgren et al., 2016))	RACMO2.3 (Meijgaard et al., 2012)
Initial and boundary conditions	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)	WRF-0.44 simulation used by other EDT models	ERA-Interim global reanalysis (resolution ~80km) (Dee et al., 2011)	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)
Coordinate system	Rotated latitude and longitude	Lambert Conformal	Latitude and longitude	Rotated latitude and longitude	Rotated latitude and longitude with a South Pole at 47S and 10E.
Horizontal setting / number of zonal and meridional grid cells	0.44° x 0.44° (120-117)	25 km x 25 km (176-197)	Approx. 25 km x 25 km (144-154)	Approx. 22km x 22km (326-341)	0.22x0.22 (306x220)
Vertical setting	31 layers	31 layers	34 layers	60 layers eta coordinates	40 layers hybrid coordinates
Microphysics	Morrison DM (Morrison et al., 2009)	Morrison DM (Morrison et al., 2009)	Morrison DM (Morrison et al., 2009)	Large-scale condensation with Rasch-Kristjansson scheme (Rasch and Kristjánsson, 1998)	Prognostic cloud scheme (Tiedtke, 1993), Large-scale condensation (Tompkins et al., 2007), boundary-layer clouds (Neggers, 2009)
LW,RW radiation	RRTMG - (Iacono et al., 2008)	RRTMG - (Iacono et al., 2008)	RRTMG - (Iacono et al., 2008)	(Savijärvi, 1990)	Short wave radiation (Clough et al., 2005; Morcrette et al., 2008) Long wave radiation (Mlawer et al., 1997; Morcrette et al., 2001)

Cumulus scheme	Tiedtke - (Tiedtke, 1989;Zhang et al., 2011)	Tiedtke - (Tiedtke, 1989;Zhang et al., 2011)	Grell 3D scheme ¹² (Grell and Dévényi, 2002)	Convective processes Kain-Fritsch scheme (Kain, 2004)	Mass flux scheme (Tiedtke, 1989;Nordeng, 1994;Neggers et al., 2009;Siebesma et al., 2007)
Boundary & Surface layer	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	Turbulence CBR scheme (Cuxart J. et al., 2000); adaptions for moist CBR (Tijm and Lenderink, 2003)	Eddy-Diffusivity Mass Flux Scheme with TKE prognostic variable (Lenderink and Holtslag, 2004;Siebesma et al., 2007)
Soil	NOAH (Tewari et al., 2004)	NOAH (Tewari et al., 2004)	NOAH (Tewari et al., 2004)	Further developed ISBA scheme (Noilhan and Planton, 1989;Noilhan J. and J.-F., 1996;Gollvik and Samuelsson, 2010)	TESSEL (Van den Hurk et al., 2000), HTESSEL (Balsamo et al., 2009)

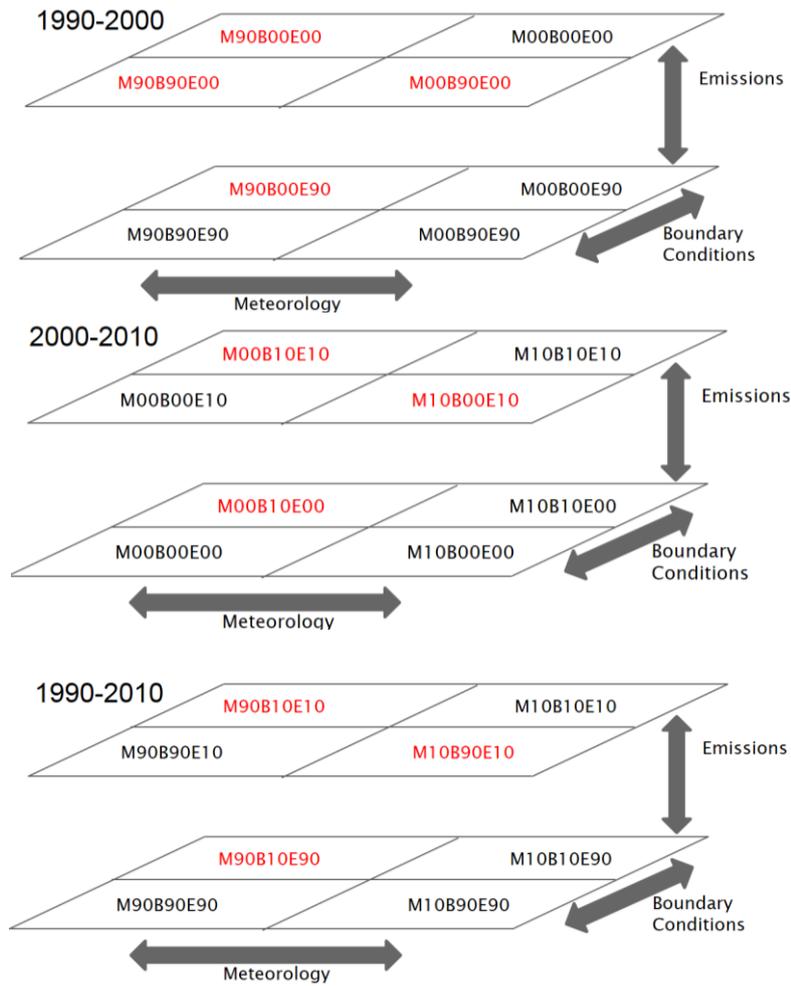
1

¹² A different scheme was chosen for compatibility with chemistry, in particular so that there would be subgrid convective transport of chemical species.

O3_DL	daily ozone computed on the basis O3_HL as the mean value for each day between 00:00 and 23:00 UTC.
O3Aot40_DL	accumulated ozone over 40ppbv computed on the basis of O3_HL, for each day (from 1 May until 31 July) as the sum of all the daylight hourly O3_HL values exceeding the value of 40 ppb (80 ug/m ³). Note that hourly values in the models correspond to instantaneous values: e.g. O3_HL(0) is for 00:00, O3_HL(23) is for 23:00). Therefore, the accumulation of AOT between 8hr and 20hr was taken as the sum of O3_HL between 8:00 and 19:00, included. O3Aot40_DL is a daily quantity that must be cumulated over a given period of the year, e.g. May-June-July in the European Air Quality Directive (EC, 2008). Its units are: (µg/m ³)*hours
O3Aot60_DL	same as before, but with a threshold of 60 ppb (120 µg/m ³) and to be accumulated over the period 1 April - 30 September. Its units are: (µg/m ³)*hours
O3hr8_HL	the 8-hr running mean hourly ozone computed from O3_HL. To each hour ih in O3hr8_HL the running mean is that of the 8 past values of O3_HL : O3hr8_HL(ih) = [O3_HL(ih) + ... O3_HL(ih-7)] /8
O3hr8Somo35_DL	Sum of ozone means over 35ppbv computed from O3hr8_HL for each day of the year as the exceedance of the daily max O3hr8_HL with respect to 35 ppb (70 µg/m ³). The accumulated value used in the Air Quality Directive is the sum over all days of the year. Its units are: (µg/m ³)*days
O3hr8Max_DL	Maximum daily value of O3hr8_HL, sometimes also referred to as MDA8 as Ozone Maximum Daily Average
O3hr8Exc60_DL	computed from O3hr8_HL. For each day of the year a value of 1 is assigned if the maximum daily value of O3hr8_HL exceeds 60 ppb (120 µg/m ³), otherwise equal to zero. The value mentioned in the Directive is the sum over all days of the year. Units are: days.
NO2_DL	Computed from NO2_HL, same as O3_DL
NO2hr1Max_DL	Computed from NO2_HL, Maximum daily value of NO2_HL
NO2hr1Exc200_DL	Computed from NO2_HL, for each day of the year a value of 1 is assigned if the maximum daily value of NO2_HL exceeds 200 ppb, otherwise equal to zero. The value mentioned in the Directive value is the sum over all days of the year. Its units are: days
NOx-ppb DL and HL	Sum of NO and NO2 in ppb, i.e. NO(µg/m ³)*22.4/30 + NO ₂ (µg/m ³)*22.4/46
PM10Exc50_DL	Computed from daily mean PM ₁₀ (PM10_DL), for each day of the year a value of 1 is assigned if the (daily) value of PM10_DL exceeds 50 ug/m ³ , otherwise equal to zero. The value in the Directive is the sum over all days of the year. Units are: days
TNO3-N	Sum of NO ₃ -10 and HNO ₃ in µgN/m ³ , i.e. NO ₃ -10(µg/m ³)*14/62 + HNO ₃ (µg/m ³)*14/63
TNH4-N	Sum of NH ₄ -10 and NH ₃ in µgN/m ³ : i.e. NH ₄ -10(µg/m ³)*14/18 + NH ₃ (µg/m ³)*14/17
TSO4-S	Sum of SO ₄ -10 and SO ₂ in µgS/m ³ : SO ₄ -10(µg/m ³)*32/96 + SO ₂ (µg/m ³)*32/64
NOz	Sum of HNO ₃ , and PAN in ppb. Conversion factors from µg/m ³ to ppb: [24/63,24/53]
NOy	Sum of NO ₂ , NO, HNO ₃ , and PAN in ppb. Conversion factors from µg/m ³ to ppb: [24/46,24/30,24/63,24/53]

