

Technical Note: Anthropogenic and natural offline emissions and the online Emissions and dry DEPosition submodel EMDEP of the Modular Earth Submodel system (MESSy)

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Abstract

We present the online calculated Earth's surface trace gas and aerosol emissions and dry deposition in the Modular Earth Submodel System (MESSy) submodel EMDEP as well as the currently applied anthropogenic and natural emissions inventories. These inventories, being read-in by the MESSy submodel OFFLEM, include the industrial, fossil fuel, agricultural and biomass burning emissions considering emission height profiles as a function of the source category based on the EDGAR v3.2 fast track 2000 inventory. Terrestrial and marine emissions of a selection of trace gases and aerosols are calculated online in EMDEP using climate model parameters such as wind speed, temperature and land cover and land use parameters. The online dry deposition calculation includes gases and aerosols, where the default selection for the trace gases for the dry deposition scheme can be easily extended using a commonly applied method based on trace gas solubility and reactivity. In general, the simulated global annual emissions agree with previously reported inventories, although differences exist, partly dependent on the applied model resolution. A high sensitivity of the simulated dry deposition to the applied emission height profiles stresses the importance of a realistic and consistent representation of the spatial and temporal variability in surface exchange processes in Earth system models.

1 Introduction

A key aspect of comprehensive models to study atmospheric chemistry and its role in the Earth system is the treatment of the surface boundary conditions, i.e., anthropogenic and natural emissions and dry deposition. The representation of these emissions and dry deposition is not only important for simulating realistic global burdens but also for the model evaluation in comparison to near-surface observations, which largely reflect the role of the temporal and spatial variability in surface exchanges and local photochemistry. Consequently the model representation of surface exchange

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processes should realistically incorporate some of the major drivers that determine this variability such as land cover and meteorological, biogeochemical and hydrological parameters. Here we provide a description of state-of-the-art representations of anthropogenic and natural emissions and dry deposition of reactive trace gases and aerosols in the Modular Earth Submodel System (MESSy, see Jöckel et al., 2005) submodel EMDEP. This includes an anthropogenic and natural offline emission inventory (hereafter referred to as offline emission inventory), distributed with the first official release of MESSy (version 1.1), and the online calculated emissions and dry deposition surface fluxes with MESSy's submodel EMDEP. The later is the extensive development and testing model for the representation of online emissions, dry deposition and canopy interactions (coupled representation of in-canopy dry deposition, biogenic emissions, turbulence and chemistry) (Ganzeveld et al., 2002) in MESSy.

The dry deposition and online emission routines of EMDEP actually provided the basis for the development of a distribution submodel for online emissions (ONLEM, Kerkweg et al., 2006a) and one for dry deposition (DRYDEP, Kerkweg et al., 2006b¹). The processing of the tracer emission fluxes of the offline emission inventory to update the atmospheric tracer concentrations is being done with MESSy's submodel OFFLEM (Kerkweg et al., 2006a).

More details about the emissions included in the offline emission inventory are presented in Sect. 2 followed by a description in Sect. 3 of some of the fundamental concepts of online calculated emissions and dry deposition. In Sect. 4 we present the online emission budgets simulated with EMDEP with the major drivers of these emissions, e.g., surface temperature, wind speed, precipitation, being provided by the climate model ECHAM5 (Roeckner et al., 2003). To indicate the sensitivity of the online emissions to differences in the simulation of major drivers as a function of ap-

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plied GCM's model resolution, we present the calculated budgets for three resolutions. Moreover, we present some first analysis of the sensitivity of dry deposition calculations to the inclusion of emissions heights in the offline emission inventory, followed by a summary and conclusions.

2 Offline emission inventory

The representation of surface emissions in EMDEP includes a selection of terrestrial and marine natural emissions which are calculated online whereas technologically driven emissions and a selection of natural emissions, some with an anthropogenic component such as biomass burning, are included in preprocessed input files, referred to as offline emissions, and read in for use in MESSy using the submodel OFFLEM.

These offline sources include a large selection of technological-, agricultural, biomass burning- and terrestrial biogenic and oceanic emissions of mostly trace gases. For the technological, agricultural and biomass burning emissions we use the EDGAR (Emission Database for Global Atmospheric Research, <http://www.mnp.nl/edgar/>) v3.2 fast-track update of the emissions for the year 2000 (hereafter referred to as EDGARv3.2-FT2000). For more details we refer to Olivier et al. (2005), Van Aardenne et al. (2005) and Van der Werf et al. (2003), discussing here some specific features relevant to the compiled inventory.

The EDGARv3.2-FT2000 dataset differs from the EDGARv3.2 dataset representing the 1995 emissions such that it is based on the activity data for the year 2000 applying the same emission factors as in EDGARv3.2 except for countries for which new information about emission control measures has become available. A new feature of the offline emissions is that emission height profiles have been assigned to the different EDGAR source categories distinguishing six effective emission heights (45, 140, 240, 400, 600 and 800 m a.g.l.) based on profiles used in the EMEP (Co-operative Programme on the Long-Range Transmission of Air Pollutants in Europe, <http://www.emep.int>) model. For those categories not mentioned in the EMEP work,

we arbitrarily assumed that the effective emission height is less than 45 m, except of ship emissions for which we assume that a substantial fraction is emitted at a higher altitude of 145 m.

