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Parameterization of wet scavenging and heterogeneous glyoxal sinks in the IMAGESv2 global chemistry-transport model

Supplement to "The continental source of glyoxal estimated by the synergistic use of spaceborne measurements and inverse modelling"

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1 Wet Scavenging Parameterization

The parameterization adopted in IMAGESv2 for wet scavenging of gases and aerosols is based on cloud and precipitation fields (analyses and forecasts) provided by the European Centre for Medium-range Weather Forecasts (ECMWF). The treatment of washout by convective precipitation (section 1.1) is included in the convective transport scheme, whereas scavenging in and below large-scale stratiform clouds (section 1.2) is represented as a first-order loss process. The effect of gravitational settling of (non-precipitating) cirrus clouds (section 1.3) is also considered for HNO_3 and for aerosols.

1.1 Scavenging by Convective Precipitation

As pointed out by Balkanski et al. (1993), scavenging in convective clouds is better represented as part of the model operator for convective transport, rather than separately as a first-order loss rate. The numerical implementation of convective transport in IMAGES follows a probabilistic approach first proposed by Chatfield and Crutzen (1984) and further developed by Costen et al. (1988) and Müller and Brasseur (1995). Transition probability densities (TPDs) are defined for the ascendence (updraft) of boundary layer air to the different free tropospheric levels, as well as for the compensating subsidence (downdraft) of air from each level j to the level directly underneath, $j-1$. The updraft TPDs for insoluble chemical compounds are obtained from the distributions of updraft mass fluxes provided by the ECMWF analyses. The downdraft TPDs are calculated from the mass conservation equation (Müller and Brasseur, 1995). Untill 2001, daily ERA40 fields are used; after this date, monthly climatological mass fluxes are used, based on the 1996-2001 ERA40 values. Monthly averaged diurnal cycles for these fluxes are also obtained from the 3-hourly fields of the ERA40 analyses, and used in the diurnal calculations with the IMAGES model. Time-stepping is implicit. This transport scheme is stable and mass-conserving for insoluble tracers.

Now soluble compounds entering convective updraft are partly or entirely incorporated into (liquid or solid) water condensates, a fraction of which end their life as precipitation at the earth's surface. Therefore the updraft TPDs for a soluble compound i are multiplied by $(1 - E \cdot s_i)$, where E is the precipitation efficiency, defined as the ratio of precipitation to the total water vapor inflow, and s_i is a species-specific scavenging efficiency ratio depending on the compound solubility in water and on the retention efficency of the tracer during conversion of liquid cloudwater to precipitation (Mari et al., 2000). The product $E \cdot s_i$, the rained out fraction, is the scavenging efficiency defined by Mari et al. (2000). The precipitation efficiency (E) is parameterized according to Fritsch and Chappell (1980) as a function of the vertical shear of the horizontal wind (V) between the base and top of the convective cloud:

$$E = \min(0.9, 1.591 - 0.639\frac{\Delta V}{\Delta z} + 0.0953(\frac{\Delta V}{\Delta z})^2 - 0.00496(\frac{\Delta V}{\Delta z})^3), \tag{1}$$

where $\frac{\Delta V}{\Delta z}$ is expressed in 10^{-3} s^{-1}. The cloud top altitude is taken as the uppermost model level with non-zero updraft flux according to ECMWF.

The scavenging efficiency ratio (s_i) is taken equal to 1 for the very soluble nitric acid, 0.9 for hydrophilic aerosols (e.g. SO_4 and SOA) (Textor et. al, 2006; Stier et al., 2005), and 0.36 for SO_2, based on results by Mari et al. (2000). For other soluble gases, it is taken equal to the dissolved fraction of the total concentration, calculated for a typical liquid water content in convective cells (L_{Cb} = 2 g m^{-3}) and for a temperature (T) of 280 K:

$$s_i = 1/(1 + \frac{1}{{K^H}_i \cdot L_{Cb} \cdot R \cdot T}, \tag{2}$$

where ${K^H}_i$ is the Henry's law constant of species i (M atm^{-1}), obtained from the compilation by R. Sander (http://www.mpch-mainz.mpg.de/ sander/res/henry.html) and R is the gas constant.