1 Table 5: List and definition of air pollution indicators derived from the model results and available in the project
2 database

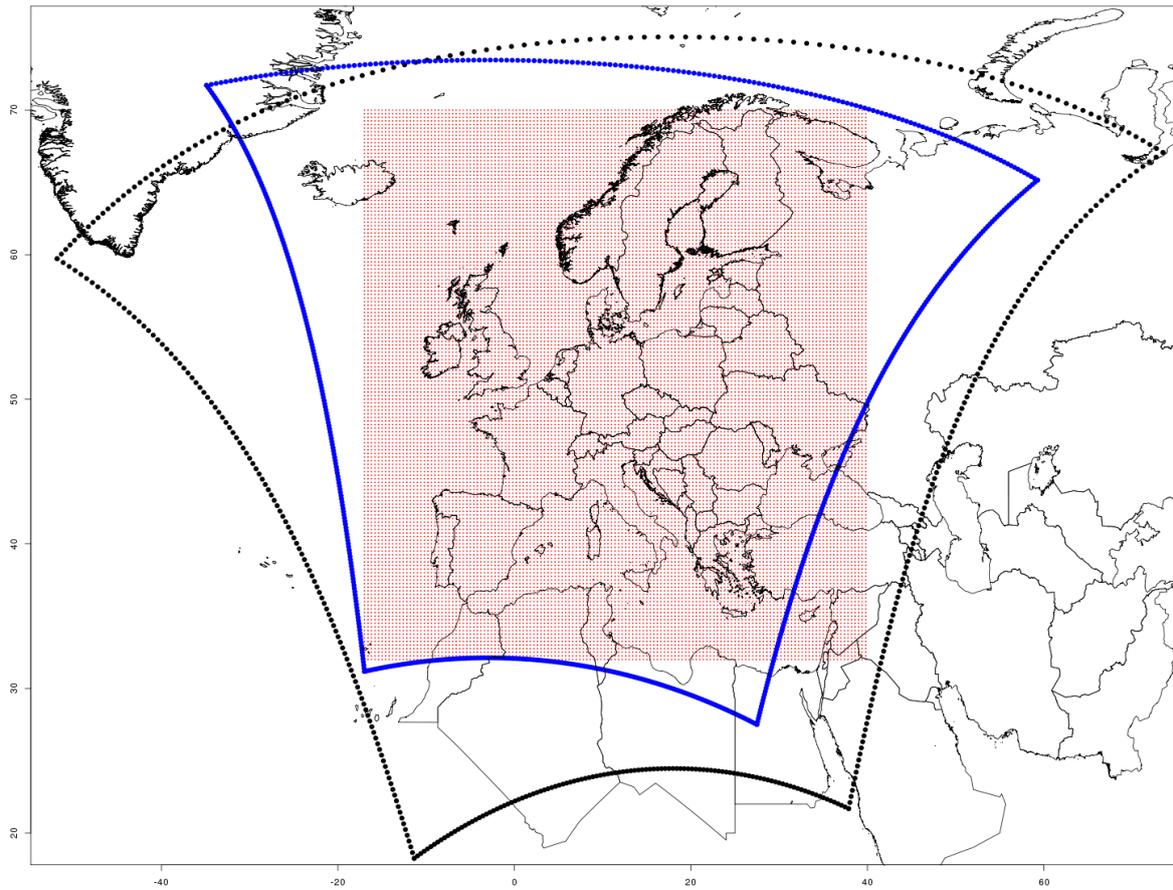
1



2

3 Figure 1 : Combination of sensitivity simulations required to perform the analysis of the contribution of (i)
4 meteorology, (ii) boundary conditions, and (iii) emission changes for the 1990-2000, 2000-2010, and 1990-2010
5 years from the top to the bottom. The key to EDT model simulations provides the 2-digit modelled year for
6 meteorology (M), boundary conditions (B) and emissions (E). Black labels are for the simulations included in the
7 experiment, and red labels are the combinations not produced in any of the tiers of experiments.

8



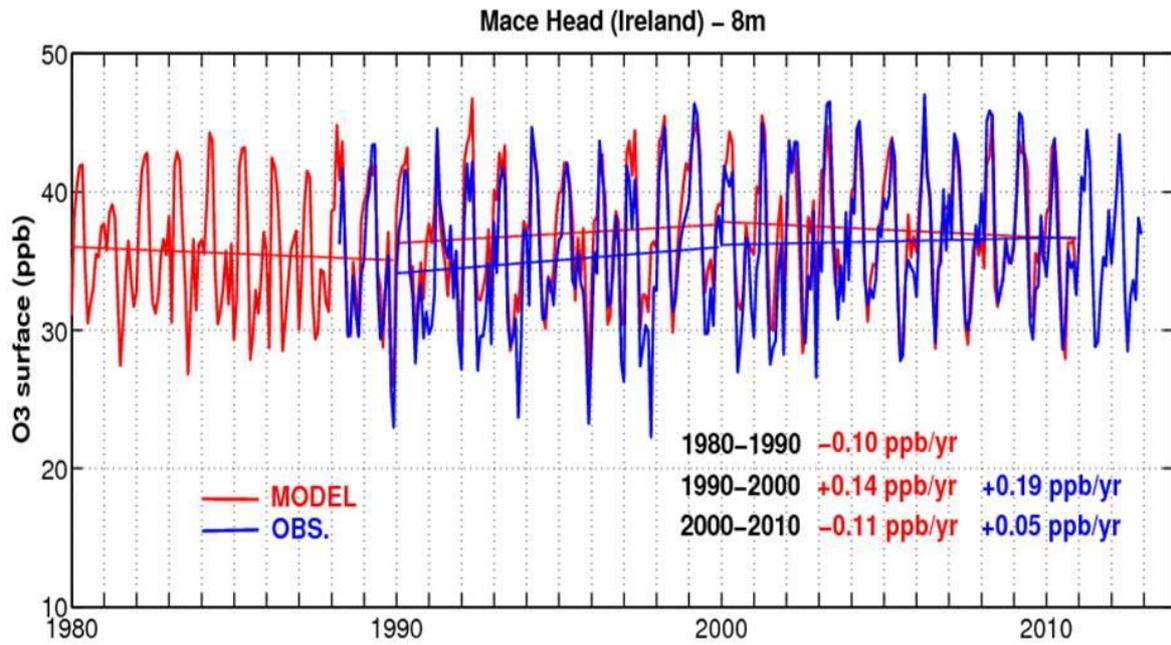
1

2 Figure 2: Modelling grid used by all the chemistry transport models involved in Eurodelta-Trends (red dots)
 3 with the exception of CMAQB that could not implement a regular latitude/longitude grid (outer grid cell of the
 4 modelling domain displayed with blue dots). The outer grid cells of the meteorological forcing data on the
 5 EuroCordex grid is also displayed (black dots).