The emission profiles are shown in Table 1. The sensitivity of simulated atmospheric chemistry to biomass burning emission height profiles, as recommended by van Aardenne et al. (2005), considering a larger vertical extent of emission heights compared to the domain described above, is currently being tested and will be included in a future release of the offline emission inventory.

The EDGARv3.2-FT2000 provides only emissions of total non-methane volatile organic compounds (NMVOC). To obtain the emissions of each individual NMVOC represented in the offline emission inventory, this total mass of NMVOC is first converted to a mass of carbon using the ratios from Table 4.7(b) of the IPCC Third Assessment Report. This total mass of carbon is then partitioned between the NMVOC species using the speciation ratios from von Kuhlmann et al. (2003) which in turn are for anthropogenic emissions based on the EDGAR v2.0 inventory and for biomass burning and biofuel emissions based on Andrea and Merlet (2001) for biomass burning and biofuel emissions.

The terrestrial biogenic and oceanic emissions of NMVOC's are similar to the emissions in the inventory applied in the offline chemistry and tracer transport model MATCH-MPIC (Von Kuhlmann et al., 2003; Lawrence et al., 1999, hereafter MATCH). Although the biogenic emissions of terpenes and other reactive volatile organic compounds (ORVOC's) are calculated online (see Sect. 3.1), which could be used to provide the first-order estimate of biogenic CO and acetone emissions, we use the Guenther et al. (1995) global annual estimates of terpene and ORVOC emissions to arrive at the biogenic emission budgets of CO, acetone, methanol, alkanes, alkenes, aldehydes and acids. Oceanic emissions of CO and NMVOC's are also included. In addition, we apply the Bouwman (1997) dataset for ammonia (NH₃) emissions, made available through the Global Emission Inventory Activity (GEIA, Guenther et al., 1995).

To indicate the source strength of the anthropogenic and natural offline emissions

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we present in Table 2 the global annual technological, biofuel, biomass burning and total emissions (with the biogenic component). Note that biomass burning emissions refer to the year 2000, based on average activity data over the period 1997–2001. For specific years within this period the actual biomass burning estimates are now available as well but have not yet been applied here. Also shown are the budgets of a selection of offline terrestrial and oceanic biogenic emissions, including CO and NMVOC's, based on the MATCH model emission dataset, about which more details can be found in von Kuhlmann et al. (2003). The MATCH emission budgets are based on the 1990 EDGARv2 inventory for technological emissions and the biomass burning inventory by Galanter et al. (2000). Consequently, the comparison of global emission budgets indicates about the differences between the EDGARv2 and v3.2-F2000 as well as the biomass burning emissions. The largest differences occur in the NO_x, CO and C₄H₁₀ global emissions, with changes (relative to the MATCH inventory) of +33, -13 and +54%. The significantly larger total NO_x source is mostly due to larger ship emissions in EDGARv3.2-FT2000 (Eyring et al, 2005) compared to EDGARv2 and larger biomass burning emissions, both about 4 TgN yr⁻¹ larger compared to the MATCH inventory. These differences do not only reflect differences in inventory compilation but obviously also differences between 1990 and 2000. The smaller CO emissions in the presented inventory compared to the MATCH inventory are mostly due to an about 50 TgC yr⁻¹ smaller technological emission flux of the EDGARv3.2-FT2000 inventory compared to EDGARv2 whereas EDGARv3.2-FT2000 includes an about 20 TgC yr⁻¹ larger C₄H₁₀ emission flux compared to EDGARv2. The latter difference is explained by EDGARv3.2-FT2000 technological NMVOC emissions being about 30 TgC yr⁻¹ larger compared to EDGARv2 for an NMVOC speciation such that C₄H₁₀ makes up 70% of the technological NMVOC emissions. The biogenic NH₃ emission reflects the agricultural NH₃ inventory by Bouwman et al. (1997).

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3 Online emissions and dry deposition in EMDEP

3.1 Online emissions

For more details concerning the description of the emissions mostly relevant to aerosol modeling, including the emissions of marine DiMethylSulfide (DMS) and sea salt, terrestrial DMS, volcanic SO₂, mineral dust, and fossil fuel and biomass burning organic and black carbon, we refer to Dentener et al. (2006) and Stier et al. (2005). The implementation of a selection of algorithms to calculate the natural emissions of DMS, sea salt and mineral dust allows that the model can be used at different resolutions to study the sensitivity for a range of global flux estimates as well as controlling parameters such as the surface layer wind speed. The DMS emissions, which are calculated from the 10m wind speed and a climatology of the ocean water DMS concentrations according to Kettle and Andreae (2000), can be calculated by selecting one of different available parameterizations of the piston velocity according to Liss and Merlivat (1986), Wanninkhof (1992) or Nightingale (2000). Similarly, sea salt emissions can be simulated using algorithms based on Monahan (1986) or Schulz et al. (2004). Mineral dust emissions are simulated according to Balkanski et al. (2004) where alternative algorithms, e.g. based on Tegen et al. (2002), will become available in future updated model versions (see also Sect. 4). Other online calculated natural sources involve soil-biogenic NO_x and vegetation emissions of NMVOC's. Soil-biogenic NO_x emissions are calculated according to a modified version of the Yienger and Levy (1995) algorithm initially implemented and applied in the chemistry-climate model ECHAM4 to study the role of canopy processes for global soil-biogenic NO_x emissions (Ganzeveld et al., 2002). To actually consider the role of deposition, chemical transformations and turbulence within the canopy, a Canopy Reduction Factor (CRF) (Yienger and Levy (1995) is used as a default since the study by Ganzeveld et al. (2002) indicated that the ecosystem specific CRF provides reasonable estimates of the atmosphere-biosphere NO_x flux compared to an explicit multi-layer canopy model. One further modification relevant to mention is that in the version implemented in EMDEP not only the contribution to soil NO emis-