1.2 Scavenging by Large-Scale Precipitation

1.2.1 General Formulation

Scavenging by large-scale (stratiform) clouds is represented as a first-order process. It is a subgrid phenomenon, since only a fraction of a given model grid cell is subject to precipitation scavenging. For a weakly soluble compound (e.g. CH_2O), scavenging does not cause large differences between the gridscale concentration of the compound and its concentration within the precipitating cloud (or in the precipitation region underneath), so that the effective loss rate due to scavenging can be written

$$\lambda = a_{in} \cdot \lambda_{in} + a_{below} \cdot \lambda_{below} \tag{3}$$

where λ (s^{-1}) is the loss rate applicable to the grid cell concentration of the compound, a_{in} and a_{below} are the fractional areas where in-cloud and below-cloud scavenging take place, respectively, and λ_{in} and λ_{below} are the loss rates (s^{-1}) due to in-cloud and below-cloud scavenging, respectively. In-cloud scavenging (also called washout) is associated with the conversion of cloud water into precipitation, whereas below-cloud scavenging (rainout) is due to the absorption of chemical compounds by falling rain or snow. Note that rainout occurs (not only below, but) also within precipitating clouds, and therefore $a_{below} > a_{in}$.

In the more general case, however, scavenging depletes the compound concentration in the scavenging area, and Eq. (3) overestimates the effective sink applicable to the grid-averaged concentration of the chemical compound. Different approaches have been proposed to account for this effect, among which the popular methods introduced by Rodhe and Grandell (1972) (RG72) and Giorgi and Chameides (1986). In both methods, λ depends on the average duration of precipitation events (t_p). For example, RG72 expresses the grid-average loss rate as

$$\lambda = \frac{a_{in} \cdot \lambda_{in}}{1 + (1 - a_{in})^2 \cdot t_p \cdot \lambda_{in}}, \tag{4}$$

where below-cloud scavenging has been neglected for simplicity. The RG72 parameterization predicts residence times of chemical compounds in good agreement with the results of a time-dependent numerical model using randomly distributed precipitation events (Stewart et. al, 1990). t_p is poorly constrained from the observations, with typical values over the northeast United States ranging between 4 hours in summer and 7 hours during winter (Thorp, 1986), whereas larger values have been reported over Russia in wintertime (Feigelson, 1984). We parameterize the seasonality of t_p (seconds) by the expression

$$t_p = 3600 \cdot (4 + \min(14, \max(0, 7 \cdot \frac{288.15 - T_s}{20}))), \tag{5}$$

where T_s is the surface temperature (in K). This parameter probably exhibits a much larger variability in the real atmosphere, and can be expected to be a large source of error in the overall scavenging parameterization.

We generalize the formulation of RG72 (Eq. (4)) in order to account for both in-cloud and below-cloud scavenging:

$$\lambda = \frac{a_{in} \cdot (\lambda_{in} + \lambda_{below})}{1 + (1 - a_{in})^2 \cdot t_p \cdot (\lambda_{in} + \lambda_{below})} + \frac{(a_{below} - a_{in}) \cdot \lambda_{below}}{1 + (1 - (a_{below} - a_{in}))^2 \cdot t_p \cdot \lambda_{below}} \tag{6}$$

The scavenging rate is expressed as the sum of two independent terms, one for the total scavenging within precipitating clouds, and the other for the scavenging by falling precipitation outside these clouds.

1.2.2 Precipitating Cloud Fractions

The determination of both a_{in}, the fraction covered by precipitating clouds, and a_{below}, the fraction covered by falling precipitation, relies on analysed cloud and large-scale precipitation fields provided by the European Centre for Medium-Range Weather Forecasts. In the ECMWF model, the conversion of condensed water to rain or snow follows the parameterization of Sundqvist (1978) (see also Tiedtke (1982)), where the generation of precipitation (G_{prec}, in g kg^{-1} s^{-1}) is calculated with

$$G_{prec} = a_{cld} \cdot c_0 \cdot L_{cld} \cdot [1 - \exp(-(\frac{L_{cld}}{L_{crit}})^2)], \tag{7}$$