6

7

1

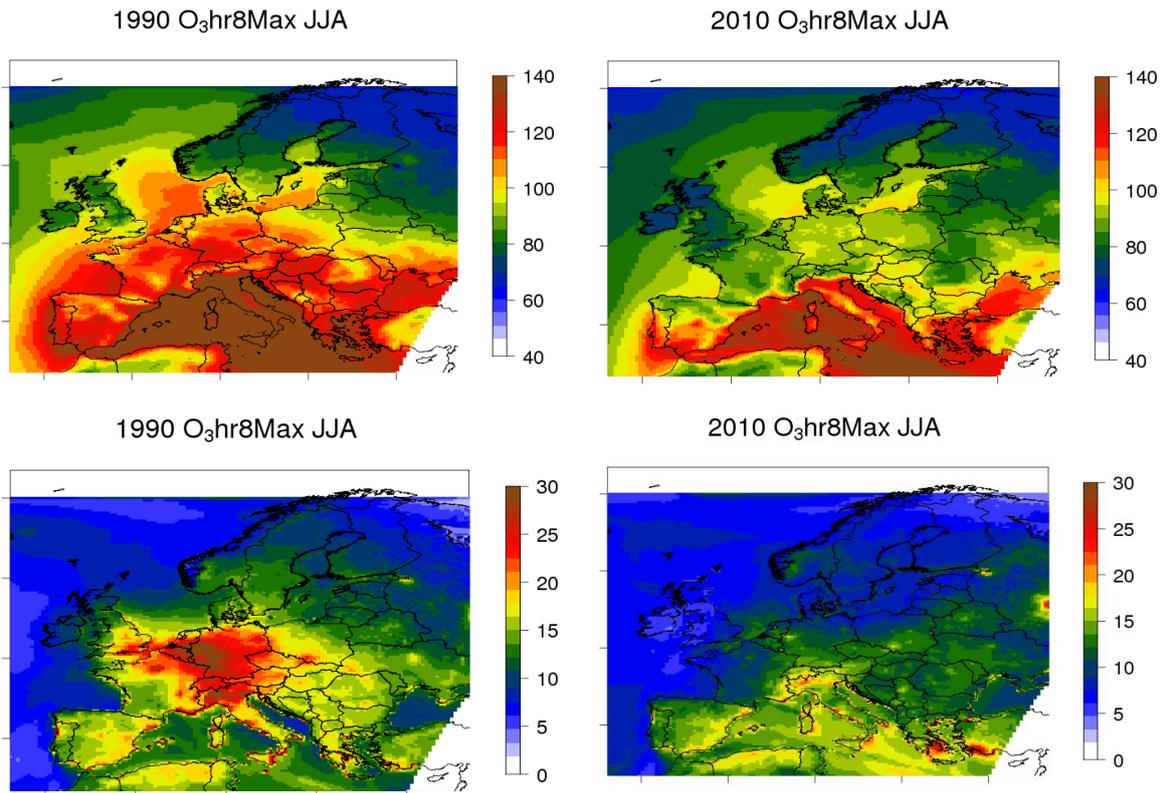


2

3

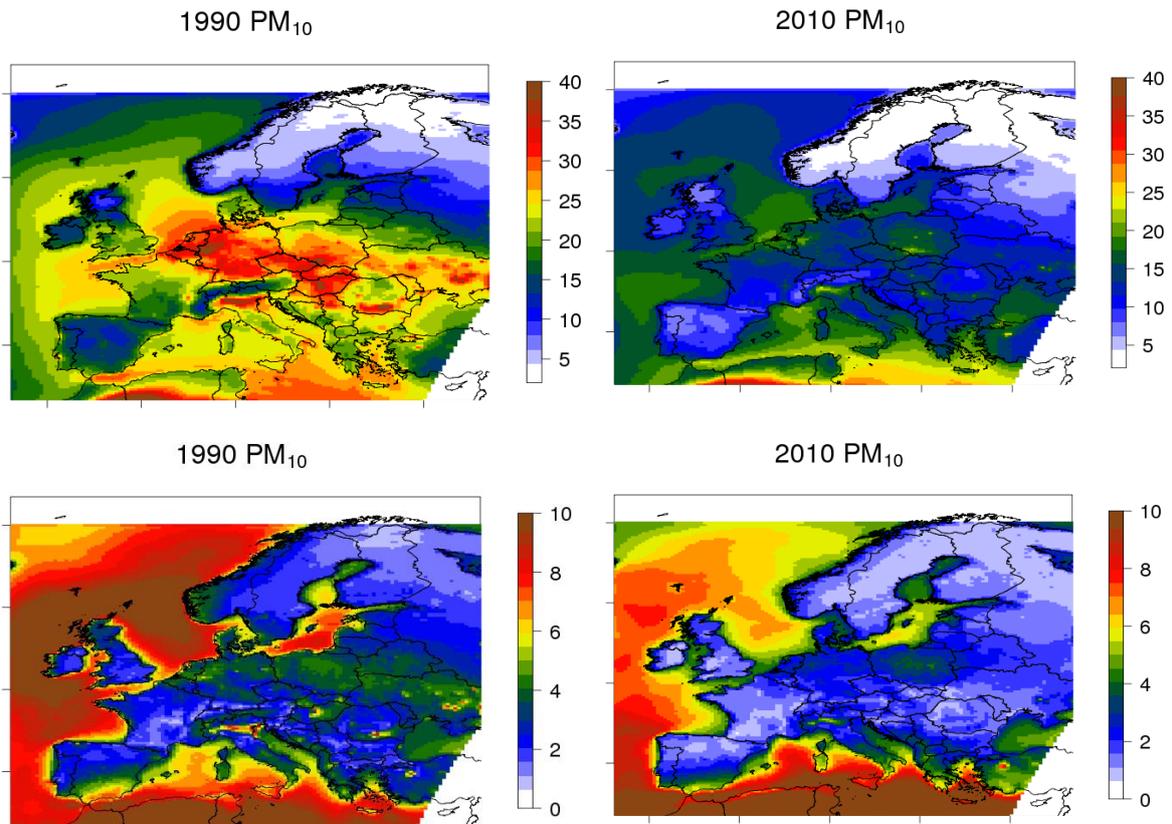
Figure 3 : Monthly variation of surface ozone (in ppb/year) at the Mace Head station observed (blue) and modelled (red) in the CamChem member of the Climate-Chemistry Model Initiative (CCMI)

4



1 Figure 4: Eight-model ensemble results for 1990 (first column) and 2010 (second column) for
 2 summertime ozone peaks (June-July-August means of 8-hr mean daily maxima, $\mu\text{g}/\text{m}^3$). Top:
 3 ensemble median, bottom: ensemble spread (standard deviation).

4



2 Figure 5: Eight-models ensemble results for 1990 (first column) and 2010 (second column) for annual
 3 mean PM₁₀ (µg/m³). Top: ensemble median, bottom: ensemble spread (standard deviation).

4

5

6 Bibliography

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- 8 Ackermann, I. J., Hass, H., Memmesheimer, M., Ebel, A., Binkowski, F. S., and Shankar, U.: Modal
 9 aerosol dynamics model for Europe: development and first applications, *Atmospheric Environment*,
 10 32, 2981-2999, [http://dx.doi.org/10.1016/S1352-2310\(98\)00006-5](http://dx.doi.org/10.1016/S1352-2310(98)00006-5), 1998.
- 11 Amann, M., Bertok, I., Borcken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z.,
 12 Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W.: Cost-
 13 effective control of air quality and greenhouse gases in Europe: Modeling and policy applications,
 14 *Environmental Modelling and Software*, 26, 1489-1501, 2011.
- 15 Amann, M., Borcken-Kleefeld, J., Cofala, J., Heyes, C., Klimont, Z., Rafaj, P., Purohit, P., Schoepp, W.,
 16 and Winiwarter, W.: Future emissions of air pollutants in Europe - Current legislation baseline and
 17 the scope for further reductions., IIASA, Laxenburg, Austria, 2012.
- 18 Andersson, C., Langner, J., and Bergstrom, R.: Interannual variation and trends in air pollution over
 19 Europe due to climate variability during 1958 – 2001 simulated with a regional CTM coupled to the
 20 ERA-40 reanalysis, *Tellus B*, 59, 77-98, 2007.

1 Balsamo, G., Viterbo, P., Beljaars, A., van den Hurk, B. J. J. M., Hirschi, M., Betts, A., and Scipal, K.: A
2 revised hydrology for the ECMWF model: Verification from field site to terrestrial water storage and
3 impact in the Integrated Forecast System, *J. Hydrometeor.*, 623-643, doi:10.1175/2008JHM1068.1,
4 2009.