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sions by synthetic fertilizer application but also that due to animal manure is considered (A. F. Bouwman, personal communication, 2004). However, despite a near doubling of the N input, there is actually a decrease in the global annual NO emissions related to agricultural activities due to the fact that the fractional loss of the amount of fertilizer, which was previously set at 2.5% (Yienger and Levy, 1995), has been reduced to 0.7% (Bouwman et al., 2002).

Biogenic NMVOC emissions are calculated according to the Guenther et al. (1995) algorithm based on the global distribution of the 72 ecosystems of the Olson (1992) database. The latter, in combination with a Normalized Difference Vegetation Index (NDVI) satellite dataset, provides monthly estimates of the biomass (foliar density and Leaf Area Index, LAI), which are applied not only to calculate the emissions of isoprene, monoterpenes and other VOC's, but also dry deposition and soil-biogenic NO_x emissions.

Finally, in EMDEP there is the option to consider the emissions of methane (CH₄), which are, in the current implementation, not calculated online but inferred from the difference in the simulated surface layer and observed surface layer CH₄ concentrations. The latter are based on observations from the NOAA/CMDL cooperative air sampling network (Dlugokencky, 1994) processed using the methodology of Butler (2004).

3.2 Gaseous and aerosol dry deposition

A major limitation to introduce improved representations of the dry deposition process in large-scale models such ECHAM5/MESSy is the fact that dry deposition has only been directly measured for a small selection of gases and aerosols such as ozone (O₃), sulfur dioxide (SO₂) ammonia (NH₃) and sulfate (SO₄²⁻). Merely a limited number of dry deposition measurements of other species such as peroxides (hydrogen peroxide, H₂O₂), nitric acid (HNO₃) and other oxidized nitrogen species such as nitrogen (di)oxide (NO₂ and NO) have been performed, whereas measurements are also generally biased towards vegetation dry deposition. Observations of dry deposition over the oceans, snow-ice covered and bare soil surfaces are sparse, which not only limits a

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better quantification of the role of dry deposition in tracer budgets over these surfaces, but also the identification of controlling mechanisms required for the further advancement of model representations. The understanding of the mechanisms is mostly limited to that of turbulent transport and diffusion to the surface and some surface related mechanisms such as the stomatal uptake of gases such as O₃ and SO₂. For a comprehensive overview of the status of dry deposition research we refer to Wesely and Hicks (2000).

To at least provide first-order estimates of the potential role of dry deposition of gases for which no direct dry deposition measurements are available Wesely (1989) introduced a commonly applied approach by which surface uptake resistances are estimated based on solubility and reactivity by scaling to those of O₃ and SO₂. A non-soluble but reactive gas is expected to deposit like O₃ whereas a more soluble and less reactive gas should deposit like SO₂. The default selection of gases included in the dry deposition scheme of EMDEP (and DRYDEP) are O₃, SO₂, HNO₃, NO and NO₂, complemented with bulk sulfate aerosol dry deposition (Ganzeveld and Lelieveld, 1995; Ganzeveld et al., 1998) for studies that include sulfur chemistry but do not explicitly resolve aerosol mass size distributions (see below).

Dry deposition fluxes are calculated as the product of the surface layer concentration and the dry deposition velocity, which reflects the efficiency of the transport to- and destruction at the surface. The dry deposition velocity (V_d) is calculated according to the so-called "big leaf" approach where the vegetation canopy, or other surface, is represented as a bulk substrate without considering in detail the different removal processes at different levels in the canopy. In analogy to the resistance against electrical conductivity, V_d is calculated as the reciprocal of three serial resistances that represent the turbulent transport to the surface, molecular diffusion through a quasi-laminar boundary layer and finally uptake and destruction by the surface. The turbulent transport or so-called aerodynamic resistance (R_a) is calculated from the wind speed, surface roughness and atmospheric stability whereas the quasi-laminar boundary layer resistance (R_b) is calculated from the same parameters and the trace gas specific molec-

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ular diffusivity. The vegetation surface uptake resistance (R_c) is calculated from the leaf/needle resistance, scaled up to the canopy scale by using the Leaf Area Index (LAI, single-sided leaf area per soil area), a species-specific soil uptake resistance and a resistance that accounts for turbulent transport through the canopy to the soil. The leaf resistance in EMDEP is calculated from the stomatal uptake resistance, generally being provided by the driver model (e.g., in ECHAM5 to calculate evapotranspiration according to Sellers, 1986), corrected for differences in diffusivity between H₂O and the gas, and species-specific mesophyll and cuticular uptake resistances. For details concerning the selected values for the default selection of gases of the dry deposition scheme, we refer to Ganzeveld and Lelieveld (1995) and Ganzeveld et al. (1998). For any other trace gas to be added to the dry deposition scheme, e.g., oxygenated compounds or acids, the only modification to be made is to define the tracer specific Henry Law's coefficient [m atm^{-1}], a reactivity coefficient [0, 0.1, 1] (with 0 for species that are poorly reactive on surfaces, e.g. acetaldehyde, 0.1 for semi-reactive species, e.g., peroxyacetylnitrate and 1 for highly reactive species, e.g., O₃) and its molar mass (see Appendix A).

