An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution


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Abstract

The Danish Eulerian Hemispheric Model (DEHM) is a 3D long-range atmospheric chemistry-transport model with a horizontal domain covering the Northern Hemisphere. For the AQMEII (Air Quality Modelling Evaluation International Initiative) inter-comparison exercise, the model was set up with two two-way nested domains simultaneously – one covering Europe and one covering North America. In this paper, the model configuration used in AQMEII will be described, including a discussion of model results and evaluation for the year 2006 against available measurements in Europe for different chemical species. The evaluation of DEHM for Europe shows that the model gives satisfying results for species such as ozone, nitrogen-dioxide, sulphur-dioxide and secondary inorganic aerosols. The evaluation also points to certain processes where DEHM can be improved, such as biogenic emissions of isoprene, mass closure for particulate matter, wet deposition, and description of vertical mixing during winter. Furthermore, special attention is given to the intercontinental transport of air pollution between North America (NA) and Europe (EU). We estimate the contributions to the total air pollution levels at continental scale from the anthropogenic emissions in the two areas, with a focus on ozone and particulate matter using a tagging method, taking into account the non-linear effects of atmospheric chemistry. We conclude that for this specific year, the intercontinental transport between NA and EU is small for the annual or seasonal mean values – for ozone the contributions are typically around 3% (~1 ppb) from NA to EU and around 1% (~0.3 ppb) from EU to NA. For particles the contributions from NA to EU is around 0.9% (~0.05 μg m⁻³) and from EU to NA around 1.4% (~0.05 μg m⁻³).

1. Introduction

The Danish Eulerian Hemispheric Model (DEHM) is a three-dimensional, offline, large-scale, Eulerian, atmospheric chemistry-transport model (CTM) developed to study long-range transport of air pollution in the Northern Hemisphere. The model domain used in previous studies covers most of the Northern Hemisphere, discretized on a polar stereographic projection, and includes a two-way nesting procedure with several nests with higher resolution over Europe, Northern Europe and Denmark (Frohn et al., 2002).

DEHM was originally developed in the early 1990's to study the atmospheric transport of sulphur and sulphate into the Arctic (Christensen, 1997; Heidam et al., 1999, 2004). The model has been modified, extended and updated several times since then. The original simple sulphur-sulphate chemistry has been replaced by a more comprehensive chemical scheme, including 58 chemical species, 9 primary particles and 122 chemical reactions. In the last decade the model has been expanded to describe atmospheric transport of other atmospheric compounds such as lead (Christensen, 1999; Heidam et al., 2004), atmospheric chemistry and transport of mercury (Christensen et al., 2004; Heidam et al., 2004; Skov et al., 2004), fluxes and atmospheric transport of CO₂ (Geels et al., 2001, 2002; 2004, 2006, 2007), emissions and atmospheric transport of pollen (Skjøth et al., 2007, 2008a; 2008b, 2009; Sikoparija et al., 2009; Smith et al., 2008; Stach et al., 2007), as well as atmospheric transport and environmental fate of persistent organic pollutants (POPs) (Hansen et al., 2004, 2008a, 2008b). The latest development was to include the POP and Hg modules in the chemistry version to allow for the option to simulate concurrently the traditional photo-chemistry and the mercury chemistry.
Together as well as the more inert POPs (McLachlan et al., 2010; Genuard et al., 2011). Furthermore, a simple data assimilation scheme has been coupled to the model based on the optimum interpolation method (Fryendall et al., 2009).

Other important developments include a two-way nesting capability for obtaining higher resolution over limited areas, coupling DEHM to several different emission databases and extending the description of the physical properties in the model such as wet and dry deposition (Frohn et al., 2001, 2002; 2003; Frohn, 2004). The model version used in this study has been validated against measurement data in Europe over a 10-years period (Geels et al., 2005).

The present basic chemistry version of the model, including photo-chemistry and particles has been used in a range of applications. For example, it was applied to study the effect of climate change on future air pollution levels where it was coupled to data from the general circulation model ECHAM5 (Hedegaard et al., 2008, 2011). It has been used for many years as a part of the Danish monitoring programme (NOVANA), where the focus has been on the optimal integration of measurement and modelling data for assessment of, for example, nitrogen loads to eco-systems or human exposure to harmful gases and particles. The coupled approach for combining model results and measurements used within NOVANA is called “integrated monitoring” (Hertel et al., 2007), and is based on the fact that measurements are relatively precise, but have a poor spatial coverage while the model is relatively less accurate but has greater spatial coverage. In the monitoring programme, the model is used to make an intelligent interpolation between the measurement stations and it can be used to explain what is measured.

The DEHM model is furthermore used in the integrated model system, THOR (Brandt et al., 2001a, 2001b, 2001c, 2003, 2007, 2009), including several meteorological and air pollution models capable of operating for different applications and at different scales. In the THOR system, meteorological data, obtained from either the MM5v3.7 model or the EtA model, are used to drive the air pollution models, including the Danish Eulerian Hemispheric Model, DEHM, the Urban Background Model, UBM (Berkowicz, 2000a), and the Operational Street Pollution Model, OSPM (Berkowicz, 2000b). The THOR system runs operationally four times every day, producing three-day forecasts of weather and air pollution from regional scale over urban background scale and down to individual street canyons in cities — on both sides of the streets. The coupling of models over different scales makes it possible to account for contributions from local, near-local and remote emission sources in order to describe the air quality at a specific location — e.g. in a street canyon, in a park or in rural areas. The THOR system also includes the accidental release model, DREAM (Brandt et al., 2002), used to forecast the releases from point sources at European scale.

The DEHM model is also included in the integrated EVA model system (Economic Valuation of air pollution, Frohn et al., 2006, 2007; Brandt et al., 2011) based on the impact pathway chain. The EVA system consists of the regional-scale DEHM, address-level or gridded population data, exposure-response functions for health impacts and economic valuations of the impacts from air pollution. The system was originally developed to evaluate site-specific health costs related to air pollution, such as from specific power plants (Andersen et al., 2007), but was recently extended to assess health cost externalities at the national level for Denmark and Europe (Brandt et al., 2011).

DEHM is furthermore a part of the Danish Ammonia Model System (DAMOS) (Hertel et al., 2006; Geels et al., in review), which includes a coupling between DEHM and a local scale Gaussian plume model OML-DEP (Olesen et al., 1992; Sommer et al., 2009). The system is used to calculate deposition of ammonia with high resolution around emission sources, which is necessary information in relation to regulation of ammonia emissions from the agricultural sector. A key part of the DAMOS system is a dynamical ammonia emission model, estimating the time dependent emissions of ammonia (Skjøth et al., 2004; Gyldenkærne et al., 2005). This emission model is included in the DEHM model (Skjøth et al., 2011).

The various versions of DEHM have participated in several model inter-comparison studies, and the model was shown to perform well compared to other models for the compounds sulphur (Barrie et al., 2001; Lohmann et al., 2001; Roelofs et al., 2001), O3 (Van Loon et al., 2007; Vautard et al., 2006), O3, NO2 and secondary inorganic aerosols (SIA) (Vautard et al., 2009), CO2 (Geels et al., 2007; Law et al., 2008; Fatra et al., 2008; Rivier et al., 2010), Hg (Ryaboshapko et al., 2007) and POPs (Hansen et al., 2006). The model was also included in the EURODELTIA inter-comparison, which focussed on aerosol and gas concentrations in different emission scenarios in Europe (Cuvetilier et al., 2007).

DEHM model results were included in the AQMEII model inter-comparison for North America and Europe, and they form the basis of this study. The purpose of the paper is threefold: (1) to give an extensive description of the DEHM model, (2) to make a thorough evaluation of the model for Europe for the base year 2006, including all available measured species, and (3) to quantify the intercontinental transport of air pollution between North America and Europe for the year 2006. All three aims are relevant to the AQMEII inter-comparison since information about the model and the basic model performance is necessary for interpretation of the inter-comparison results provided in the common papers of this special issue. Furthermore, it is interesting to assess the contributions from the anthropogenic emissions within the two continents to the total air pollution levels in the context of AQMEII.