5 Banzhaf, S., Schaap, M., Kerschbaumer, A., Reimer, E., Stern, R., van der Swaluw, E., and Bultjes, P. J.
6 H.: Implementation and evaluation of pH-dependent cloud chemistry and wet deposition in the
7 chemical transport model REM-Calgrid, *Atmos. Environ.*, 49, 2012.

8 Banzhaf, S., Schaap, M., Kranenburg, R., Manders, A. M. M., Segers, A. J., Visschedijk, A. J. H., Denier
9 van der Gon, H. A. C., Kuenen, J. J. P., van Meijgaard, E., van Ulft, L. H., Cofala, J., and Bultjes, P. J. H.:
10 Dynamic model evaluation for secondary inorganic aerosol and its precursors over Europe between
11 1990 and 2009, *Geosci. Model Dev.*, 8, 1047-1070, 10.5194/gmd-8-1047-2015, 2015.

12 Beltman, J. B., Hendriks, C., Tum, M., and Schaap, M.: The impact of large scale biomass production
13 on ozone air pollution in Europe, *Atmos. Environ.*, 71, 352-363, 2013.

14 Bergström, R., Denier van der Gon, H. A. C., Prévôt, A. S. H., Yttri, K. E., and Simpson, D.: Modelling of
15 organic aerosols over Europe (2002–2007) using a volatility basis set (VBS) framework: application of
16 different assumptions regarding the formation of secondary organic aerosol, *Atmos. Chem. Phys.*, 12,
17 8499-8527, doi:10.5194/acp-12-8499-2012, 2012.

18 Bessagnet, B., Pirovano, G., Mircea, M., Cuvelier, C., Aulinger, A., Calori, G., Ciarelli, G., Manders, A.,
19 Stern, R., Tsyro, S., García Vivanco, M., Thunis, P., Pay, M. T., Colette, A., Couvidat, F., Meleux, F.,
20 Rouil, L., Ung, A., Aksoyoglu, S., Baldasano, J. M., Bieser, J., Briganti, G., Cappelletti, A., D'Isodoro, M.,
21 Finardi, S., Kranenburg, R., Silibello, C., Carnevale, C., Aas, W., Dupont, J. C., Fagerli, H., Gonzalez, L.,
22 Menut, L., Prévôt, A. S. H., Roberts, P., and White, L.: Presentation of the EURODELTA III inter-
23 comparison exercise – Evaluation of the chemistry transport models performance on criteria
24 pollutants and joint analysis with meteorology, *Atmos. Chem. Phys. Discuss.*, 2016, 1-61,
25 10.5194/acp-2015-736, 2016.

26 Binkowski, F., and Shankar, U.: The Regional Particulate Matter Model .1. Model description and
27 preliminary results, *J. Geophys. Res.*, 100, 26191–26209, 1995.

28 Binkowski, F. S.: The aerosol portion of Models-3 CMAQ. In Science Algorithms of the EPA Models-3
29 Community Multiscale Air Quality (CMAQ) Modeling System. Part II: Chapters 9-18, National
30 Exposure Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC,
31 1999.

32 Bott, A.: A Positive Definite Advection Scheme Obtained by Nonlinear Renormalization of the
33 Advective Fluxes, *Mon. Wea. Rev.*, 117, 1006-1015, 1989.

34 Byun, D. W., and Schere, K. L.: Review of the governing equations, computational algorithms, and
35 other components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System,
36 *Appl. Mech. Rev.*, 59, 51-77, 2006.

37 Carlton, A. G., Bhawe, P. V., Napelenok, S. L., Edney, E. O., Sarwar, G., Pinder, R. W., Pouliot, G. A., and
38 Houyoux, M.: Model representation of secondary organic aerosol in CMAQv4.7, *Environ. Sci.*
39 *Technol.*, 44, 8553-8560, 2010.

40 Implementation of the SAPRC-99 Chemical Mechanism into the Models-3 Framework:
41 <http://www.cert.ucr.edu/~carter/absts.htm#s99mod3>.

42 Carter, W. P. L.: Condensed atmospheric photooxidation mechanisms for isoprene, *Atmospheric*
43 *Environment*, 30, 4275-4290, [http://dx.doi.org/10.1016/1352-2310\(96\)00088-X](http://dx.doi.org/10.1016/1352-2310(96)00088-X), 1996.

44 Carter, W. P. L.: Documentation of the SAPRC-99 Chemical Mechanism for VOC Reactivity
45 Assessment, 2000.

46 Chang, J. S., Brost, R. A., Isaksen, I. S. A., Madronich, S., Middleton, P., Stockwell, W. R., and Walcek,
47 C. J.: A Three-dimensional Eulerian Acid Deposition Model: Physical Concepts and Formulation, *J.*
48 *Geophys. Res.*, 92, 14,681-614,700, 1987.

49 Clough, S. A., Shephard, M. W., Mlawer, E. J., Delamere, J. S., Iacono, M. J., Cady-Pereira, K.,
50 Boukabara, S., and Brown, P. D.: Atmospheric radiative transfer modeling: A summary of the AER
51 codes, *J. Quant. Spectrosc. Radiat. Transfer*, 91, 233–244, 2005.

1 Colette, A., Granier, C., Hodnebrog, O., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B., D'Angiola, A.,
2 D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouil, L., Russo, F., Solberg, S.,
3 Stordal, F., and Tampieri, F.: Air quality trends in Europe over the past decade: a first multi-model
4 assessment, *Atmos. Chem. Phys.*, **11**, 11657-11678, 2011.

5 Colette, A., Aas, W., Banin, L., Braban, C. F., Ferm, M., González Ortiz, A., Ilyin, I., Mar, K., Pandolfi,
6 M., Putaud, J.-P., Shatalov, V., Solberg, S., Spindler, G., Tarasova, O., Vana, M., Adani, M., Almodovar,
7 P., Berton, E., Bessagnet, B., Bohlin-Nizzetto, P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A.,
8 Cuvelier, K., Derwent, R., D'Isidoro, M., Fagerli, H., Funk, C., Garcia Vivanco, M., Haeuber, R., Hueglin,
9 C., Jenkins, S., Kerr, J., de Leeuw, F., Lynch, J., Manders, A., Mircea, M., Pay, M. T., Pritula, D., Querol,
10 X., Raffort, V., Reiss, I., Roustan, Y., Sauvage, S., Scavo, K., Simpson, D., Smith, R. I., Tang, Y. S.,
11 Theobald, M., Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M., and Wind, P.: Air pollution
12 trends in the EMEP region between 1990 and 2012, NILU, Oslo, 2016.

13 Couvidat, F., Debry, E., Sartelet, K., and Seigneur, C.: A Hydrophilic/Hydrophobic Organic (H²O)
14 model: Model development, evaluation and sensitivity analysis, *J. Geophys. Res.*, **117**, 304,
15 doi:10.1029/2011JD017214, 2012.

16 Couvidat, F., and Sartelet, K.: The Secondary Organic Aerosol Processor (SOAP) model: a unified
17 model with different ranges of complexity based on the molecular surrogate approach, *Geosci.*
18 *Model Dev.*, **8**, 1111-1138, 2015.

19 Cuxart J., Bougeault P., and J.-L., R.: A turbulence scheme allowing for mesoscale and large-eddy
20 simulations, *Q. J. R. Meteorol. Soc.*, **126**, 1-30, doi: 10.1002/qj.49712656202, 2000.

21 Dahlgren, P., Landelius, T., Kållberg, P., and Gollvik, S.: A high-resolution regional reanalysis for
22 Europe. Part 1: Three-dimensional reanalysis with the regional High-Resolution Limited-Area Model
23 (HIRLAM), *Quarterly Journal of the Royal Meteorological Society*, **142**, 2119-2131, 10.1002/qj.2807,
24 2016.

25 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda,
26 M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N.,
27 Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V.,
28 Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J. J.,
29 Park, B. K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J. N., and Vitart, F.: The ERA-Interim
30 reanalysis: configuration and performance of the data assimilation system, *Quarterly Journal of the*
31 *Royal Meteorological Society*, **137**, 553-597, 2011.

32 Denier van der Gon, H., Jozwicka, M., Hendriks, E., Gondwe, M., and Schaap, M.: Mineral dust as a
33 component of particulate matter Delft, The Netherlands, 2010.