A main new feature of the dry deposition calculations in EMDEP is the online calculation of aerosol dry deposition as a function of aerosol properties, meteorology and surface cover. Aerosol dry deposition velocities (V_{daerosol}) and fluxes are calculated with a modified version of the aerosol dry deposition model that has been used to develop a parameterization of the bulk sulfate aerosol dry deposition in the chemistry-climate model ECHAM4 (Ganzeveld et al., 1998). The online aerosol dry deposition code has also been implemented and applied in the aerosol-climate model ECHAM5-HAM (Stier et al., 2005).

The model calculates V_{daerosol} over land and over sea considering turbulence, Brownian diffusion, impaction and sedimentation as a function of radius and density. Over sea the effect of particle growth on dry deposition due to the large relative humidity close to the sea surface is accounted for as well as bubble bursting that enhances dry deposition through the breakdown of the laminar boundary layer and the scaveng-

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ing of the particles by sea spray (Hummelshøj, 1992). The main modification in the aerosol dry deposition model is the calculation of the vegetation dry deposition velocities, whereas in the Ganzeveld et al. (1998) model we applied a parameterization of sulfate aerosol dry deposition as a function of the friction velocity for stable and unstable conditions developed by Wesely et al. (1985). In the new model aerosol dry deposition to the vegetation is calculated according to Gallagher et al. (2002), which is a further modification of a parameterization of aerosol dry deposition by Slinn (1982). In contrast to Zhang et al. (2001) we have selected a smallest and a largest collector size of $10\ \mu\text{m}$ and $1\ \text{mm}$, respectively, independent of land cover, partly associated with the large uncertainty involved in the definition of these parameters. The largest collector size is used to calculate the Stokes number, which is not only used to calculate impaction but also a rebound correction factor, whereas the small collector size is used to calculate the interception collection efficiency (Gallagher et al., 2002). More details concerning the use of the aerosol dry deposition model are found in the Appendix A.

4 Global emission budgets and dry deposition

Table 3 shows the global annual emissions for a selection of reactive trace gases and aerosols simulated with EMDEP, applied in ECHAM5/MESSy (version 1.1), for an 1-year climatological model integration (1 month spin-up, reference year 1987–1988) with the T42L19 (i.e., 2.8° grid resolution, 19 vertical levels) resolution of ECHAM5. The global isoprene (C_5H_8) emission flux of $607\ \text{TgC yr}^{-1}$ is significantly larger compared to the estimate by Guenther et al. (1995) and the online simulated source of $500\ \text{TgC yr}^{-1}$ with the chemistry-climate model ECHAM4 (Ganzeveld et al., 2002). The latter was calculated at a T31L19 spatial resolution with significantly larger estimates of vegetation biomass compared to that used in the current implementation of biogenic emission and dry deposition calculations in EMDEP. In ECHAM4 the maximum Leaf Area Index (LAI) for tropical forest even exceeded $10\ \text{m}^2\ \text{m}^{-2}$ whereas observations show maximum values up to about 6–7. Consequently we apply a function that provides an

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asymptotic increase in LAI to a maximum of 7 for the maximum observed Normalized Differential Vegetation Index (NDVI) (Ganzeveld et al., 2002). In addition, we have consistently corrected the foliar density (leaf biomass in g m^{-2}) which is used to calculate the biogenic VOC emissions. Interestingly, a relative increase in the simulated fraction of sunlit leaves partly compensates this decrease in foliar density. The budgets of the monoterpene ($\text{C}_{10}\text{H}_{16}$) and ORVOC's emissions of 143 and $255\ \text{TgC yr}^{-1}$, that are calculated by EMDEP using ECHAM5's surface temperature, agree reasonable well with the estimates by Guenther et al. (1995) of 127 and $260\ \text{TgC yr}^{-1}$, respectively. This suggests that EMDEP's larger isoprene emission flux, compared to the Guenther et al. (1995) inventory, is mostly due to differences between the EMDEP online simulated surface and in-canopy radiation regime, using ECHAM5's surface net radiation, compared to the offline approach by Guenther et al. (1995), and not so much due to differences in temperature. Note that the simulated isoprene emission budget is nearly a factor two larger compared global isoprene flux of $350\ \text{TgC yr}^{-1}$ by von Kuhlmann et al. (2003), which applied a global scaling factor to the online calculated emission fluxes and which is a common approach widely applied in global scale atmospheric chemistry model analysis. This is generally being justified by the fact that use of the $500\ \text{TgC yr}^{-1}$ inventory results in simulated tropical boundary layer isoprene concentrations that are significantly larger compared to observations. This misrepresentation is likely also due to the models representations of boundary layer mixing, dry deposition (of isoprene and the oxidation products) and chemical destruction (von Kuhlmann et al., 2004) and not only a possible overestimation of emissions. Interestingly, a new inventory of the global isoprene emissions with the MEGAN model (Guenther et al., 2006), which incorporates the newly acquired information on biological, physical and chemical drivers since the release of the Guenther et al. (1995) inventory, provides a global isoprene emission budget of $\sim 530\ \text{TgC yr}^{-1}$.