In section 2 a more detailed description of the DEHM model is given. In section 3 the model setup used in AQMEII is described together with some typical model results for ozone and PM2.5. The model was validated against measurements from the EMEP database for the European domain for the base year of 2006 and the results of this evaluation are also presented in Section 3. Section 4 focuses on the intercontinental transport between North America to Europe using a tagging method (also described in section 4) and finally Section 5 gives the conclusions.

2. The Danish Eulerian Hemispheric Model

In this section the DEHM model is described, including an overview of the numerical schemes, the chemical scheme and the deposition as well as the input data of meteorology and emissions. In DEHM, the continuity equation is solved:

\[
\frac{\partial c}{\partial t} = -\left( \frac{\partial c}{\partial x} u + \frac{\partial c}{\partial y} v + \frac{\partial c}{\partial z} \nu + \frac{\partial c}{\partial \sigma} \right) + K_v \frac{\partial^2 c}{\partial x^2} + K_{y'} \frac{\partial^2 c}{\partial y^2} + \frac{\partial}{\partial \sigma} \left( K_c \frac{\partial c}{\partial \sigma} \right) + P(c, t) - L(c, t)
\]

where \(c\) is the mixing ratio, \(t\) is time, \(u, v\), and \(\nu\) are the wind-speed components in the \(x\), \(y\) (horizontal) and \(\sigma\) (vertical) directions, respectively, \(K_v\), \(K_{y'}\), and \(K_c\) are dispersion coefficients, while \(P\) and \(L\) are production and loss terms, respectively.

2.1. Numerical schemes

In the model the basic continuity equation for each chemical component is divided into sub-models using a simple non-symmetric splitting procedure based on the ideas of McRae et al.
The horizontal advection is solved numerically using the higher-order Accurate Space Derivatives scheme, documented to be very accurate (Dabdub and Seinfeld, 1994), especially when implemented in combination with a Forester filter (Forester, 1977). An alternative approach for solving the advection is under development and based on a semi-Lagrangian scheme. This scheme has been tested against the ASD scheme in Hansen et al. (2011). The method for calculating boundary conditions is described by Frohn et al. (2002). The vertical advection as well as the dispersion sub-models are solved using a finite elements scheme (Pepper et al., 1979) for the spatial discretization. For the temporal integration of the dispersion, the 0-method (Lambert, 1991) is applied and the temporal integration of the three dimensional advection is carried out using a Taylor series expansion to third order. Frohn et al. (2002) provides further details of the splitting procedure, including a detailed description of the numerical methods used in each sub-model.

Time integration of the advection is controlled by the Courant-Friedrich-Lewy (CFL) stability criterion. For each advection time step, the maximum time step for the whole 3D domain is found from the local grid resolution and 3D wind-speed in every grid point so that the CFL criterion is fulfilled everywhere in the domain. For the mother domain the time step will typically be between 10 and 20 min. The time step in each sub-nest is typically approximately one-third of that for the parent nest. A detailed description of the numerical methods as well as the testing of the methods can be found in (Christensen, 1997; Frohn et al., 2002), and references herein.

The chemistry sub-model is solved using a combination of the Euler Backward Iterative method (Hertel et al., 1993) and the two-step method (Verwer and Simpson, 1995), implementing a very thorough error check procedure, which includes time step adjustment based on concentration change; for example, this can entail smaller time steps around sun-rise and sun-set at any location in the model domain. The two-step method is found to be the most accurate chemical solver (Verwer and Simpson, 1995), however, it requires two initial fields in order to get started, and for this purpose the Euler Backward Iterative (EBI) method is used.

The Forester and Bartnicki filters are applied to resolve Gibbs oscillations and negative concentration estimates, respectively (Forester, 1977; Bartnicki, 1989), however, the Bartnicki filter is only used for the background field and not for the tagged field described in Section 4. Boundary conditions (BCs) for the outermost domain depend on the direction of the wind, such that free BCs are used for sections where the wind is blowing out of the domain. Constant BCs are used for sections of the boundary where the wind is blowing into the domain; in this case, the boundary value is set to the annual average background concentration. For ozone, the initial and boundary conditions are based on ozonesonde measurements, interpolated to global monthly 3D values with a resolution of 4° × 5° (Logan, 1999).

2.2. Chemical scheme

The basic chemical scheme in DEHM includes 67 different species and is based on the scheme by Strand and Hov (1994). The original scheme has been modified in order to improve the description of, amongst other things, the transformations of nitrogen containing compounds. The chemical scheme has been extended with a detailed description of the ammonia chemistry through the inclusion of ammonia (NH₃) and related species: ammonium-nitrate (NH₄NO₃), ammonium bisulphate (NH₄HSO₄), ammonium sulphate ([NH₄]₂SO₄) and particulate nitrate (NO₃⁻) formed from nitric acid (HNO₃) using an aerosol equilibrium approach with reaction rates dependent on the equilibrium (Frohn, 2004). Furthermore, reactions concerning the wet-phase production of particulate sulphate have been included. Several of the original photolysis rates as well as rates for inorganic and organic chemistry have been updated with rates from the chemical scheme applied in the EMEP model (Simpson et al., 2003). The current model describes concentration fields of 58 photo-chemical compounds (including NOₓ, SOₓ, VOC, NHₓ, CO, etc.) and 9 classes of particulate matter (PM₁₀, PM₂·₅, sea-salt <2·₅ µm, sea-salt >2·₅ µm, smoke from wood stoves, fresh black carbon, aged black carbon, and organic carbon). A total of 122 chemical reactions are included. Furthermore, the model has options for an additional chemical scheme for mercury (Hg) and a module for emissions and transport of persistent organic pollutants (POPs), including an extensive description of air-surface exchange of POPs for soil, water, vegetation and snow.

2.3. Dry and wet deposition

Dry deposition velocities are calculated at each time step for each grid cell in the surface layer for 16 different land-surface classes. The calculation of the dry deposition velocity is based on the resistance method including the aerodynamic resistance (r_a), the laminar boundary layer resistance (r_b) and the surface resistance (r_s). For vegetative surfaces, the surface conductance (the reciprocal of the resistance) is composed of two parts; the stomatal conductance and the non-stomatal conductance. The land use information for Denmark is obtained from the AIs database (Area Information System) which covers Denmark with very high resolution (100 m × 100 m). The land use information for the rest of the world is based on the Olson World Ecosystem Classes v1.4D (Olson, 1992). The implementation of the deposition module follows the overall methodology of the implementation in the EMEP module (see Simpson et al., 2003 and Emberson et al., 2000).

The parameterisation of wet deposition is based on a simple scavenging ratio formulation with in-cloud and below-cloud scavenging coefficients for both gas and particulate phase based on Simpson et al. (2003).

2.4. Meteorology

The DEHM model is driven with meteorological data from numerical weather prediction models and is currently coupled to data from two different numerical weather prediction models, the MM5v3·7 model (Grell et al., 1995) or theEta model (Janjic, 1994), both driven by global data from either ECMWF or NCEP. More details on the setup of the meteorological model used for the AQMEII exercise are found in Section 3.

2.5. Emissions

Emissions are based on several inventories, including global emission databases used in the Northern Hemispheric domain, namely EDGAR2000 Fast track (Emission Database for Global Atmospheric Research; Olivier and Berdowski, 2001), GtJ (Global Emission Inventory Activity; Graedel et al., 1993) both with a 1° × 1° resolution and RCP (Representative Concentration Pathways) with a 0·₅° × 0·₅° resolution for historical data (Lamarque et al., 2010) and future scenarios (Clarke et al., 2007; Smith and Wigley, 2006; Wise et al., 2009). Emissions from the EMEP expert database (European Monitoring and Evaluation Programme) with
50 km × 50 km resolution (Mareckova et al., 2008) are used in the nested domain over Europe.