where a_{cld} is the cloud fraction, c_0 (s^{-1}) is a characteristic rate for the conversion of condensed water to precipitation, L_{cld} is the cloud water content (g kg^{-1}), and L_{crit} is a typical cloud water content at which the generation of precipitation begins to be efficient. c_0 and L_{crit} depend on temperature and on the local cloudy precipitation rate (Sundqvist, 1978; European Centre for Medium-Range Weather Forecasts, 2007). Note that Eq. (7) implies that all clouds precipitate in the ECMWF formulation, i.e. $a_{in} = a_{cld}$. Since this would lead to a large overestimation of precipitation fractions observed at meteorological stations (see below), we calculate a_{in} by re-writing Eq. (7) as

$$G_{prec} = a_{in} \cdot \frac{c_0 \cdot L_{cld}}{\alpha_1} \tag{8}$$

with

$$a_{in} = a_{cld} \cdot \alpha_1 \cdot [1 - \exp(-(\frac{L_{cld}}{L_{crit}})^2)] \tag{9}$$

which expresses that only a fraction of the cloud precipitates, and that this fraction increases with the condensed water content of the cloud. The coefficient α_1 is an adjustable parameter (see below). The maximum overlap assumption (European Centre for Medium-Range Weather Forecasts, 2007) relates the precipitating cloud fraction (i. e. the fraction of the grid-cell covered by precipitating clouds, ${a^j}_{in}$, where j denotes the vertical level index) to the precipitation fraction (i. e. the fraction of the grid-cell where precipitation occurs, ${a^j}_{precip}$):

$$(1 - {a^j}_{precip}) = (1 - {a^{j+1}}_{precip}) \cdot \frac{1 - \max({a^j}_{in}, {a^{j+1}}_{in})}{1 - \min({a^{j+1}}_{in}, 1 - \delta)} \tag{10}$$

with δ being a tiny number set to 10^{-6}. The factor α_1 is adjusted so that the surface-level precipitation fraction ${a^1}_{precip}$ matches the observed average precipitation fraction over the N-E United States (Thorp, 1986). Due to an upgrade of the Integrated Forecast System (IFS) of ECMWF in early 2005, the adjustment is year-dependent: before 2005, α_1 = 0.25 and 0.75 for mixed-phased clouds and ice clouds, respectively, whereas since 2005, α_1 = 0.5 and 1, respectively.

1.2.3 In-cloud scavenging rates

In-cloud scavenging (rainout) follows from the conversion of condensed water into precipitation. The in-cloud scavenging rate is equal to the condensed phase fraction (r_{cond}) of the species considered multiplied by the rate of conversion of cloud water into precipitation:

$$\lambda_{in} = r_{cond} \cdot \frac{Q}{L}, \tag{11}$$

where Q is the water condensation rate (kg m^{-3} s^{-1}) and L is the condensed water content (kg m^{-3}), both provided by ECMWF. The condensed fraction for gases in liquid clouds is calculated according to Henry's law (similar to Equation 2). For nitric acid, the condensed fraction in both liquid and ice clouds is always equal to 1. Riming (freezing of liquid droplets) being the main process responsible for the conversion of liquid water to precipitation in mixed-phase clouds, the condensed fraction is multiplied by the retention efficiency R (i. e. the fraction remaining in the condensed phase when the droplet freezes) below 268 K (Mari et al., 2000). R is equal to 0.05 for all gases, nitric acid excepted. Gas/ice partitioning for H_2O_2 follows Mari et al. (2000). Other gases do not partition to ice particles. The condensed fraction for soluble aerosols follows Stier et al. (2005): r_{cond}=0.1 in ice clouds, 0.85 in liquid clouds, 0.75 in mixed clouds.