The LAI is also used in the calculation of the Canopy Reduction Factor (CRF), which provides estimates of the soil-biogenic NO_x emissions comparable to those calculated with an explicit multi-layer canopy model implemented in ECHAM4 (Ganzeveld et al.,

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2002). Table 3 shows that the online simulated global soil NO flux of 12.4 TgN yr^{-1} is reduced by about 35% with maximum decreases in tropical forested regions up to about 75%. Pulsing, which is the enhancement in emissions in response to rainfall after a period of drought, enhances the soil NO emission by about 2 TgN yr^{-1} whereas
5 fertilizer application and animal manure contribute about 0.7 TgN yr^{-1} . The latter is about the half the contribution by fertilizers to global soil NO emissions inferred from a statistical modeling approach by Bouwman et al. (2002). This can be explained by the fact that the fertilizer application is corrected for a prescribed global distribution of cultivation intensity determining the timing of fertilizer application. Consequently, we
10 apply in EMDEP a fractional fertilizer loss of 1.5% to arrive at a contribution by fertilizers to global soil-biogenic NO_x emissions of 1.5 TgN yr^{-1} .

The global annual DMS emission flux, calculated in this simulation with the Liss and Merlivat (1986) parameterization for the piston velocity, is 17.3 TgS yr^{-1} . This is about
15 6 TgS less compared to the number reported by Stier et al. (2005). This also reflects the sensitivity to the parameterization applied for the piston velocity (Nightingale, 2000) that was used by Stier et al. (2005). Note that in addition to this major biogenic source of sulfur, terrestrial DMS and volcanic SO_2 emissions, amounting to about 1 TgS yr^{-1} (Spiro et al., 1992) and 8 TgS yr^{-1} (Graf et al., 1997), respectively, are also included in EMDEP. The sea salt emission flux of $5906 \text{ Tg sea salt yr}^{-1}$, calculated according
20 to Schulz et al. (2004), is larger compared to the emissions budgets presented by Stier et al. (2005) (5050 Tg yr^{-1}). One possible explanation for this discrepancy are differences in the surface wind speed as the emissions were obtained with the T42L19 model resolution whereas Stier et al. (2005) applied an T63L31 (ECMWF) resolution. In addition, our emission budget reflects an ECHAM5 climatological simulation whereas
25 the results by Stier et al. (2005) reflect emissions for a simulation where ECHAM5 has been nudged towards the ECMWF ERA40 reanalysis data. The nudging results in a slightly smaller sea salt emission burden compared to climatological simulations due to a relative decrease in the wind speeds. This has an even more pronounced impact on the simulated mineral dust emissions, which partly explains an calculated

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global annual emission flux according to our implementation of the scheme according to Balkanski et al. (2004) with EMDEP of 1432 Tg yr^{-1} , about 680 Tg larger compared than that presented by Stier et al. (2005). This is consistent with other simulations of
5 mineral dust emissions, calculated according to Tegen et al. (2002), which also showed a significant decrease in the nudged model simulations compared to a climatological simulation with the same model resolution.

In Table 3 we not only present global emission burden for biogenic NO_x , VOC, DMS and sea-salt and mineral dust aerosol mass for the T42L19 model resolutions but also include the budgets for the T21L19 ($\sim 5.6^\circ$) and T63L19 ($\sim 1.8^\circ$) model resolution. Note
10 that for the T63 resolution, the 31 layer (L31) vertical structure is generally recommended, but here we have consistently applied the L19 vertical resolution. In general the emissions increase with an increase in resolution from T21 to T63 with relative increases (T63-T21 relative to T42 emissions) up to about 10% for isoprene and DMS and about 20% for monoterpenes, ORVOC and sea salt, whereas soil-biogenic NO_x
15 emissions actually show a small decrease. These changes reflect differences in the simulated net surface radiation, temperature, wind speed, precipitation and soil moisture for the different model resolutions.

The mineral dust emissions are extremely sensitive to the applied resolution suggesting that the implementation of the emission algorithm by Balkanski et al. (2004) in
20 EMDEP should only be applied for the T42 resolution since only for this resolution it produces emissions fluxes comparable to those reported by Tegen et al. (2002) in a climatological run with ECHAM5. For other resolutions, for example the T63 simulation we simulate a global emission strength $>8000 \text{ Tg yr}^{-1}$ mostly due to very large emissions in Pakistan and north of the Himalaya contributing $>85\%$ of the global emissions also due to simulated monthly mean 10 m wind speeds exceeding 20 m s^{-1} . Alternative
25 algorithms to simulate mineral dust emissions, e.g., the one by Tegen et al. (2002), potentially being less sensitive to the applied resolution will be implemented in EMDEP in the near future.