Emissions for Denmark are based on the national emission inventory for NH₃, NO₂ and SO₂ (http://www.dmu.dk/luft/emissioner/air-pollutants). For NH₃ the basic spatial resolution is 100 m × 100 m which is then aggregated to the 50 km × 50 km grid. The temporal distribution of the ammonia emissions is calculated using a dynamical ammonia emission model (Skjøth et al., 2004; Gyldenkerne et al., 2005). For NO₂ and SO₂ the spatial resolution is 50 km × 50 km and the data include about 70 larger point sources. Ship emissions both around Denmark with very high resolution (Olesen et al., 2009) and global (Corbett and Fischbeck, 1997) are included.

With respect to emissions from natural sources, emissions from retrospective wildfires are included based on Schultz et al. (2008). The current version of DEHM includes the temporal allocation of emissions from the IGAC-GEIA biogenic emission model for biogenic isoprene (International Global Atmospheric Chemistry – Global Emission Inventory Activity; Guenther et al., 1995) as an in-line module. Natural emissions of NOₓ from lightning and soil as well as emissions of NH₃ from soil/vegetation based on GEIA are also implemented in the model.

The emissions in all domains are distributed in time and with height above the surface following patterns depending on the source categories.

3. Model setup in AQMEII and evaluation for Europe

For the AQMEII inter-comparison exercise, the DEHM model was set up with a mother domain covering the Northern Hemisphere and limited parts of the Southern Hemisphere and two nested domains simultaneously—one covering Europe and one covering North America—in the following referred to as the EU domain and as the NA domain, respectively, see Fig. 1. The model is defined on a polar stereographic projection true at 60° N with a resolution in the mother domain of 150 km × 150 km and in the nested domains of 50 km × 50 km. Both the mother domain and the two nested domains consist of 150 × 150 grid cells in the horizontal plane. The vertical grid is defined using the σ-coordinate system, with 29 vertical layers extending up to a height of 100 hPa. The model was run for the full year of 2006, which is the basic year for the AQMEII intercomparison. Data from a previous model run for the year 2005 was used as initial conditions.

In the present study, the model is driven with meteorological data from the MM5v3.7 model using global analyzed meteorological data from NCEP (1° × 1° spatial and 6 h temporal resolution) as input and defined on the same domains and resolutions both horizontally and vertically as in DEHM. The simulation was performed as a single model run, and was forced towards the 6-hourly global analysis using nudging. A spin-up time of 24 h was used to initiate the meteorological simulation. The meteorological data are archived at 1 h intervals.

In the MM5 simulation, we used the following schemes: the mixed phase Reisner 1 explicit moisture scheme (Reisner et al., 1998), the Betts–Miller cumulus scheme (Betts and Miller, 1993), the Mellor-Yamada-Janic planetary boundary layer scheme (Janjic, 1990, 1994), the CCM2 radiation cooling scheme (Hack et al., 1993), the Noah land-surface scheme with 4 snow/soil layers (Chen and Dudhia, 2001), and a shallow convection scheme to handle non-precipitating clouds. Snow depth was calculated dynamically as a prognostic variable.

For the mother domain and the North American domain, anthropogenic emissions based on RCP with a 0.5° × 0.5° resolution were applied for the year 2005. For the European nested domain, emissions from EMEP were used for the year 2006. Natural emissions were included as described in section 2.5.

3.1. Model results

In this sub-section, annual mean values for ozone and total PM₂.₅ are shown for the three domains used in the AQMEII intercomparison exercise. These species were chosen to illustrate the model results since they are both subject to long-range transport and are both harmful to human health. The total PM₂.₅ consists in the model of the sum of the following species: primary emitted mineral dust, black carbon (fresh and aged), organic carbon, and the secondary formed particles H₂SO₄, NH₄NO₃, NH₄HSO₄ and (NH₄)₂SO₄. Secondary formed organic aerosols (SOA) are not presently included in the model, and we therefore expect the total PM₂.₅ levels to be underestimated compared to measurements.

In Fig. 2, the annual mean concentrations for ozone and PM₂.₅ are shown for the Northern Hemisphere. High values of ozone are especially seen over Asia due to relatively higher emissions of ozone precursors in these areas and over the oceans due to the low deposition of ozone over water surfaces. High values over central Africa are due to a high level of biogenic isoprene emissions, whereas the high values over Greenland are due to the high altitude of the surface (up to 3 km above sea level). For PM₂.₅ high annual mean values are seen in the eastern part of China as well as over the highly populated areas as eastern USA, central Europe and India.

In Fig. 3, the annual mean values for ozone and PM₂.₅ are shown for the NA domain. Background levels of ozone vary between 25 ppb and more than 45 ppb in the USA with the highest values simulated over the western parts of the USA. In Canada the annual mean ozone concentrations are relatively lower—between 17 and 30 ppb. The annual mean value for the whole NA domain over land
is estimated to 32 ppb. For PM$_{2.5}$ high annual mean values are seen in the eastern part of USA of around 8–12 µg m$^{-3}$. The mean value for the whole nested domain over land is 3.3 µg m$^{-3}$.

In Fig. 4, the annual mean values for ozone and PM$_{2.5}$ are shown for the EU domain. The mean background levels of ozone over the land covered part of the EU domain is 34 ppb with a range from around 20 ppb in the area of Berlin to 45 ppb in the Alps, however, higher values are seen in the eastern part of the domain and over Greenland. For PM$_{2.5}$ typical annual values range from 8 to 20 µg m$^{-3}$ with the highest values in Eastern Europe as well as Belgium, the Netherlands and the Po Valley. Furthermore, high values are seen around Sicily due to the activity from the volcano Etna emitting sulphur-oxides. The annual mean value over the land covered part of the domain covering Europe is 5.1 µg m$^{-3}$.

Fig. 2. Annual mean concentrations of ozone (left figure) and total PM$_{2.5}$ (right figure) for the year 2006 calculated by the DEHM model. Results are shown for the mother domain.

Fig. 3. Annual mean concentrations for ozone (left figure) and total PM$_{2.5}$ (right figure) for the year 2006 calculated by the DEHM model. Results are shown for the nested NA domain.
Examining the results in Figs. 3 and 4 (nested domains) relative to Fig. 2 (hemispheric domain) the added detail of the nested grid versus the hemispheric model is clearly seen. This is especially true for city-scale “hot spots” of ozone and PM in Europe.

### 3.2. Evaluation against measurements in Europe and North America

In Figs. 5–9, the DEHM model is evaluated for Europe by comparing model results with measurements from the EMEP (European Monitoring and Evaluation Programme, www.emep.int) monitoring network. In Figs. 10–13, the model is evaluated for Europe and North America based on data from the Ensemble database (Bianconi et al., 2004).

In Figs. 5–8, daily mean values for 12 different chemical compounds for the year 2006 are applied in the evaluation. Model results are interpolated to the locations of 208 monitoring sites, using the nearest four grid points and average values over all the measurement stations for each day are calculated for the model results as well as the measurements whenever measurements are available. The time series in Figs. 5–8 are therefore calculated as a spatial mean showing the time evolution as a mean over Europe. Even though the measurement network includes a large number of stations, not all the stations measure all the species all the time. For the interpretation of the results below, the number of stations actually measuring the different chemical compounds (for at least part of the year) is given for each compound in the figures. The spatial variability of the annual values is shown in Figs. 2–4.

In Fig. 5, results are given for the chemical species NH$_3$, NH$_4^+$, SNH (≡ NH$_3$ + NH$_4^+$), NH$_4^+$ WD (wet deposition) and precipitation. NH$_3$ (ammonia) can be considered a local chemical compound with a short atmospheric life time, reacting relatively fast to form ammonium-sulphate and ammonium-nitrate and with a high dry deposition rate. As seen in Fig. 5, the model nevertheless shows fine skill in terms of the overall annual variability with a correlation of 0.68 and only some underestimation during the summer months. The model is quite accurate in simulating the concentrations of ammonium (NH$_4^+$), yielding a correlation of 0.77. In general, the secondary inorganic aerosols (SIA) are expected to be simulated well by the model, since these species have a life time of several days and therefore are easier to simulate with a regional model, compared to species with a shorter life time. The overall features of SNH are also captured by the model as is the wet deposition of NH$_4^+$. The wet deposition is measured as the concentration of the species in precipitation, which also includes the wet deposition of ammonia, which is transformed into ammonium in the precipitation sample. It is also important that the daily precipitation levels are reproduced with sufficient accuracy, and this is shown as the lower graph in Fig. 5. The performance with respect to wet deposition is a little worse than the performance for concentrations of ammonium indicating that the limitation lies within the performance of ammonia. However, the reason can also be the timing of the precipitation events.