34 EC: Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient
35 air quality and cleaner air for Europe, European Commission, Brussels, 2008.

36 Emberson, L. D., Ashmore, M. R., Simpson, D., Tuovinen, J.-P., and Cambridge, H. M.: Towards a
37 model of ozone deposition and stomatal uptake over Europe, Norwegian Meteorological Institute,
38 Oslo, Norway, 57, 2000a.

39 Emberson, L. D., Ashmore, M. R., Simpson, D., Tuovinen, J.-P., and Cambridge, H. M.: Modelling
40 stomatal ozone flux across Europe, *Water, Air and Soil Pollution*, **109**, 403-413, 2000b.

41 EMEP: Transboundary acidification, eutrophication and ground level ozone in Europe. Part I: Unified
42 EMEP model description, EMEP, Oslo, Norway, 2003.

43 Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D., Granier, C.,
44 Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S.
45 L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers,
46 version 4 (MOZART-4), *Geosci. Model Dev.*, **3**, 43-67, 2010.

47 Erisman, J. W., Van Pul, A., and Wyers, P.: Parametrization of surface resistance for the quantification
48 of atmospheric deposition of acidifying pollutants and ozone, *Atmospheric Environment*, **28**, 2595-
49 2607, [http://dx.doi.org/10.1016/1352-2310\(94\)90433-2](http://dx.doi.org/10.1016/1352-2310(94)90433-2), 1994.

50 Eyring, V.: Report on the IGAC/SPARC Chemistry-Climate Model Initiative (CCMI) 2013 Science
51 Workshop, 2014.

52 Galmarini, S., Rao, S. T., and Steyn, D. G.: Preface, *Atmospheric Environment*, **53**, 1-3, 2012.

1 Galmarini, S., Koffi, B., Solazzo, E., Keating, T., Hogrefe, C., Schulz, M., Benedictow, A., Griesfeller, J. J.,
2 Janssens-Maenhout, G., Carmichael, G., Fu, J., and Dentener, F.: Technical note: Coordination and
3 harmonization of the multi-scale, multi-model activities HTAP2, AQMEII3, and MICS-Asia3:
4 simulations, emission inventories, boundary conditions, and model output formats, *Atmos. Chem.*
5 *Phys.*, 17, 1543-1555, 10.5194/acp-17-1543-2017, 2017.

6 Gollvik, S., and Samuelsson, P.: A tiled land-surface scheme for HIRLAM, SMHI 2010.

7 Gong, S., Barrie, L., and Blanchet, J.: Modelling sea-salt aerosols in the atmosphere .1. Model
8 development, *J. Geophys. Res.*, 102, 3805–3818, doi:10.1029/96JD02953, 1997.

9 Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G., Heil, A., Kaiser, J.,
10 Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F. o., Liousse, C., Masui, T., Meleux, F., Mieville, A.,
11 Ohara, T., Raut, J.-C., Riahi, K., Schultz, M., Smith, S., Thompson, A., van Aardenne, J., van der Werf,
12 G., and van Vuuren, D.: Evolution of anthropogenic and biomass burning emissions of air pollutants
13 at global and regional scales during the 1980-2010 period, *Climatic Change*, 109, 163-190, 2011.

14 Grell, G. A., and Dévényi, D.: A generalized approach to parameterizing convection combining
15 ensemble and data assimilation techniques, *Geophysical Research Letters*, 29, 38-31-38-34,
16 10.1029/2002GL015311, 2002.

17 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully
18 coupled “online” chemistry within the WRF model, *Atmospheric Environment*, 39, 6957-6975,
19 <http://dx.doi.org/10.1016/j.atmosenv.2005.04.027>, 2005.

20 Guenther, A., Zimmerman, P., Harley, P., Monson, R., and Fall, R.: Isoprene and monoterpene rate
21 variability: model evaluations and sensitivity analyses, *J. Geophys. Res.*, 98, 12609–12617, 1993.

22 Guenther, A., Zimmerman, P., and Wildermuth, M.: Natural volatile organic compound emission
23 rate estimates for U.S. woodland landscapes, *Atmos. Environ.*, 28, 1197–1210, 1994.

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

27 Hicks, B. B., Artz, R. S., Meyers, T. P., and Hosker, R. P.: Trends in eastern U.S. sulfur air quality from
28 the Atmospheric Integrated Research Monitoring Network, *Journal of Geophysical Research:*
29 *Atmospheres*, 107, ACH 6-1-ACH 6-12, 10.1029/2000JD000165, 2002a.

30 Hicks, B. B., Artz, R. S., Meyers, T. P., and Hosker, R. P.: Trends in eastern US sulfur air quality from
31 the Atmospheric Integrated Research Monitoring Network, *Journal of Geophysical Research:*
32 *Atmospheres*, 107, 2002b.

33 Hong, S.-Y., Dudhia, J., and Chen, S.-H.: A revised approach to ice microphysical processes for the bulk
34 parameterization of clouds and precipitation, *Monthly Weather Review*, 132, 103-120, 2004.

35 Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins, W. D.:
36 Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer
37 models, *J. Geophys. Res.*, 113, doi: 10.1029/2008JD009944, 2008.

38 IEA: Energy Statistics of OECD Countries, International Energy Agency Paris, France, 2012.

39 Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baró, R., Bellasio, R., Brunner,
40 D., and Chemel, C.: Evaluation of operational on-line-coupled regional air quality models over Europe
41 and North America in the context of AQMEII phase 2. Part I: Ozone, *Atmospheric Environment*, 115,
42 404-420, 2015.

43 Jacob, D., Petersen, J., Eggert, B., Alias, A., Christensen, O., Bouwer, L., Braun, A., Colette, A., Déqué,
44 M., Georgievski, G., Georgopoulou, E., Gobiet, A., Menut, L., Nikulin, G., Haensler, A., Hempelmann,
45 N., Jones, C., Keuler, K., Kovats, S., Kraner, N., Kotlarski, S., Kriegsmann, A., Martin, E., Meijgaard, E.,
46 Moseley, C., Pfeifer, S., Preuschmann, S., Radermacher, C., Radtke, K., Rechid, D., Rounsevell, M.,
47 Samuelsson, P., Somot, S., Soussana, J.-F., Teichmann, C., Valentini, R., Vautard, R., Weber, B., and
48 Yiou, P.: EURO-CORDEX: new high-resolution climate change projections for European impact
49 research, *Regional Environmental Change*, 1-16, 2013.

50 Janjic, Z. I.: Nonsingular implementation of the Mellor-Yamada Level 2.5 Scheme in the NCEP Meso
51 model, National Centers for Environmental Prediction, College Park, MD, 61, 2002.

1 Jeričević, A., Kraljević, L., Grisogono, B., Fagerli, H., and Večenaj, Ž.: Parameterisation of vertical
2 diffusion and the atmospheric boundary layer height determination in the EMEP model, *Atmos.*
3 *Chem. Phys.*, 10, 341-364, doi:10.5194/acp-10-341-2010, 2010.

4 Jonson, J. E., Simpson, D., Fagerli, H., and Solberg, S.: Can we explain the trends in European ozone
5 levels?, *Atmos. Chem. Phys.*, 6, 51-66, 2006.

6 Kain, J. S.: The Kain–Fritsch Convective Parameterization: An Update, *Journal of Applied*
7 *Meteorology*, 43, 170-181, doi:10.1175/1520-0450(2004)043<0170:TKCPAU>2.0.CO;2, 2004.

8 Kelly, J. T., Bhave, P. V., Nolte, C. G., Shankar, U., and Foley, K. M.: Simulating emission and chemical
9 evolution of coarse sea-salt particles in the Community Multiscale Air Quality (CMAQ) model, *Geosci.*
10 *Model Dev.*, 3, 257-273, 2010.

11 Klimont, Z., Hoeglund-Isaksson, L., Heyes, C., Rafaj, P., Schoepp, W., Cofala, J., Purohit, P., Borken-
12 Kleefeld, J., Kupiainen, K., Kiesewetter, G., Winiwarter, W., Amann, M., Zhao, B., Wang, S., Bertok, I.,
13 and Sander, R.: Global scenarios of air pollutants and methane: 1990-2050, in prep., 2016a.