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4.1 Dry deposition and emission height profiles

An important feature of the application of the emission inventory in OFFLEM is the consideration of emission height profiles for tracer transport calculations. To indicate the sensitivity of the simulated trace gas exchanges to the applied height profiles, we present in Figs. 1a and b the relative difference in annual mean SO₂ and O₃ dry deposition fluxes between a simulation with ECHAM5/MESSy in which the precursors have been released at the surface and one applying the vertical redistribution of emissions up to 800m altitude, as described above (Table 1). These results reflect a simulation with a setup similar to an that used for an evaluation of ECHAM5/MESSy (Jöckel et al., 2006²) including tracer transport, gas-phase chemistry (Sander et al., 2005) and scavenging (Tost et al., 2006) except of a different vertical resolution (L19 versus L90), no nudging and using EMDEP instead of the submodels ONLEM and DRYDEP. The simulated SO₂ dry deposition fluxes resulting from emitting all SO₂ in the surface layer are up to 80% higher in the vicinity of the sources over the continents in low and middle latitudes, whereas at larger distances from the sources e.g., over the eastern Sahara and Arctic regions, large relative (but small absolute) decreases in dry deposition occur as a result of the decreased supply by long-range transport. Figure 1b shows a relative smaller sensitivity of O₃ deposition to the vertical emissions profiles of the precursor gases CO, NO_x and NMVOC's compared to SO₂. There are relative decreases >10% in O₃ dry deposition in the main source regions with large technological and biomass burning emissions when those emissions are emitted away from the surface whereas the more effective long-range transport of the precursor gases explains the increases in the O₃ deposition up to 15% in the northern hemisphere high latitude regions. The

²Jöckel, P., Tost, H., Pozzer, A., Brühl, Ch., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkmann, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: Consistent simulation of ozone from the surface to the mesosphere, Atmos. Chem. Phys. Discuss., submitted, 2006.

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application of the emission height profiles results in a relative decrease in the global annual SO₂ dry deposition flux up to about 30% compared to surface emissions simulations associated with a significant decrease in surface layer concentrations. This results in a significant increase in the SO₂ lifetime and it enhances the long-range transport of sulfur pollution. This sensitivity of dry deposition to the emission height is also relevant to the evaluation of ECHAM5/MESSy, addressed in our first extensive model evaluation publication (Jöckel et al., 2006²), which will also include a direct comparison of observed and simulated surface concentrations and dry deposition fluxes.

5 Summary/conclusions

We have presented the state-of-the-art representation of surface reactive trace gas and aerosol exchanges describing an anthropogenic and natural offline emission inventory, distributed with the first official release of MESSy, and MESSy's submodel EMDEP. This includes a discussion of the mechanisms underlying the emissions and dry deposition calculations, global online calculated emission budgets with EMDEP and an assessment of the sensitivity of dry deposition to the emission height profiles. This emission height is included in the offline emission inventory that includes the EDGARv3.2-FT2000 technological and biomass burning emission and prescribed terrestrial and oceanic emissions. The large sensitivity of the simulated dry deposition fluxes to the emission heights that are based on a limited amount of studies and expert assumptions, and therefore accompanied with large uncertainty, indicates the priority of a better understanding and representation of the effective emission heights in models such as MESSy.

The online calculated gaseous and aerosol natural and biogenic emissions use the driver models micrometeorological and surface cover parameters complemented with land cover and land use properties inferred from satellite data and global scale ecosystem and land use datasets.

Aerosol dry deposition is calculated considering turbulence, Brownian diffusion, im-

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paction, interception and sedimentation as a function of the models micrometeorology and land cover and aerosol radius. Gaseous dry deposition is calculated online for many species in addition to those for which dry deposition is known to be an important sink based on scarcely available measurement data. The scheme can be easily
5 extended to other species expected to be removed by dry deposition based on their chemical and physical properties, using the approach proposed by Wesely (1989). However, it should be noted that recent studies indicate that first-order estimates of the surface uptake resistances according to that approach seem to result in a significant underestimation of dry deposition of a selection of oxygenated species, measured
10 in the tropical forest of Costa Rica (Karl et al., 2004) and peroxides (Ganzeveld et al., 2006). This suggests that the approach needs modification which, however, is severely limited by the availability of observational data. Actually, the implementation of these first-order estimates of dry deposition in models such as EMDEP can help obtain indications about the potential relevance of the dry deposition process, and consequently
15 provide suggestions to the experimental community on the priority of further research focusing on dry deposition.

We have demonstrated the large impact of applying the EDGARv3.2-FT2000 recommended emission heights; for example, on the SO₂ dry deposition flux. This is not only relevant to atmospheric chemistry with respect to a change in the lifetime and
20 atmospheric burden but also to the evaluation of models such as ECHAM5/MESSy. This evaluation relies on the comparison of observed and simulated concentrations and fluxes, including many surface observations, obviously being strongly affected by emission and deposition parameterisations. This underscores the need for improved and consistent representations of the spatial and temporal variability of surface exchange processes in Earth system models, including e.g. diel cycles in technological
25 and biomass burning emissions.