In Fig. 6, results are given for the chemical species HNO$_3$, NO$_3^-$, SNO$_3$ (≡ HNO$_3$ + NO$_3^-$), NO$_3^-$ WD (wet deposition) and precipitation. Like NH$_3$, HNO$_3$ is a relatively short-lived species; however, the model captures the concentrations in the summer half year well, but underestimates during the winter season. NO$_3^-$ is also well simulated in the summer season, but overestimated in the winter season, suggesting a possible deficiency in the chemistry with a too low reaction rate from HNO$_3$ to NO$_3^-$ during the winter season. For the sum SNO$_3$, the model captures the overall variability well with only a smaller fractional bias of 9%. The annual average levels of NO$_3^-$ WD are well reproduced, however, with a lower correlation than the air concentration of NO$_3^-$, indicating that the uncertain parameter is the concentrations of HNO$_3$, which are included in the wet deposition measurements of NO$_3$.

In Fig. 7, results are given for the chemical species NO$_2$, O$_3$, SO$_2$, SO$_4^{2-}$ and SO$_4^{2-}$ WD (wet deposition). Concentrations of these species in the air are all reproduced well with correlations above
NO₂ is underestimated in the winter months, which could be due to meteorological situations of low vertical mixing in these months or even nocturnal inversion layers, which are difficult to simulate with the meteorological model. In particular, this has impacts on the primary emitted species released at ground-level. O₃ is somewhat underestimated in the summer months, which is partly due to an underestimation at the stations in Spain (not shown). Based on yet unpublished experiments with the implementation of the MEGAN model for biogenic emissions of isoprene, it seems that emissions of isoprene are too low in the present version of DEHM, which in turn has a considerable impact on ozone levels especially in the Mediterranean region. SO₄²⁻ is one of the chemical compounds that is best simulated by the DEHM model. For both the temporal variability and the annual mean, there is a good agreement between observed and modelled values. The wet deposition of SO₄²⁻ has a much lower correlation and is underestimated by 39%, with a performance worse than the concentrations in air and the precipitation levels, indicating that the parameterization of wet deposition for this species could be improved.

In Fig. 8, we have collected the results for particulate matter. Results are given for the chemical species of total PM₂.₅, total PM₁₀, sea salt, wet deposition of sea salt and secondary inorganic aerosols (SIA). SIA are measured at only 13 stations, where all the components of SIA are included. The total PM₂.₅ in the model consists, as mentioned in the previous section, of the sum of the species:

![Graph showing the evaluation of DEHM model results as daily mean values for the year 2006 compared to available measurements from the EMEP network taken as average values in space over all the measurement stations for each day. Results are given for the chemical species: NH₃ (19), NH₄⁺ (26), SNH (NH₃ + NH₄⁺) (35), NH₄⁺ WD (wet deposition) (38) and precipitation (35). The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE).](image-url)
primary emitted mineral dust, black carbon (fresh and aged), organic carbon, sea salt, and the secondary formed particles included in SIA: \( \text{SO}_4^{2-} \), \( \text{NO}_3^- \), \( \text{NH}_4\text{NO}_3 \), \( \text{NH}_4\text{HSO}_4 \) and \((\text{NH}_4)_2\text{SO}_4\). Secondary formed organic aerosols are not yet included in the model, and therefore we expect the total PM2.5 and total PM10 levels to be underestimated compared to measurements, this is also the case as can be seen from the figure. To improve the model’s ability to simulate particulate matter and especially the total particle mass in order to obtain mass closure for PM2.5 and PM10, we are currently working on implementing a scheme for emissions of biogenic mono-terpenes and extending the chemical scheme of the model to simulate SOA. The simulation of SIA and sea salt seems to be in good agreement with measurements, with correlations of 0.68 and 0.78, respectively and small overall bias.

In the evaluations carried out in the following (Figs. 9–13), observations and modelled values were extracted from the Ensemble database (Bianconi et al., 2004). All stations which were classified as “rural”, below 1500 m altitude and with less than 25% missing data were included. For the European domain, the observational data were originally from the EMEP or AIRBASE networks. For the North American domain, the observations were collected by the AIRS, CAST and NAPS networks.

In Fig. 9, an evaluation of DEHM model results as a scatter plot including the annual mean values for each measurement station in

![Fig. 6. Evaluation of DEHM model results as daily mean values for the year 2006 compared to available measurements from the EMEP network taken as average values in space over all the measurement stations for each day. Results are given for the chemical species HNO3 (16), NO3 (25), \( \text{SNO}_3^- \) (35), NO3 WD (wet deposition) (38) and precipitation (35). The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE).]
Europe for the year 2006 compared to available measurements from the EMEP is shown. Results are given for the chemical species $O_3$, $NO_2$, $SO_4^{2-}$, and $PM_{2.5}$ as examples. For $O_3$ the results are given only for the summer half year (April to September) as this is the period where ozone is most interesting. As seen in the results for $O_3$, the spatial correlation coefficient (0.39) is less than for the time series (0.85), showing that the model performs better in the time variation than the spatial variation. For $NO_2$, the time series showed an underestimation during the whole year, and from the scatter plot it is seen that this originates from specific measurement sites in Italy (I), Switzerland (CH) and Czech Republic (CS), while the rest of the sites show relatively good agreement in the annual values.

The performance for sulphate is showing relatively good agreement in the spatial correlation (0.70) and almost no overall bias, while the results for $PM_{2.5}$ show that the model underestimates the annual values.

An evaluation of DEHM model results for the daily maximum values for each measurement station in Europe for the year 2006 compared to available measurements from the EMEP and AIRBASE networks (taken from the Ensemble database) are given in Fig. 10. In the EMEP database hourly values are only provided for ozone, so therefore we have used the Ensemble database for this evaluation. Results are given for the chemical species $O_3$, $NO_2$, and $PM_{2.5}$. The scatter-plot for all stations and all days is represented as a two-

![Graph]

**Fig. 7.** Evaluation of DEHM model results as daily mean values for the year 2006 compared available measurements from the EMEP network taken as average values in space over all the measurement stations for each day. Results are given for the chemical species $NO_2$ (38), $O_3$ (83), $SO_2$ (51), $SO_4^{2-}$ (54) and $SO_4^{2-}$ WD (wet deposition) (39). The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE).
dimensional density plot (shown in blue). For each station, the average of the daily maxima is shown as a circle. The solid line corresponds to observed = modelled, while the dashed line is a line of best fit through the origin using a robust linear regression. The results show a good agreement for the daily maximum values for ozone, which are also the most important with respect to regulation of this species as it is associated with hourly limit values with respect to human health. For NO₂, the model seems to capture the annual mean values for most stations correctly (Fig. 9), but underestimates the daily maxima in general.

In Fig. 11, the DEHM model results are evaluated for North America as daily mean values for the year 2006 compared to available measurements from the AIRS, CAST and NAPS networks (taken from the Ensemble database) taken as average values in space over all the measurement stations for each day. Results are given for the chemical species PM_{2.5} (12), PM_{10} (21), Sea salt (Sea) (24), wet deposition of sea salt (Sea WD) (39) and secondary inorganic aerosols (SIA) (13), which includes the sum of NH₄⁺, NO₃⁻ and SO₄²⁻. The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE).
however with relatively high correlation coefficients. The emission inventory used for the North American domain is based on the RCP emissions for the year 2005, and either the emissions for NOx are too low for this area or the measurement sites are located in areas with urban chemistry.