1.2.4 Below-cloud scavenging rates

The rate of removal by rain (washout) of an irreversibly soluble gas such as HNO_3 can be obtained by evaluation of the integral of this rate over the drop-size distribution (Levine and Scwartz, 1982):

$$\lambda_{below} = \int k_g \cdot (\pi D^2) \cdot (\frac{dN}{dD}) \cdot dD, \tag{12}$$

where D is the droplet diameter, $(dN/dD \cdot dD)$ (m^{-4}) is the number of drops per unit volume in the diameter interval dD, and k_g (m s^{-1}) is the gas-phase mass transfer coefficient. The size distribution is related to the intensity of precipitation I (mm h^{-1}) (de Wolf, 2001) with

$$\frac{dN}{dD} = 1.98 \cdot 10^7 \cdot I^{-0.384} \cdot D^{2.93} \cdot \exp[-5.38 \cdot I^{-0.186} \cdot D], \tag{13}$$

where D is in millimeters. The mass transfer coefficient is given by the semi-empirical Frössling equation

$$k_g = \frac{D_g}{D} \cdot (2 + 0.6 \cdot Re^{1/2} \cdot Sc^{1/3}), \tag{14}$$

where D_g is the diffusivity of the gaseous compound in air (m^2 s^{-1}) (Fuller et al., 1969), parameterized as a function of temperature T and molecular mass m,

$$D_g = \frac{1.9 \cdot 10^{-4}}{m^{2/3}} \cdot T^{1.75}, \tag{15}$$

and Re and Sc are the Reynolds and Schmidt numbers given by

$$Re = D \cdot \frac{U_t}{D_a}, Sc = \frac{D_a}{D_g}, \tag{16}$$

where D_a is the diffusivity of air (=0.133·10^{-4} m^2 s^{-1}) and U_t is the terminal velocity of rain droplets obtained from Atlas et al. (1973):

$$U_t = 9.65 - 10.3 \cdot \exp(-0.6 \cdot D) \tag{17}$$

Since the explicit determination of λ_{below} using Eq. (12) would be computationally expensive in the CTM, the following regression is used

$$\lambda_{below} = 4.9 \cdot 10^{-5} \cdot I^{0.65} \cdot (T/298)^{1.26} \tag{18}$$

which reproduces the rate given by Eq. (12) to better than 2% in the range 0.5–5 mm h^{-1}.

For less soluble gases, the scavenging rate given by Eq. (18) is multiplied by the dissolved fraction calculated using Henry's law (analogous to Eq. (2)), where the below-cloud water content L_{below} (g m^{-3}) is calculated using a regression of the water content determined from the size distribution of de Wolf (2001):

$$L_{below} = 0.113 \cdot 10^{-6} \cdot I^{0.91} \tag{19}$$

which reproduces the water content of de Wolf (2001) to 1% or less in the range 0.1–5 mm h^{-1}.

The below-cloud scavenging rate of accumulation mode aerosol is adapted from Henzing et al. (2006):

$$\lambda_{below} = A_0 \cdot (e^{A_1 P^{A_2}} - 1), \tag{20}$$

with parameters $A_1 = 1.41 \cdot 10^{-6}$, $A_2 = 0.218$ and $A_3 = 0.25$ obtained through fitting of their results for 0.1 μm particle radius (their Figure 3, green curve).

1.3 Impact of cirrus gravitational settling

The vertical redistribution of key soluble compounds (e. g. HNO_3) resulting from the gravitational settling of cirrus clouds (Lawrence and Crutzen, 1998) is represented in the model based on the ECMWF parameterizations for ice clouds. Since the removal of soluble compounds due to precipitation scavenging is already taken into account in the stratiform scavenging parameterization (section 1.2), we consider here the effect of gravitational settling due to the non-precipitating part of ice cloud cover fractions, obtained as the difference between a_{cld} and a_{in} (Eq. (9)). The ice sedimentation velocity (w^{ice} in m s^{-1}) is calculated according to European Centre for Medium-Range Weather Forecasts (2004):

$$w^{ice} = f_{<100} \cdot w_{<100} + (1 - f_{<100}) \cdot w_{>100}, \tag{21}$$

where $f_{<100}$ is the mass fraction of ice particle smaller than 100 μm, $w_{<100}$=0.15 m s^{-1} is the terminal velocity of the smaller ice particles, and $w_{>100}$ is the terminal velocity (m s^{-1}) of the larger particles (>100 μm) given by

$$w_{>100} = 2.3 \cdot (\frac{IWC_{>100}}{1000})^{0.16}, \tag{22}$$

where $IWC_{>100}$ is the ice water content of large particles (g m^{-3}). The mass fraction of smaller particles is given by

$$f_{<100} = \min(1, 0.252 \cdot (\frac{IWC_0}{IWC_{tot}})^{0.163}), \tag{23}$$

where IWC_0 is set to 1 g m^{-3} and IWC_{tot} is the total ice water content in g m^{-3}.