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Appendix A

The emission and dry deposition calculations in EMDEP are included in three different sub-submodels identified by the names EMDEP_XTSURF, EMDEP_DRYDEP and EMDEP_EMIS. The EMDEP_XTSURF routines deal with the initialization of land cover
5 and land use parameters, the tracer specific surface uptake resistances, and the actual calculations of the gaseous and aerosol “big leaf” dry deposition velocities from those surface resistances. These dry deposition velocities are then subsequently applied in EMDEP_DRYDEP to actually calculate the dry deposition fluxes, where alternative algorithms, e.g., based on prescribed deposition velocities, can also be included. In
10 EMDEP_EMIS the online emission are calculated from the driver model parameters and the land cover and land use parameters initialized in EMDEP_XTSURF. Note that the presented budgets reflect calculations with the EMDEP model, which is continuously further developed. Consequently differences in calculated emission and deposition budgets between EMDEP and ONLEM and DRYDEP are to be expected. Further
15 improvements in the online emission- and dry deposition calculations in EMDEP are intended to be implemented later into ONLEM and DRYDEP, respectively, as well, after they have been thoroughly tested. One of the future extensions of EMDEP will be the release of a version that includes a multi-layer canopy model to study the role of in-canopy interactions between biogenic emissions, dry deposition, chemistry and turbulent transport, previously used to study global soil-biogenic NO_x exchanges (Ganzeveld
20 et al., 2002).

A1 Adding a tracer to the dry deposition calculations

In order to include the explicit dry deposition calculations tracer specific properties need to set in the tracer definition. If the tracer is defined by the submodel MECCA (Sander
25 et al., 2005) in the file tracdef.tex, the dry deposition calculation switch NDRYDEP has to be set to ON. In addition, for gases the molecular mass, Henry law's coefficient [mol l⁻¹] and a reactivity coefficient must be defined. The latter has only three values

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with 0 reflecting a non-reactive gas (e.g., acetone), 0.1 for an intermediate reactive gas (e.g., PAN), and 1 for a reactive gas (e.g., monoterpenes). Otherwise, these tracer properties must be set by definition of the respective tracer being considered in the various submodels. For aerosol dry deposition calculations information is required on the aerosol density [kg m^{-3}] and the aerosol number and mass radii [m]. The aerosol dry deposition code is programmed to simulate aerosol dry deposition for the implementation of a modal aerosol model conducting the calculations for the number of modes that are characterized by a median or mean mode radius, the sigma and wet aerosol density. However, the model can easily be modified for application in a bin-resolving aerosol model or in a bulk aerosol model by simply defining representative bulk aerosol properties. By default the code calculates V_{daerosol} for a more computational efficient method using the modal mean number and mass radii but the option exists to calculate V_{daerosol} integrating over the number- and mass size distributions.

A2 Integration in vertical diffusion scheme

The calculation of the concentration change due to surface exchanges can be done according to two different approaches which allow studying the possible impact of operator splitting involving surface exchanges and vertical turbulent diffusion. In the default set-up (set by the switch lturb_emis in the EMDEP namelist) the calculated surface exchange fluxes [$\text{molecules m}^{-2} \text{s}^{-1}$] are used as the lower boundary condition in the vertical diffusion scheme of ECHAM5 (parameter ZXTEMS, [$\text{mol mol}^{-1} \text{kg m}^{-2} \text{s}^{-1}$] for tracer defined in volume mixing ratio). The alternative is to assign fluxes to the tracer tendency (parameter PXTTE, [$\text{mol mol}^{-1} \text{s}^{-1}$]).

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Table 1. Emission height profiles for the EDGARv3.2-FT2000 emission source categories, distinguishing 6 emission heights, 45, 140, 240 400 600 and 800m, where the numbers in the columns reflect the percentage of the total emission flux assigned to the specific height. The info source EMEP indicates profiles based on the EMEP model whereas AA indicates that these profiles are based on an arbitrarily assumption.

Source category	Source name	45 m	140 m	240 m	400 m	600 m	800 m	Info source
B10	Industry	0	4	19	41	30	6	EMEP
B20	Power generation	0	0	8	46	29	17	EMEP
B30	Charcoal prod.	100	0	0	0	0	0	AA
B40	RCO	50	50	0	0	0	0	EMEP
F10	Industry	0	4	19	41	30	6	EMEP
F20	Power generation	0	0	8	46	29	17	EMEP
F30	OTS	0	4	19	41	30	6	EMEP
F40	RCO	50	50	0	0	0	0	EMEP
F51	Road transport	100	0	0	0	0	0	EMEP
F54	Non-Road transport	100	0	0	0	0	0	EMEP
F58	Inter. Shipping	50	50	0	0	0	0	AA
F60	Chem. Feedstock	100	0	0	0	0	0	AA
F70	Coal production	90	10	0	0	0	0	EMEP
F80	Oil production	90	10	0	0	0	0	EMEP
F90	Gas production	90	10	0	0	0	0	EMEP
I10	Iron and steel	90	10	0	0	0	0	EMEP
I20	Non-ferrous metals	90	10	0	0	0	0	EMEP
I30	Chemicals	90	10	0	0	0	0	EMEP
I40	Building materials	90	10	0	0	0	0	EMEP
I50	Pulp and paper	90	10	0	0	0	0	EMEP
I60	Food, bev, tobacco	90	10	0	0	0	0	EMEP
I70	Solvents	100	0	0	0	0	0	EMEP
I90	Misc. industry	100	0	0	0	0	0	EMEP
L10	Arable land	100	0	0	0	0	0	EMEP
L15	Rice cultivation	100	0	0	0	0	0	EMEP
L20	Animals	100	0	0	0	0	0	EMEP
L30	Animal waste man.	100	0	0	0	0	0	EMEP
L41	Deforestation	0	100	0	0	0	0	AA
L42	Savannah burning	0	100	0	0	0	0	AA
L43	Agric. waste burning	0	100	0	0	0	0	AA
L44	Vegetation fires	0	100	0	0	0	0	AA
L45	Deforestation indirect	0	100	0	0	0	0	AA
L50	Crop products	100	0	0	0	0	0	AA
L70	Indirect N ₂ O	100	0	0	0	0	0	AA
W10	Landfills	100	0	0	0	0	0	AA
W20	Wastewater treatment	10	15	40	35	0	0	EMEP
W30	Human waste disposal	10	15	40	35	0	0	EMEP
W40	Waste incineration	10	15	40	35	0	0	EMEP
W50	Misc. waste handling	10	15	40	35	0	0	EMEP

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Table 2. Global annual offline emissions per source category and total emission flux for the emission inventory and the one applied in MATCH.