An evaluation of DEHM model results as annual mean values for each measurement station in North America for the year 2006 compared to available measurements from the AIRS, CAST and NAPS networks (taken from the Ensemble database) is given in Fig. 12. Results are given for the chemical species O3, NO2, SO4\(^2-\) and PM\(\text{2.5}\). N denotes the number of stations where measurements were available. Statistics shown are the mean values, the standard deviations, the correlation coefficient (Corr), including a t-test for significance, the bias, the Fractional Bias (FB), the Fractional Standard Deviation (FSD) and the Normalised Mean Square Error (NMSE).

Finally in Fig. 13, DEHM model results are evaluated for the daily maximum values for each measurement station in North America as in Fig. 12. As in Fig. 10, the scatter-plot for all stations and all days is represented as a two-dimensional density plot (shown in blue) and the average of the daily maxima is shown as a circle. The model captures the daily maximum values for O\(_3\) quite well for this area, while the NO\(_2\) and PM\(\text{2.5}\) are underestimated for reasons explained previously.

4. Intercontinental transport between NA and EU

In order to estimate the intercontinental transport, we performed two simulations to calculate the contribution from the
anthropogenic emissions in the two sub-domains in the model to
the total concentration levels in the hemispheric domain. We have
calculated the contribution from the anthropogenic emissions in
NA and EU to the total concentration levels in both NA and EU using
a land-sea mask, so only changes in the average concentrations
over land are taken into account in the two sub-domains. In order
to calculate the contributions, we have used a recently imple-
mented tagging method described below. Apart from the tagging,
these simulations differed from the model configuration given in
section 3 in that no nesting was used (i.e. only the hemispheric
domain was simulated, but results are extracted for the land areas
covered by the nested domains).

4.1. Tagging

To calculate the contribution from a specific source area to the
total concentrations (the so-called $\delta$-concentrations), we have
used a newly implemented tagging method, taking into account
the non-linearity of atmospheric chemistry (Brandt et al., 2011).

In order to do this, one can in principle run an Eulerian CTM
twice — once with all emissions and once with all emissions
minus the specific source. The difference between the two
resulting annual mean concentration fields gives an estimate of
the $\delta$-concentration. We will refer to this as the “subtraction
method”.

Modern Eulerian CTMs rely on higher-order numerical methods
to solve the atmospheric advection in order to avoid numerical
diffusion. Although higher-order algorithms are relatively accurate,
they nevertheless introduce a certain amount of spurious oscillations
or noise — called the Gibbs phenomenon. These unwanted
oscillations can cause major problems for estimating $\delta$-concentra-
tions via the subtraction method. We have found through a number
of experiments that the $\delta$-concentrations may be of similar or even
smaller order of magnitude compared to the numerical oscillations.
To avoid this problem, we implemented a more accurate method
for estimating the contribution from specific emissions. We call this
method “tagging”, denoting that we keep track of contributions to
the concentration field from a particular emission source or sector.

Fig. 10. Evaluation of DEHM model results for the daily maximum values for each measurement station in Europe for the year 2006 compared to available measurements from the EMEP and AIRBASE networks (taken from the Ensemble database). Results are given for the chemical species $O_3$ (459), $NO_2$ (306), and $PM_{2.5}$ (17). The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE). The scatter-plot for all stations and all days is represented as a two-dimensional density plot (shown in blue). For each station, the average of the daily maxima is shown as a circle. The solid line corresponds to observed — modelled, while the dashed line is a line of best fit through the origin using a robust linear regression. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
The idea is that we model the $\delta$-concentrations explicitly, rather than calculating them post-hoc (i.e. by subtraction). Tagging takes advantage of the fact that the numerical noise is typically proportional to the concentrations being modelled. Even if the $\delta$-concentrations are much smaller than the "background" concentrations (i.e. for some baseline scenario), they will generally be orders of magnitude larger than the oscillations using the tagging method. Tagged simulations have also been made with other chemical transport models (e.g., Fisher et al., 2009; Wu et al., 2011; Iversen and Seland, 2002), however, these typically calculate $\delta$-concentrations for a small number of species. By contrast, in the present study the full non-linear effects of the chemistry are incorporated by using the complete chemical scheme in both the background and the tagged part of the model.

Tagging involves modelling the background concentrations and the $\delta$-concentrations in parallel. This is straightforward for the linear processes, but special treatment is required for the non-linear process of atmospheric chemistry, since the $\delta$-concentrations are strongly influenced by the background concentrations in such processes; although this treatment involves taking the difference of two concentration fields, it does not magnify the oscillations, which are primarily generated in the advection step. Thereby the non-linear effects (e.g. from the background chemistry) can be accounted for in the $\delta$-concentrations without losing track of the contributions arising from the specific emission source or sector due to the Gibbs phenomenon.

Technically, the concentration field for a specific emission source (tag) is modelled in parallel with the background field (bg) in the CTM. For the linear processes: emissions, advection, atmospheric diffusion, wet deposition, and dry deposition the concentrations are calculated in parallel, doubling the computing time and memory requirements. For the non-linear process, the tagged concentration fields are estimated by first adding the background and tag concentration fields, then applying the non-linear operator (chemistry). The concentration field obtained by applying the non-linear operator to the background field alone is then subtracted. Thus the contribution from the specific emission source is accounted for appropriately without assuming linearity of the non-linear atmospheric chemistry. In the following this method is applied for the anthropogenic emissions both in North America and Europe.

4.2. Contribution to the total air pollution levels from emissions in NA and EU

In Figs. 14 and 15, the contribution to the mean annual air pollution levels ($\delta$-concentrations) calculated using the DEHM
model due to all anthropogenic emissions from the whole of NA (Fig. 14) and from the whole of EU (Fig. 15) for all anthropogenic emissions for the year 2006 is presented. Results are shown for ozone and the total anthropogenic PM$_{2.5}$ for the hemispheric domain. Furthermore, the contributions are given in Tables 1–3 for selected species. In Table 4 the total annual anthropogenic emissions are given for the two domains. The tagged concentration fields are calculated for all the chemical species in the model and the results for the two species shown depend highly on the full atmospheric chemistry as well as the other processes, such as atmospheric transport.

Fig. 14 shows that the contribution from anthropogenic emissions in the NA domain to the ozone concentration level is more than 10 ppb in the eastern part of USA and between 5 and 10 ppb in the rest of the country. In Canada the contribution from these sources is between 2 and 5 ppb. The mean contribution to the land based area in the NA sub-domain is 4.5 ppb. As the mean concentration of ozone in the same domain is 32 ppb, the contribution from emissions in NA to the same land based area is 14% on annual basis (see Table 1). The remaining 86% are due to all other anthropogenic emissions in the Northern Hemisphere (except NA) and natural emissions in general. The contribution to EU from the NA emissions is between 1 and 2 ppb with a mean of 1.1 ppb. With a mean value over land in the EU domain of 34 ppb, the contribution from the emissions in the NA domain is 3.1%.

For PM$_{2.5}$ the largest contribution from NA is in the eastern part of the USA (which is the same as for ozone), with typical values around 5–10 $\mu$g m$^{-3}$. For the rest of the USA, the contribution ranges between 1 and 5 $\mu$g m$^{-3}$ with a mean value of 2.1 $\mu$g m$^{-3}$ for the whole land based area in the sub-domain. The contribution from the NA emissions in the NA domain is 63%. The contribution from NA to EU is ranging between 0.02 and 0.2 $\mu$g m$^{-3}$ with a mean contribution of 0.05 $\mu$g m$^{-3}$ for the whole land based domain. With a mean over the EU sub-domain of 5.1 $\mu$g m$^{-3}$, the contribution from NA is 0.9%. The annual mean values for the two chemical compounds due to all emissions in the Northern Hemisphere, anthropogenic as well as natural, are shown in Fig. 3.