Only nitric acid (Lawrence and Crutzen, 1998) and aerosols (Liu et al., 2001) are allowed to partition on ice particles. As mentioned in the previous section, the condensed fraction is equal to 1 for HNO_3 and 0.1 for water-soluble aerosols. Since nitric acid partitions to ice particles only in absence of liquid water, sedimentation is applied only when ice water represents more than 90% of the total condensed water content. Moreover, partitioning to ice is also suppressed at very low temperatures ($T < 205$ K) (Krämer et al., 2003).

2 Heterogeneous sink in cloud droplets

The conversion of glyoxal to SOA being assumed to be complete in liquid clouds due to oligomer formation upon evaporation of cloud droplets (Loeffler et al., 2006), the overall conversion rate is limited by the time needed for processing of air by clouds (Lelieveld, 1990). We follow the approach of Rodhe and Grandell (1972) (see Section 1.2.1) and express the conversion rate as

$$\lambda_{cl} = \frac{\lambda_{in} \cdot LCC}{1 + (1 - LCC)^2 \cdot t_{av} \cdot \lambda_{in}}, \tag{24}$$

where LCC is the liquid cloud cover obtained from ECMWF analyses, λ_{in} is the in-cloud conversion rate, determined by gas-phase diffusion of glyoxal to the cloud droplets (Jacob, 2000), t_{av} is the average time an air parcel spends inside the cloud (taken equal to 1 hour).

3 Heterogeneous sink on aqueous aerosols

The rate for the heterogeneous uptake of glyoxal to an aqueous aerosol component (OC, BC or inorganic) is given by

$$\lambda_{aer} = A/(\frac{r_n}{D_g} + \frac{4}{\omega \cdot \gamma}), \tag{25}$$

where A is the aerosol surface density expressed as

$$A = 4\pi r_{wet}^2 \cdot \frac{C_{dry}}{\rho_{dry}(\frac{4\pi \cdot r_{dry}^3}{3})}, \tag{26}$$

r_n is the number mean particle radius, D_g is the gas diffusivity (Eq. (15)), ω is the mean molecular speed of the compound, γ $(= 2.9 \cdot 10^{-3})$ is the reactive uptake coefficient (Liggio et al, 2005), r_{dry} and r_{wet} are respectively the effective dry and wet radius of the aerosol, ρ_{dry} is the dry aerosol density (1.75 g cm^{-3} for sulfates, 1.3 g cm^{-3} for OC, 1.77 g cm^{-3} for BC) (Jimenez, 2007) , and C_{dry} is the mass concentration of the dry aerosol component calculated by the CTM.

The dry effective radius of inorganic aerosol (160 nm) is obtained from the Global Aerosol Data Set (GADS) (Köpke et al., 1997). For organic aerosol (also black carbon aerosol), we use a value of 145 nm based on field campaigns over Amazonia (Rissler et al., 2006) and Africa (Bellouin et al., 2007).

Since γ is much lower than one, the precise value of the number mean radius r_n is unimportant. The growth factor (ratio of r_{wet} to the r_{dry}) for inorganic aerosols is obtained from the EQSAM module (Metzger et al., 2002). For organic aerosol, it is calculated by assuming an activity coefficient of water equal to 1.1, typical of α-pinene SOA (Compernolle et al., 2009):

$$r_{wet} = r_{dry} \cdot (1 + \frac{18 \cdot \rho_{dry} \cdot RH}{MW_{OA} \cdot (\zeta_{H_2O} - RH)})^{1/3}, \tag{27}$$

where MW_{OA} is the molecular weight of the organic aerosol, and ζ_{H_2O} is the activity coefficient of water in the organic aerosol. This formula leads to growth factors of typically 1.13–1.18 at 90% RH, in good agreement with field campaigns over Amazonia (Rissler et al., 2006; Mircea et al., 2006).

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