Chemical Compound	Technological	Biofuel	Biomass burning	Biogenic	Total	MATCH ¹
NO _x	31.4	2.5	9.3	0.0 ²	43.1	32.4 ³
CO	120.7	107.3	193.8	48.2	470.1	541
SO ₂	73.2	3.1	1.3		76.0	
C ₂ H ₄	3.3	4.4	4.1	10.7	22.6	25.1
C ₂ H ₆	5.0	2.3	2.2	0.4	10.0	10.2
C ₃ H ₆	1.7	2.0	1.9	3.0	8.5	9.6
C ₃ H ₈	8.1	0.7	0.7	0.3	9.8	7.3
C ₄ H ₁₀	59.9	0.1	0.9	0.3	62.1	40.4
CH ₃ CHO	0.0	1.1	1.1	0.0	2.2	2.5
CH ₃ COCH ₃	1.9	1.2	1.1	25.6	29.8	28.6
CH ₃ COOH	0.0	2.6	2.5	1.4	6.5	7.2
CH ₃ OH	1.2	2.5	2.3	23.2	29.1	28.7
HCHO	0.4	1.4	1.3	0.0	3.1	3.6
HCOOH	0.0	0.9	0.9	1.5	3.3	3.5
MEK	2.8	2.9	2.8	0.0	8.5	8.4
NH ₃				53.9	53.9	

Units are in Tg C/N/S

¹ von Kuhlmann et al. (2003)

² Calculated online

³ Total technological and biomass burning, excluding the soil-biogenic, aircraft and lightning contribution

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Table 3. Global annual online emissions calculated by EMDEP, applied in ECHAM5/MESy, as function of three different model resolutions and the relative difference between the high and coarse resolution simulations resembling an 1-year integration from 1 February 1987 until 31 January 1988. The budgets are based on a 25-h out-put frequency of the instantaneous emissions.

Compound	T42	T21	T63	T63-T21 [%] ¹
C ₅ H ₁₀	607.1	556.2	626.4	12.6
C ₁₀ H ₁₆	143.1	128.6	148.5	15.5
ORVOC's	254.6	227.2	266.2	17.2
NO _x , soil	12.4	12.7	12.3	-3.1
NO _x , canopy	8.4	8.6	8.3	-3.5
DMS	17.3	16.3	17.6	8.0
Dust	1432 ²	-	-	-
Seasalt	5906 (64, 5842) ³	5199 (56, 5143)	6258 (68, 6190)	

Units are in Tg C/N/S/dust/sea salt

¹ Relative difference relative to T21 budget

² The dust emissions appear to be highly resolution dependent and are only presented for the T42 resolution, the resolution for which the emission scheme has been developed

³ The numbers in parentheses reflect accumulation- and coarse mode emissions, respectively

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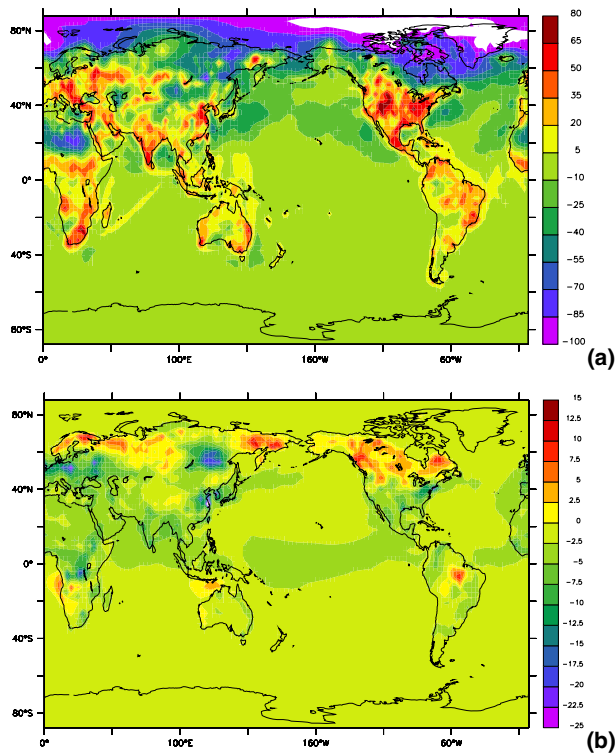


Fig. 1. Relative change in annual mean SO₂ (a) and O₃ (b) dry deposition fluxes due to emitting all SO₂ and O₃ precursors in the surface layer instead of including the recommended emission heights for technological and biomass burning emissions according to EDGARv3.2-FT2000. The white areas in the Arctic in (a) denote relative differences $\leq 100\%$.