Fig. 15 shows that the contribution from the EU emissions to the ozone concentrations is between 5 and 10 ppb in the Mediterranean area and between 2 and 5 ppb in the central parts of Europe. The mean contribution to the EU sub-domain from EU emissions is 1.8 ppb, corresponding to 5.2%. According to these results, EU emissions contribute to the ozone background levels with less than 2 ppb on the hemispheric scale. The mean contribution from EU sources to the NA domain is 0.3 ppb or 0.9%.
For PM$_{2.5}$, the contribution from sources in the EU domain has a maximum in central Europe with typical values around $5-10$ µg m$^{-3}$. The mean contribution over the whole EU land based domain is 1.8 µg m$^{-3}$, corresponding to 35%. A large area corresponding to a contribution of less than 1 µg m$^{-3}$ is seen in a large part of the hemisphere, but the contribution to the NA area is less than 0.1 µg m$^{-3}$ with a mean of 0.05 µg m$^{-3}$ or 1.4% of the total concentrations in the domain. The annual mean values for the two chemical compounds due to all emissions in the Northern Hemisphere, anthropogenic as well as natural, are shown in Fig. 4.

When estimating the intercontinental contributions, all the anthropogenic emissions for all species are tagged in the model, including the non-linear chemistry. In Tables 1–3, we present results for selected species in the model related to ozone and particulate matter as annual mean concentrations (Table 1), mean over the summer period (Table 2) and mean over the winter period (Table 3) over the land areas within by the two AQMEII domains. The tables also show the contributions (in percentage) from the anthropogenic emissions in the areas covered by the two sub-domains to the mean concentrations in the two areas for selected chemical species.

As seen in these tables, there are some differences between the different chemical species. This should come as no surprise, as the atmospheric chemistry is non-linear and since there are large differences in the atmospheric lifetimes of the species listed.

The contribution from the anthropogenic emissions of VOCs and NO$_x$ in the EU domain to the annual mean ozone concentrations in the EU domain is 5.2% on the annual basis. During the summer period the contribution is 7.8% due to enhanced photo-chemistry and during winter it is only 2.6%. The contribution from emissions in NA to the annual mean concentrations in EU is 3.1%, while the contribution the other way is only 0.9% and these contributions do not vary much in the summer and winter periods. The higher contribution from NA to EU than the other way fits with the prevailing pattern of surface winds from west to east. Furthermore, the total anthropogenic emissions of the ozone precursors are similar in the two domains, emphasizing that the contribution from NA to EU is largest due to the shorter transport pathway. The contribution

![Fig. 13. Evaluation of DEHM model results for the daily maximum values for each measurement station in North America for the year 2006 compared to available measurements from the AIRS, CAST and NAPS networks (taken from the Ensemble database). Results are given for the chemical species O$_3$ (327), NO$_2$ (95), and PM$_{2.5}$ (15). The number of stations where measurements were available is given in parentheses. Statistics shown are the mean values, the Fractional Bias (FB), the correlation coefficient (Corr) and the Normalised Mean Square Error (NMSE). The scatter-plot for all stations and all days is represented as a two-dimensional density plot (shown in blue). For each station, the average of the daily maxima is shown as a circle. The solid line corresponds to observed = modelled, while the dashed line is a line of best fit through the origin using a robust linear regression. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image-url)
Fig. 14. Contribution from the anthropogenic emissions in the NA domain to the mean annual air pollution levels (μ-concentrations) in the whole mother domain for the year 2006 calculated using the DEHM model with tagging. The compounds are ozone (left figure) and total PM$_{2.5}$ (right figure).

Fig. 15. Contribution from the anthropogenic emissions in the EU domain to the mean annual air pollution levels (μ-concentrations) in the whole mother domain for the year 2006 calculated using the DEHM model with tagging. The compounds are ozone (left figure) and total PM$_{2.5}$ (right figure).
Table 1
Contributions in percent from anthropogenic emissions for the year 2006 in the European domain to the annual mean concentrations in the European domain (EU → EU), from emissions in North America to concentrations in North America (NA → NA), from emissions in Europe to concentrations in North America (EU → NA) and from emissions in North America to concentrations in Europe (NA → EU) for selected chemical species in DEHM using the tagging method. The first two columns give the mean concentrations over land of the species in the EU and NA domains. Units for the mean values are ppb for the gases and % for the particles. BC is black carbon (fresh and aged). The “Total SO4” is a sum of SO4$^2-$, NH4HSO4 and (NH4)2SO4. “Total NO3” is a sum of NO3 and NH4NO3. Total PM2.5 is the sum of SIA, BC, OC and primary emitted mineral dust.

<table>
<thead>
<tr>
<th>Chemical species</th>
<th>Mean EU</th>
<th>Mean NA</th>
<th>EU → EU [%]</th>
<th>NA → EU [%]</th>
<th>NA → NA [%]</th>
<th>EU → NA [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH</td>
<td>6.85E-05</td>
<td>6.65E-05</td>
<td>-0.1</td>
<td>32.6</td>
<td>-0.2</td>
<td>32.6</td>
</tr>
<tr>
<td>C2H6</td>
<td>1.15E-00</td>
<td>9.53E-01</td>
<td>12.9</td>
<td>14.9</td>
<td>3.6</td>
<td></td>
</tr>
<tr>
<td>H2O2</td>
<td>6.00E-01</td>
<td>4.93E-01</td>
<td>4.4</td>
<td>3.5</td>
<td>-0.9</td>
<td>1.0</td>
</tr>
<tr>
<td>O3</td>
<td>3.37E-01</td>
<td>3.18E-01</td>
<td>5.2</td>
<td>14.1</td>
<td>0.9</td>
<td></td>
</tr>
<tr>
<td>NO2</td>
<td>7.30E-01</td>
<td>8.30E-01</td>
<td>34.8</td>
<td>-1.4</td>
<td>65.4</td>
<td>-0.3</td>
</tr>
<tr>
<td>PAN</td>
<td>2.09E-01</td>
<td>2.16E-01</td>
<td>36.8</td>
<td>11.0</td>
<td>57.8</td>
<td>6.6</td>
</tr>
<tr>
<td>SO2</td>
<td>4.16E-01</td>
<td>2.27E-01</td>
<td>25.1</td>
<td>-0.7</td>
<td>83.7</td>
<td>-0.2</td>
</tr>
<tr>
<td>CO</td>
<td>1.02E-02</td>
<td>1.03E-02</td>
<td>11.1</td>
<td>4.8</td>
<td>20.2</td>
<td>2.6</td>
</tr>
<tr>
<td>BC</td>
<td>1.62E-01</td>
<td>1.01E-01</td>
<td>38.1</td>
<td>0.20</td>
<td>72.9</td>
<td>0.19</td>
</tr>
<tr>
<td>Total SO4</td>
<td>5.13E-01</td>
<td>2.99E-01</td>
<td>34.9</td>
<td>1.3</td>
<td>81.5</td>
<td>0.8</td>
</tr>
<tr>
<td>Total NO3</td>
<td>3.36E-01</td>
<td>2.30E-01</td>
<td>45.9</td>
<td>1.1</td>
<td>89.5</td>
<td>0.4</td>
</tr>
<tr>
<td>Total PM2.5</td>
<td>5.15E-00</td>
<td>3.26E-00</td>
<td>35.0</td>
<td>0.9</td>
<td>63.4</td>
<td>1.4</td>
</tr>
</tbody>
</table>

from emissions in NA to the annual mean concentrations in NA for ozone is 14.1% varying from 19.8% in summer to 9.1% during winter.

For particles, the contribution from the emissions in EU to the annual mean concentrations in EU is 35%, while the contribution from the emissions in NA to the concentrations in same area is 63%. The contributions from NA to EU for the total PM2.5 is only 0.9% and 1.4% from EU to NA. The contribution from NA to EU and vice versa are actually equal in absolute numbers – namely 0.05 µg m$^{-3}$ as mentioned above. As seen in Tables 2 and 3 the contributions do not vary much between summer and winter. As seen in Table 4, the total anthropogenic emission of primary particles is nearly twice as large in EU compared to the NA domain. This compensates for the longer transport pathways from EU to NA giving equal absolute contributions between the two domain for the primary particles. Results are also given for BC as well as for total SO4 and total NO3. While BC is an inert tracer, chemistry is involved in the secondary particle components and this gives some variation of the contributions for the different particles as seen in the table.