14 Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., and Schoepp, W.: Global
15 anthropogenic emissions of particulate matter, in preparation, 2016b.

16 Koeble, R., and Seufert, G.: Novel Maps for Forest Tree Species in Europe, *A Changing Atmosphere*,
17 8th European Symposium on the Physico-Chemical Behaviour of Atmospheric Pollutants, Torino,
18 Italy, 2001.

19 Köhler, I., Sausen, R., and Klenner, G.: NO_x production from lightning, The impact of NO_x emissions
20 from aircraft upon the atmosphere at flight altitudes 8–15 km (AERONOX), , *Deutsch Luft und*
21 *Raumfahrt, Oberpfaffenhofen, Germany*, 1995.

22 Kotlarski, S., Keuler, K., Christensen, O. B., Colette, A., Déqué, M., Gobiet, A., Goergen, K., Jacob, D.,
23 Lüthi, D., van Meijgaard, E., Nikulin, G., Schär, C., Teichmann, C., Vautard, R., Warrach-Sagi, K., and
24 Wulfmeyer, V.: Regional climate modeling on European scales: a joint standard evaluation of the
25 EURO-CORDEX RCM ensemble, *Geosci. Model Dev.*, 7, 1297-1333, 2014.

26 Lamarque, J. F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E.
27 A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAM-chem: description and
28 evaluation of interactive atmospheric chemistry in the Community Earth System Model, *Geosci.*
29 *Model Dev.*, 5, 369-411, 2012.

30 Lange, R.: Transferrability of a three-dimensional air quality model between two different sites in
31 complex terrain, *J. Appl. Meteorol.*, 78, 665-679, 1989.

32 Langner, J., Bergström, R., and Pleijel, K.: European scale modeling of sulphur, oxidized nitrogen and
33 photochemical oxidants. Model development and evaluation for the 1994 growing season, *Swedish*
34 *Met. and Hydrol. Inst., Norrköping, Sweden*, 1998.

35 Lenderink, G., and Holtslag, A. A. M.: An updated length-scale formulation for turbulent mixing in
36 clear and cloudy boundary layers., *Q.J.R. Meteorol. Soc.*, 3405–3427, doi: 10.1256/qj.03.117, 2004.

37 Logan, J. A.: An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D
38 models and development of a gridded climatology for tropospheric ozone, *J. Geophys. Res.*, 10,
39 16115–16149, 1998.

40 Maas, R., and Grennfelt, P.: Towards Cleaner Air - Scientific Assessment Report 2016, EMEP-Steering
41 body and Working Group on Effects - Convention on Long-Range Transboundary Air Pollution 2016.

42 Mallet, V., Quélo, D., Sportisse, B., Ahmed de Biasi, M., Debry, É., Korsakissok, I., Wu, L., Roustan, Y.,
43 Sartelet, K., Tombette, M., and Foudhil, H.: Technical Note: The air quality modeling system
44 Polyphemus, *Atmos. Chem. Phys.*, 7, 5479-5487, doi:10.5194/acp-7-5479-2007, 2007.

45 Manders, A. M. M., Builtjes, P. J. H., Curier, L., Denier van der Gon, H. A. C., Hendriks, C., Jonkers, S.,
46 Kranenburg, R., Kuenen, J., Segers, A. J., Timmermans, R. M. A., Visschedijk, A., Wichink Kruit, R. J.,
47 Van Pul, W. A. J., Sauter, F. J., van der Swaluw, E., Swart, D. P. J., Douros, J., Eskes, H., van Meijgaard,
48 E., van Ulft, B., van Velthoven, P., Banzhaf, S., Mues, A., Stern, R., Fu, G., Lu, S., Heemink, A., van
49 Velzen, N., and Schaap, M.: Curriculum Vitae of the LOTOS-EUROS (v2.0) chemistry transport model,
50 *Geosci. Model Dev. Discuss.*, 2017, 1-53, 10.5194/gmd-2017-88, 2017.

1 Mar, K. A., Ojha, N., Pozzer, A., and Butler, T. M.: Ozone air quality simulations with WRF-Chem
2 (v3.5.1) over Europe: model evaluation and chemical mechanism comparison, *Geosci. Model Dev.*, 9,
3 3699-3728, 10.5194/gmd-9-3699-2016, 2016.

4 Martensson, E., Nilsson, E., de Leeuw, G., Cohen, L., and Hansson, H.-C.: Laboratory simulations and
5 parameterisation of the primary marine aerosol production, *J. Geophys. Res.*, 108, 4297,
6 doi:10.1029/2002JD002263, 2003.

7 Meijgaard, E. v., van Ulft, L. H., Lenderink, G., de Roode, S. R., Wipfler, L., Boers, R., and Timmermans,
8 R. M. A.: Refinement and application of a regional atmospheric model for climate scenario
9 calculations of Western Europe, *KvR 054/12*, 44, 2012.

10 Menut, L., Bessagnet, B., Khvorostyanov, D., Beekmann, M., Blond, N., Colette, A., Coll, I., Curci, G.,
11 Foret, G., Hodzic, A., Mailler, S., Meleux, F., Monge, J. L., Pison, I., Siour, G., Turquety, S., Valari, M.,
12 Vautard, R., and Vivanco, M. G.: CHIMERE 2013: a model for regional atmospheric composition
13 modelling, *Geosci. Model Dev.*, 6, 981-1028, 2013.

14 Mircea, M., Grigoras, G., D'Isidoro, M., Righini, G., Adani, M., Briganti, G., Ciancarella, L., Cappelletti,
15 A., Calori, G., Cionni, I., Cremona, G., Finardi, S., Larsen, B. R., Pace, G., Perrino, C., Piersanti, A.,
16 Silibello, C., Vitali, L., and Zanini, G.: Impact of grid resolution on aerosol predictions: a case study
17 over Italy, *Aerosol and Air Quality Research*, 1253–1267, doi: 10.4209/aaqr.2015.02.0058, 2016.

18 Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for
19 inhomogeneous atmospheres : RRTM, a validated correlated-k model for the longwave, *J. Geophys.*
20 *Res.*, 16663-16682, 1997.

21 Monahan, E. C.: The ocean as a source of atmospheric particles, in: *The Role of Air-Sea Exchange in*
22 *Geochemical Cycling*, Kluwer Academic Publishers, Dordrecht, Holland, 129–163, 1986.

23 Morcrette, J.-J., Mlawer, E. J., Iacono, M. J., and Clough, S. A.: Impact of the radiation transfer
24 scheme RRTM in the ECMWF forecasting system, ECMWF, Reading, UK, 2–9, 2001.

25 Morcrette, J.-J., Barker, H. W., Cole, J. N. S., Iacono, M. J., and Pincus, R.: Impact of a New Radiation
26 Package, McRad, in the ECMWF Integrated Forecasting System, *Mon. Wea. Rev.*, 4773–4798,
27 dx.doi.org/10.1175/2008MWR2363.1 2008.

28 Morrison, H., Thompson, G., and Tatarskii, V.: Impact of Cloud Microphysics on the Development of
29 Trailing Stratiform Precipitation in a Simulated Squall Line: Comparison of One- and Two-Moment
30 Schemes, *Mon. Wea. Rev.*, 137, 991–1007, 2009.

31 Mozurkewich, M.: The dissociation constant of ammonium nitrate and its dependence on
32 temperature, relative humidity and particle size, *Atmospheric Environment. Part A. General Topics*,
33 27, 261-270, [http://dx.doi.org/10.1016/0960-1686\(93\)90356-4](http://dx.doi.org/10.1016/0960-1686(93)90356-4), 1993.

34 Nakanishi, M., and Niino, H.: An improved Mellor–Yamada level 3 model: its numerical stability and
35 application to a regional prediction of advecting fog, *Bound. Layer Meteor.*, 119, 397–407, 2006.

36 Nakanishi, M., and Niino, H.: Development of an improved turbulence closure model for the
37 atmospheric boundary layer, *J. Meteor. Soc. Japan*, 87, 895–912, 2009.

38 Neggers, R. A. J.: A dual mass flux framework for boundary layer convection. Part II: Clouds. , *J.*
39 *Atmos. Sci.*, 1489-1506, doi:10.1175/2008JAS2636.1, 2009.