As seen in the tables there are also some negative contributions, meaning that the emissions from one of the areas actually result in decreasing mean concentrations over the other area. The emissions in NA and EU actually result in decreased concentrations of NO2 in EU and NA, respectively. We investigated the model results for all the species and found that the decrease in NO2 is due to emissions of hydrocarbons (C2H6), which increase the mean concentrations of CH3OC(O)2 which reacts with NO2 to give more peroxy acetyl nitrate (PAN) (see the tables for these species). Negative contributions are also seen for SO4 – the contribution from sources in NA e.g. results in a decrease in SO2 of 0.7% in EU on the annual basis. This is found to be due to the increased O3 concentrations, which react with OH to give HO2. HO2 reacts with another HO2 to form H2O2 and this species together with O3 has an impact on the aqueous phase chemistry between SO2 and SO4$^2-$. These results are in agreement with previous global model simulations finding that sulphate production is sensitive to oxidant levels (Unger et al., 2006; Kloster et al., 2008; Leibensperger et al., 2011).

4.3. Comparison with similar studies

There is a long history of models conducting similar simulations for intercontinental source–receptor relationships. Recently, results from ensembles of models have been summarized in the Task Force on Hemispheric Transport of Air Pollution (TF HTAP, 2010), which includes results from the 15 models that participated in the SRES set of HTAP simulations, supplemented with results from a study including an ensemble of 21 global and hemispheric chemical transport models that were applied to estimate the source–receptor relationship for O3 (Fiore et al., 2009).

In general these ensemble studies made 20% reduction of emissions in the source regions and scaled the results linearly to 100%. Using the model ensemble approach, they found that the contribution to O3 levels from NA to EU were in the range from 1 ppb to 2.5 ppb and that the contribution from EU to NA were in the range from 0.4 ppb to 1.5 ppb. As our results are 1 ppb and 0.3 ppb, respectively for O3, we can conclude that the results presented in this paper are similar to the results summarized in ensemble studies, however in the lower end.

In TF HTAP (2010) source–receptor relationships have also been estimated for some particles based on results from six global models results for the year 2001. Contributions are given in percent as median values (and ranges given in parenthesis of the six models). The results that can be compared to results in this paper are given for

Table 2
As for Table 1, but for the summer period (April–September, 2006).

<table>
<thead>
<tr>
<th>Chemical species</th>
<th>Mean EU</th>
<th>Mean NA</th>
<th>EU → EU [%]</th>
<th>NA → EU [%]</th>
<th>NA → NA [%]</th>
<th>EU → NA [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH</td>
<td>1.03E-04</td>
<td>9.43E-05</td>
<td>7.0</td>
<td>-0.1</td>
<td>33.7</td>
<td>-0.2</td>
</tr>
<tr>
<td>C2H6</td>
<td>1.06E-00</td>
<td>9.12E-01</td>
<td>11.3</td>
<td>2.6</td>
<td>12.6</td>
<td>3.1</td>
</tr>
<tr>
<td>H2O2</td>
<td>8.81E-01</td>
<td>7.90E-01</td>
<td>4.4</td>
<td>3.1</td>
<td>0.8</td>
<td>0.9</td>
</tr>
<tr>
<td>O3</td>
<td>3.43E-01</td>
<td>2.99E-01</td>
<td>7.8</td>
<td>3.1</td>
<td>19.8</td>
<td>0.9</td>
</tr>
<tr>
<td>NO2</td>
<td>6.22E-01</td>
<td>6.99E-01</td>
<td>26.6</td>
<td>-0.4</td>
<td>53.0</td>
<td>0.0</td>
</tr>
<tr>
<td>PAN</td>
<td>1.61E-01</td>
<td>2.00E-01</td>
<td>37.5</td>
<td>7.8</td>
<td>57.8</td>
<td>4.3</td>
</tr>
<tr>
<td>SO2</td>
<td>1.92E-01</td>
<td>1.36E-01</td>
<td>21.7</td>
<td>-0.5</td>
<td>80.9</td>
<td>-0.1</td>
</tr>
<tr>
<td>CO</td>
<td>9.73E-01</td>
<td>1.02E-01</td>
<td>8.7</td>
<td>4.3</td>
<td>17.5</td>
<td>2.1</td>
</tr>
<tr>
<td>BC</td>
<td>1.16E-01</td>
<td>8.67E-02</td>
<td>37.2</td>
<td>0.1</td>
<td>70.1</td>
<td>0.2</td>
</tr>
<tr>
<td>Total SO4</td>
<td>4.82E-01</td>
<td>3.18E-01</td>
<td>37.6</td>
<td>1.0</td>
<td>82.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Total NO3</td>
<td>2.40E-01</td>
<td>1.32E-01</td>
<td>42.2</td>
<td>1.7</td>
<td>70.5</td>
<td>0.6</td>
</tr>
<tr>
<td>Total PM2.5</td>
<td>4.79E-00</td>
<td>3.18E-00</td>
<td>31.8</td>
<td>0.8</td>
<td>54.2</td>
<td>1.2</td>
</tr>
</tbody>
</table>
The Danish Eulerian Hemispheric Model (DEHM) is a 3D long-range atmospheric chemistry-transport model with a horizontal domain covering the Northern Hemisphere. The model includes a two-way nesting capability for obtaining higher resolution over selected areas. For the AQMEII inter-comparison exercise, the model was set up with two two-way nested domains simultaneously—one covering Europe and one covering North America. The model architecture as well as the configuration used in the intercomparison was described.

The model was validated for the year 2006 against available measurements in Europe and North America based on the daily mean values averaged over measurement stations as time series, and annual mean values as well as the daily maximum values as scatter plots. The evaluation of time series showed that the DEHM model gives satisfactory results for the secondary inorganic aerosols as well as for species such as ozone, nitrogen-dioxide, sulphur-dioxide, ammonium and sea salt over Europe. Limitations to the current performance of wet deposition of SIA appear to be related to the atmospheric concentrations of species such as ammonia or nitric acid, which are measured as part of the wet deposition of SIA. Better estimations of precipitation will also potentially improve the ability to simulate particulate matter and especially to obtain mass closure for PM2.5 and PM10, we are currently working on implementing a scheme for emissions of biogenic mono-terpenes based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) and extending the chemical scheme of the model to simulate SOA. The evaluation points to specific areas where DEHM can be improved, such as biogenic emissions of isoprene, mass closure for particulate matter, wet deposition, and description of vertical mixing during winter.

In this article, we focussed on the intercontinental transport between North America and Europe, estimating the contributions to the total air pollution levels at continental scale from the anthropogenic sources in the two areas. The regional and intercontinental contributions were assessed using a tagging method, taking into account the non-linear atmospheric chemistry.

We found that the contributions from NA anthropogenic emissions to the ozone levels in NA and EU are 14% and 3.1%, respectively. The contributions from EU anthropogenic sources to the ozone levels in EU and NA are 5.2% and 0.9%, respectively. The local contribution in percentage from NA to the mean concentrations in NA is considerably larger compared to the local contribution from EU to the mean concentrations in the EU domain and we found for ozone a seasonal difference between summer and winter of a factor of 3 for EU and 2 for NA.

Table 3
As for Table 1, but for the winter periods (January–March and October–December, 2006).

<table>
<thead>
<tr>
<th>Chemical species</th>
<th>Mean EU</th>
<th>Mean NA</th>
<th>EU → EU [%]</th>
<th>NA → EU [%]</th>
<th>NA → NA [%]</th>
<th>EU → NA [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH</td>
<td>3.37E+05</td>
<td>3.87E+05</td>
<td>2.4</td>
<td>-0.3</td>
<td>30.0</td>
<td>-0.1</td>
</tr>
<tr>
<td>C2H6</td>
<td>1.24E+00</td>
<td>9.94E+01</td>
<td>14.4</td>
<td>3.2</td>
<td>17.1</td>
<td>4.0</td>
</tr>
<tr>
<td>H2O2</td>
<td>3.19E-01</td>
<td>1.97E-01</td>
<td>4.6</td>
<td>4.4</td>
<td>-7.5</td>
<td>1.4</td>
</tr>
<tr>
<td>NO2</td>
<td>3.31E-01</td>
<td>3.36E-01</td>
<td>2.6</td>
<td>3.2</td>
<td>9.1</td>
<td>0.8</td>
</tr>
<tr>
<td>PAN</td>
<td>2.57E+01</td>
<td>2.32E+01</td>
<td>36.4</td>
<td>13.0</td>
<td>57.9</td>
<td>8.6</td>
</tr>
<tr>
<td>SO2</td>
<td>6.40E-01</td>
<td>3.19E-01</td>
<td>26.1</td>
<td>-0.7</td>
<td>84.9</td>
<td>-0.2</td>
</tr>
<tr>
<td>CO</td>
<td>1.06E+02</td>
<td>1.03E+02</td>
<td>13.3</td>
<td>5.3</td>
<td>22.8</td>
<td>3.0</td>
</tr>
<tr>
<td>BC</td>
<td>2.60E+01</td>
<td>1.15E+01</td>
<td>38.7</td>
<td>3.2</td>
<td>75.0</td>
<td>0.2</td>
</tr>
<tr>
<td>SO4</td>
<td>5.44E-01</td>
<td>2.80E-01</td>
<td>32.5</td>
<td>1.6</td>
<td>81.0</td>
<td>1.0</td>
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<tr>
<td>NO3</td>
<td>4.33E-01</td>
<td>3.28E-01</td>
<td>48.0</td>
<td>0.8</td>
<td>97.2</td>
<td>0.3</td>
</tr>
<tr>
<td>Total PM2.5</td>
<td>5.50E+00</td>
<td>3.33E+00</td>
<td>37.7</td>
<td>1.0</td>
<td>72.1</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Table 4
Total annual anthropogenic emissions for the year 2006 in kilotonnes for SOx, NOx, NMVOC and the primary part of PM2.5 (including mineral dust, BC and OC) for the North American (NA) and European (EU) domains.

<table>
<thead>
<tr>
<th>Chemical species</th>
<th>NA [ktonnes]</th>
<th>EU [ktonnes]</th>
</tr>
</thead>
<tbody>
<tr>
<td>SOx</td>
<td>9896</td>
<td>14103</td>
</tr>
<tr>
<td>NOx</td>
<td>9209</td>
<td>9542</td>
</tr>
<tr>
<td>NMVOC</td>
<td>18304</td>
<td>22992</td>
</tr>
<tr>
<td>PM2.5 (primary part)</td>
<td>3344</td>
<td>6097</td>
</tr>
</tbody>
</table>

isoprene emissions, which has impacts in the Mediterranean area in particular. As expected, the model underestimates PM2.5 and PM10 compared to measurements. To improve the model’s ability to simulate particulate matter and especially to obtain mass closure for PM2.5 and PM10, we are currently working on implementing a scheme for emissions of biogenic mono-terpenes based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) and extending the chemical scheme of the model to simulate SOA. The evaluation points to specific areas where DEHM can be improved, such as biogenic emissions of isoprene, mass closure for particulate matter, wet deposition, and description of vertical mixing during winter.

In this article, we focussed on the intercontinental transport between North America and Europe, estimating the contributions to the total air pollution levels at continental scale from the anthropogenic sources in the two areas. The regional and intercontinental contributions were assessed using a tagging method, taking into account the non-linear atmospheric chemistry.

We found that the contributions from NA anthropogenic emissions to the ozone levels in NA and EU are 14% and 3.1%, respectively. The contributions from EU anthropogenic sources to the ozone levels in EU and NA are 5.2% and 0.9%, respectively. The local contribution in percentage from NA to the mean concentrations in NA is considerably larger compared to the local contribution from EU to the mean concentrations in the EU domain and we found for ozone a seasonal difference between summer and winter of a factor of 3 for EU and 2 for NA.

The contributions from the NA anthropogenic emissions to the total annual PM2.5 concentrations in NA and EU are 63% and 0.9% respectively. The contributions from EU anthropogenic sources to the total PM2.5 concentrations in EU and NA are 35% and 1.4%, respectively. The contribution from anthropogenic emissions in the NA domain to the mean concentrations in the NA domain is nearly twice the contribution from anthropogenic emissions in the EU domain to the mean concentrations in the EU domain, in percentage. On the other hand, the contribution from the emissions in the EU domain to NA is larger than the contributions from the emissions in the NA to the mean concentrations in the EU domain, in percentage. However, the contribution in absolute values for PM2.5 shows that the contributions from each continent to the other is approximately the same, namely around 0.05 μg m⁻³, so the percentages emphasize that the emissions of primary particles are nearly twice as big in EU compared to NA, which compensates for the longer transport pathway. At the same time, the mean concentrations of PM2.5 is higher in Europe than in NA according to the model given a higher percentage in NA. We emphasise that the figures quoted for PM2.5 should be treated as partial estimates, since important aerosol species (e.g., secondary organic aerosols) are not included in the simulations, and the

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sulphate and BC. Their estimates for the contribution from NA to EU are 2.2% (1.1%–4.6%) for sulphate and 1.1% (0.2%–2.1%) for BC. The comparable results in this paper are 1.3% and 0.2%, respectively (see Table 1). For the contribution from EU to NA for the same species the estimates given in TF HTAP (2010) are 2.6% (0.8%–4.6%) for sulphate and 1.1% (0.2%–2.1%) for BC. Our results are 0.8% and 0.2%, respectively for the two species. Again we can conclude that the results presented in this paper are within the ranges found by other models, however in the lower end.

5. Conclusions

The Danish Eulerian Hemispheric Model (DEHM) is a 3D long-range atmospheric chemistry-transport model with a horizontal domain covering the Northern Hemisphere. The model includes a two-way nesting capability for obtaining higher resolution over selected areas. For the AQMEII inter-comparison exercise, the model was set up with two two-way nested domains simultaneously—one covering Europe and one covering North America. The model architecture as well as the configuration used in the intercomparison was described.

The model was validated for the year 2006 against available measurements in Europe and North America based on the daily mean values averaged over measurement stations as time series, and annual mean values as well as the daily maximum values as scatter plots. The evaluation of time series showed that the DEHM model gives satisfactory results for the secondary inorganic aerosols as well as for species such as ozone, nitrogen-dioxide, sulphur-dioxide, ammonium and sea salt over Europe. Limitations to the current performance of wet deposition of SIA appear to be related to the atmospheric concentrations of species such as ammonia or nitric acid, which are measured as part of the wet deposition of SIA. Better estimations of precipitation will also potentially improve the results for the calculated wet deposition. The results for sulphate indicate that the scavenging coefficients might be too small.

The mean over the EMEP stations of the daily ozone values shows an underestimation during the summer months, which is hypothesized to be due to an underestimation of the biogenic
evaluation in the European domain shows considerable underesti-
mation of the total PM$_{2.5}$.

Ozone has a longer atmospheric life time than particulate matter, especially when transported over the oceans, where the ozone deposition rate is very small. Therefore it is not a surprise that the contribution from NA to EU for ozone is larger than for PM$_{2.5}$. However, the overall mutual contributions between NA and EU are in general found to be small on an annual basis in 2006. The intercontinental exchange depends on the meteorology and emissions and some variation between different years must be expected.

In general, when estimating the contributions from a source area to the mean concentrations in a receptor area, the non-linear chemistry is very important, and therefore a tagging method taking this into account for all chemical species is necessary. The contribution from anthropogenic emissions in NA to the EU is larger than vice versa. This fits with the prevailing pattern of surface winds. However, from this study we conclude that the interconti-
tental transport between NA and EU is small for the annual or seasonal mean values – for ozone the contributions typically around 3% (~1 ppb) from NA to EU and around 1% (~0.3 ppb) from EU to NA. For particles the contributions from NA and EU to NA and EU, respectively, are typically ~0.9% and 1.4% (both ~0.05 μg m$^{-3}$). The contributions can of course, be much larger for individual transport episodes. We also found that the contributions we estimate in this study are similar to results found in other similar studies, however in the lower end of the ranges given by the ensemble estimates.

Acknowledgements

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