40 Neggers, R. A. J., Koehler, M., and Beljaars, A. C. M.: A dual mass flux framework for boundary layer
41 convection. Part I: Transport. , *J. Atmos. Sci.*, 1465-1487, doi:10.1175/2008JAS2635.1, 2009.

42 Nenes, A., Pilinis, C., and Pandis, S. N.: ISORROPIA: a new thermodynamic equilibrium model for
43 multiphase multicomponent marine aerosols., *Aquat. Geochem.*, 4, 123-152, 1998.

44 Nenes, A., Pilinis, C., and Pandis, S. N.: Continued development and testing of a new thermodynamic
45 aerosol module for urban and regional air quality models, *Atmos. Environ.*, 33, 1553–1560, 1999.

46 Noilhan, J., and Planton, S.: A Simple Parameterization of Land Surface Processes for Meteorological
47 Models, *Monthly Weather Review*, 117, 536-549, 10.1175/1520-
48 0493(1989)117<0536:ASPOLS>2.0.CO;2, 1989.

49 Noilhan J., and J.-F., M.: The ISBA land surface parameterization scheme, *Global Planet Change*, 145–
50 159, 1996.

51 Nordeng, T.-E.: Extended versions of the convection parameterization scheme at ECMWF and their
52 impact upon the mean climate and transient activity of the model in the tropics, ECMWF, 1994.

1 Ntziachristos, L., Gkatzoflias, D., Kouridis, C., and Samaras, Z.: COPERT: A European Road Transport
2 Emission Inventory Model Information Technologies in Environmental Engineering 4th International
3 ICSC Symposium, Thessaloniki, Greece, 2009, 491-504,
4 O'Dowd, C. D., Smith, M. H., Consterdine, I. E., and Lowe, J. A.: Marine aerosol, sea-salt, and the
5 marine sulphur cycle: a short review, *Atmospheric Environment*, 31, 73-80,
6 [http://dx.doi.org/10.1016/S1352-2310\(96\)00106-9](http://dx.doi.org/10.1016/S1352-2310(96)00106-9), 1997.
7 O'Brien, J. J.: A note on the vertical structure of the eddy exchange coefficient in the planetary
8 boundary layer, *J Atmos Sci*, 27, 1213–1215, 1970.
9 OECD: The OECD programme on long range transport of air pollutants. Measurements and findings,
10 Paris, 1977.
11 Pleim, J.: A combined local and nonlocal closure model for the atmospheric boundary layer. Part I:
12 model description and testing. , *J. Appl. Met. Climatology*, 46, 1383-1395, 2007.
13 Rao, S. T., Galmarini, S., and Puckett, K.: Air Quality Model Evaluation International Initiative
14 (AQMEII): advancing the state of the science in regional photochemical modeling and its applications,
15 *Bulletin of the American Meteorological Society*, 92, 23-30, 2011.
16 Rasch, P. J., and Kristjánsson, J. E.: A Comparison of the CCM3 Model Climate Using Diagnosed and
17 Predicted Condensate Parameterizations, *Journal of Climate*, 11, 1587-1614, doi:10.1175/1520-
18 0442(1998)011<1587:ACOTCM>2.0.CO;2, 1998.
19 Reis, S., Grennfelt, P., Klimont, Z., Amann, M., ApSimon, H., Hettelingh, J.-P., Holland, M., LeGall, A.-
20 C., Maas, R., Posch, M., Spranger, T., Sutton, M. A., and Williams, M.: From Acid Rain to Climate
21 Change, *Science*, 338, 1153-1154, 2012.
22 Robertson, L., Langner, J., and Engardt, M.: An Eulerian Limited-Area Atmospheric Transport Model,
23 *Journal of Applied Meteorology*, 38, 190-210, 1999.
24 Savijärvi, H.: Fast Radiation Parameterization Schemes for Mesoscale and Short-Range Forecast
25 Models, *Journal of Applied Meteorology*, 29, 437-447, doi:10.1175/1520-
26 0450(1990)029<0437:FRPSFM>2.0.CO;2, 1990.
27 Schaap, M., Timmermans, R. M. A., Roemer, M., Boersen, G. A. C., Bultjes, P., Sauter, F., Velders, G.,
28 and Beck, J.: The LOTOS-EUROS model: description, validation and latest developments, *International*
29 *Journal of Environment and Pollution*, 32, 270-290, 2008.
30 Schaap, M., Manders, A. M. M., Hendriks, E. C. J., Cnossen, J. M., Segers, A. J. S., Denier van der Gon,
31 H., Jozwicka, M., Sauter, F. J., Velders, G. J. M., Matthijsen, J., and Bultjes, P. J. H.: Regional Modelling
32 of Particulate Matter for the Netherlands Netherlands Research Program on Particulate Matter,
33 2009.
34 Schell, B., Ackermann, I. J., Hass, H., Binkowski, F. S., and Ebel, A.: Modelling the formation of
35 secondary organic within a comprehensive air quality model system, *J. Geophys. Res.*, 106, 28275-
36 28293, 2001.
37 Seinfeld, J. H., and Pandis, S. N.: *Atmospheric Chemistry and Physics, From Air Pollution to Climate*
38 *Change.*, edited by: John Wiley and Sons, I., New York, USA., 1998.
39 Siebesma, A. P., Soares, P. M. M., and Teixeira, J.: A Combined Eddy-Diffusivity Mass-Flux Approach
40 for the Convective Boundary Layer, *J. Atmos. Sci.*, 1230-1248, doi:10.1175/JAS3888.1, 2007.
41 Simpson, D., Guenther, A., Hewitt, C., and Steinbrecher, R.: Biogenic emissions in Europe 1. Estimates
42 and uncertainties, *J. Geophys. Res.*, 100, 22875–22890, 1995.
43 Simpson, D., Benedictow, A., Berge, H., Bergstrom, R., Emberson, L. D., Fagerli, H., Flechard, C. R.,
44 Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyiri, A., Richter, C., Semeena, V. S., Tsyro, S.,
45 Tuovinen, J. P., Valdebenito, A., and Wind, P.: The EMEP MSC-W chemical transport model - technical
46 description, *Atmos. Chem. Phys.*, 12, 7825-7865, 2012a.
47 Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Hayman, G. D.,
48 Gauss, M., Jonson, J. E., Jenkin, M. E., Nyiri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P.,
49 Valdebenito, A., and Wind, P.: The EMEP MSC-W chemical transport model -- Part 1: Model
50 description, *Atmospheric Chemistry and Physics Discussions*, 12, 2012b.
51 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang,
52 W., and Powers, J. G.: A Description of the Advanced Research WRF Version 3, NCAR, 2008.

1 Slinn, W. G. N.: Precipitation scavenging, US. Department of Energy, Washington, D.C., 1983.
2 Sofiev, M., Soares, J., Prank, M., de Leeuw, G., and Kukkonen, J.: A regional-to-global model of
3 emission and transport of sea salt particles in the atmosphere, *J. Geophys. Res.*, 116,
4 doi:10.1029/2010JD014713, 2011.
5 Spee, E. J.: Numerical methods in global transport-chemistry models, PhD, Amsterdam, 1998.
6 Sportisse, B., and Dubois, L.: Numerical and theoretical investigation of a simplified model for the
7 parameterization of below-cloud scavenging by falling raindrops, *Atmos. Environ.*, 36, 5719-5727,
8 2002.
9 Stegehuis, A. I., Vautard, R., Ciais, P., Teuling, A. J., Miralles, D. G., and Wild, M.: An observation-
10 constrained multi-physics WRF ensemble for simulating European mega heat waves, *Geosci. Model*
11 *Dev.*, 8, 2285-2298, 10.5194/gmd-8-2285-2015, 2015.
12 Stein, U., and Alpert, P.: Factor separation in numerical simulations, *Journal of the Atmospheric*
13 *Sciences*, 50, 2107-2115, 1993.
14 Stockwell, W. R., Middleton, P., Chang, J. S., and Tang, X.: The second generation regional acid
15 deposition model chemical mechanism for regional air quality modeling, *Journal of Geophysical*
16 *Research: Atmospheres*, 95, 16343-16367, 10.1029/JD095iD10p16343, 1990.
17 Strand, A., and Hov, O.: A two-dimensional global study of tropospheric ozone production, *J Geophys*
18 *Res* 99, 22877-22895, 1994.
19 Terrenoire, E., Bessagnet, B., Rouil, L., Tognet, F., Pirovano, G., Létinois, L., Beauchamp, M., Colette,
20 A., Thunis, P., Amann, M., and Menut, L.: High-resolution air quality simulation over Europe with the
21 chemistry transport model CHIMERE, *Geosci. Model Dev.*, 8, 21-42, 2015.
22 Tewari, M., Chen, F., Wang, W., Dudhia, J., LeMone, M. A., Mitchell, K., Ek, M., Gayno, G., Wegiel, J.,
23 and Cuenca, R. H.: Implementation and verification of the unified NOAA land surface model in the
24 WRF model, 20th conference on weather analysis and forecasting/16th conference on numerical
25 weather prediction, Seattle, WA, 2004.
26 Thunis, P., Cuvelier, C., Roberts, P., White, L., Post, L., Tarrason, L., Tsyro, S., Stern, R., Kerschbaumer,
27 A., Rouil, L., Bessagnet, B., Builtjes, J., Schaap, M., Boersen, G., and Bergstroem, R.: Evaluation of a
28 Sectoral Approach to Integrated Assessment Modelling including the Mediterranean Sea, JRC, Ispra,
29 Italy, 2008.
30 Tiedtke, M.: A comprehensive mass flux scheme for cumulus parameterization in large-scale models,
31 11, 1989.
32 Tiedtke, M.: Representation of clouds in large-scale models. , *Mon. Wea. Rev.*, 3040-3061, 1993.
33 Tilm, A., and Lenderink, G.: Characteristics of CBR and STRACO versions, 2003.
34 Tilmes, S., Lamarque, J. F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith, A. K., Neely,
35 R. R., Conley, A., Vitt, F., Val Martin, M., Tanimoto, H., Simpson, I., Blake, D. R., and Blake, N.:
36 Representation of the Community Earth System Model (CESM1) CAM4-chem within the Chemistry-
37 Climate Model Initiative (CCMI), *Geosci. Model Dev.*, 9, 1853-1890, 10.5194/gmd-9-1853-2016, 2016.
38 Tompkins, A. M., Gierens, K., and Rädcl, G.: Ice supersaturation in the ECMWF Integrated Forecast
39 System, *Q.J.R. Meteorol. Soc.*, 133, 53-63, 2007.
40 Troen, I., and Mahrt, L.: A simple model of the atmospheric boundary layer: Sensitivity to surface
41 evaporation, *Bound.-Layer Meteorol.*, 37, 129-148, 1986.
42 Tsyro, S., Aas, W., Soares, J., Sofiev, M., Berge, H., and Spindler, G.: Modelling of sea salt
43 concentrations over Europe: key uncertainties and comparison with observations, *Atmos. Chem.*
44 *Phys.*, 11, 10367–10388, doi:10.5194/acp-11-10367-2011, 2011.
45 Tuovinen, J.-P., Ashmore, M., Emberson, L., and Simpson, D.: Testing and improving the EMEP ozone
46 deposition module, *Atmos. Environ.*, 38, 2373–2385, 2004.
47 UNECE: Protocol to the 1979 Convention on Long-Range Tansboundary Air Pollution to abate
48 acidification, eutrophication and ground-level ozone, 1999.
49 Van den Hurk, B. J. J. M., Viterbo, P., Beljaars, A. C. M., and Betts, A. K.: Offline validation of the
50 ERA40 surface scheme, ECMWF, 2000.

1 van Leer, B.: Multidimensional explicit difference schemes for hyperbolic conservation laws, in:
2 Computing Methods in Applied Sciences and Engineering VI, edited by: Lions, R. G. a. J. L., Elsevier,
3 Amsterdam, 1984.

4 van Loon, M., Vautard, R., Schaap, M., Bergström, R., Bessagnet, B., Brandt, J., Builtjes, P. J. H.,
5 Christensen, J. H., Cuvelier, C., Graff, A., Jonson, J. E., Krol, M., Langner, J., Roberts, P., Rouil, L., Stern,
6 R., Tarrasón, L., Thunis, P., Vignati, E., White, L., and Wind, P.: Evaluation of long-term ozone
7 simulations from seven regional air quality models and their ensemble, *Atmospheric Environment*,
8 41, 2083-2097, 2007.

9 Van Zanten, M. C., Sauter, F. J., Wichink Kruit, R. J., Van Jaarsveld, J. A., and Van Pul, W. A. J.:
10 Description of the DEPAC module: Dry deposition modelling with DEPAC_GCN2010, Bilthoven, the
11 Netherlands, 2010.

12 Vautard, R., Bessagnet, B., Chin, M., and Menut, L.: On the contribution of natural Aeolian sources to
13 particulate matter concentrations in Europe: Testing hypotheses with a modelling approach,
14 *Atmospheric Environment*, 39, 3291-3303, 10.1016/j.atmosenv.2005.01.051, 2005.

15 Vautard, R., Szopa, S., Beekmann, M., Menut, L., Hauglustaine, D. A., Rouil, L., and Roemer, M.: Are
16 decadal anthropogenic emission reductions in Europe consistent with surface ozone observations?,
17 *Geophys. Res. Lett.*, 33, L13810, 10.1029/2006GL026080, 2006.

18 Venkatram, A., and Pleim, J.: The electrical analogy does not apply to modelling dry deposition of
19 particles, *Atmos. Environ.*, 33, 3075-3076, 1999.

20 Walcek, C. J., and Taylor, G. R.: A theoretical method for computing vertical distribution of acidity and
21 sulphate production within cumulus clouds, *J. Atmos. Sci.*, 43, 339-355, 1986.

22 Walcek, C. J.: Minor flux adjustment near mixing ratio extremes for simplified yet highly accurate
23 monotonic calculation of tracer advection, *Journal of Geophysical Research*, 105, 9335-9348, 2000.

24 Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale
25 numerical models, *Atmospheric Environment (1967)*, 23, 1293-1304, 1989.

26 Wichink Kruit, R. J., Schaap, M., Sauter, F. J., van der Swaluw, R., E., and Weijers, R.: Improving the
27 understanding of the secondary inorganic aerosol distribution over the Netherlands,, TNO, Utrecht,
28 The Netherlands, 2012.

29 Wilson, R. C., Fleming, Z. L., Monks, P. S., Clain, G., Henne, S., Konovalov, I. B., Szopa, S., and Menut,
30 L.: Have primary emission reduction measures reduced ozone across Europe? An analysis of
31 European rural background ozone trends 1996-2005, *Atmos. Chem. Phys.*, 12, 437-454, 2012.

32 Yamartino, R. J.: Nonnegative, conserved scalar transport using grid-cell-centered, spectrally
33 constrained Blackman cubics for applications on a variable-thickness mesh, *Mon. Wea. Rev.*, 121,
34 753-763, 1993.

35 Yarwood. G., Rao, S., Yocke, M., and Whitten, G. Z.: Updates to the Carbon Bond chemical
36 mechanism: CB05, 2005.

37 Yuan, H., Dai, Y., Xiao, Z., Ji, D., and Shanguan, W.: Reprocessing the MODIS Leaf Area Index
38 Products for Land Surface and Climate Modelling, *Remote Sensing of Environment*, 155, 1171-1187,
39 doi:10.1016/j.rse.2011.01.001, 2011.

40 Zhang, C., Wang, Y., and Hamilton, K.: Improved representation of boundary layer clouds over the
41 southeast pacific in ARW-WRF using a modified Tiedtke cumulus parameterization scheme, *Mon.*
42 *Weather Rev.*, 139, 3489-3513, 2011.

43 Zhang K.M., Knipping E.M., Wexler A.S., Bhave P.V., and Tonnesen, G. S.: Size distribution of sea-salt
44 emissions as a function of relative humidity, *Atm. Env.*, 39, 3373-3379, 2005.

45 Zhang, L., Gong, S., Padro, J., and Barrie, L.: A size-segregated particle dry deposition scheme for an
46 atmospheric aerosol module, *Atmos. Environ.*, 35, 549-560, 2001.

47 Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry deposition in air-
48 quality models, *Atmos. Chem. Phys.*, 3, 2067-2082, 2003.

49

50