Modeling of dispersion and scavenging in the Polyphemus platform. Applications to passive tracers.

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Contents

The aim of this document is to briefly describe the physical models used in Polyphemus for the dispersion of passive tracers, with a focus on scavenging processes. The report is organized as follows. The eulerian model is described in section 1. The parameterizations for scavenging used in the eulerian model are given in section 2. The gaussian model is described in section 3. The related parameterizations for scavenging are given in section 4.

1 Eulerian model

The Eulerian model, called Polair3D [Boutahar et al., 2004] allows the modeling of gaseous and particulate matter air pollution over many scales ranging from urban to continental. It simulates the emission, dispersion, chemical reactions (radioactive decay for instance) and removal of pollutants in the lower troposphere.

Polair3D requires inputs to describe the surface characteristics, initial and boundary conditions, emission rates, point source release description, physical and chemical properties of the species and various meteorological fields over the entire modeling domain. This step may be achieved by using the preprocessing tools included in Polyphemus which transforms raw data to final input files. The physical parameterizations involved in preprocessing are described in AtmoData scientific documentation [Njomgang et al., 2005].

1.1 Main equation

Polair3D [Boutahar et al., 2004], is a numerical solver for the chemistry-transport equation:

$$\frac{\partial c_i}{\partial t} = \underbrace{-\mathrm{div}(Vc_i)}_{\text{advection}} + \underbrace{\mathrm{div}\left(\rho K \nabla \frac{c_i}{\rho}\right)}_{\text{diffusion}} + \underbrace{\chi_i(c)}_{\text{chemistry}} + S_i - L_i \tag{1}$$

which is satisfied by all involved chemical species. The concentration of the i-th species is c_i. The transport driven by wind V is the advection term. The diffusion term $\mathrm{div}\left(\rho K \nabla \frac{c_i}{\rho}\right)$ essentially accounts for turbulent mixing in the vertical. Chemical production and losses of the i-th species are introduced with χ_i. Additional sources (S_i, emissions) and losses (L_i, wet and dry deposition) are included.

For particles, the same equation is solved for each component represented in every size section. The number of size sections and their size ranges are chosen following a log-normal distribution. In practice, it means that the effects of condensation/evaporation, coagulation, nucleation and aqueous-phase chemistry are neglected. The size of the particles plays a major role in the deposition processes as explained in sections 2 and 4.

1.2 Transport

The flow is prescribed by meteorological fields computed by an independent meteorological model. There is no feedback between the chemistry-transport model and the meteorological model. This is referred as off-line coupling with a meteorological model.

Diffusion coefficients (3×3-matrix K) is assumed to be diagonal with a constant horizontal diffusion, usually set to $10,000\ \mathrm{m}^2 \cdot \mathrm{s}^{-1}$. Vertical diffusion coefficients are estimated with Louis parameterization [Louis, 1979].

1.3 Chemistry

Radioactive decay may be seen as a first-order chemistry. Much more complex chemical mechanism involving many chemical species are also managed in Polair3D, e.g., RACM [Stockwell et al., 1997].

1.4 Dry and wet deposition

Trace gases and small particles are removed from the atmosphere via deposition to the surface. Dry deposition velocity of gases is based on the resistance model of Zhang [Zhang et al., 2003].

Wet deposition refers to the impact of clouds and the transfer to the Earth's surface by precipitation.

More details on parameterizations used in Polair3D may be found in section 4.

1.5 Numerical issues

The simulation domain is discretized with a regular grid, with fixed step in latitude/longitude coordinates or in meters (recommended at local scale). The vertical discretization is defined with layers of increasing thickness, but with constant altitudes over the domain, whatever the orography may be.

Numerical schemes are:

- for the time integration, a first-order operator splitting, the sequence being advection–diffusion–chemistry;

- a direct space-time third-order advection scheme with a Koren flux-limiter, as recommended in Verwer et al. [2002];
- a second-order order Rosenbrock method for diffusion.

2 Parameterizations for the scavenging processes in the eulerian model

2.1 Gravitational settling

Gravitational settling is taken into account as an additional term for vertical advection of particles. The vertical velocity for an advected particle is then $w - v_g$, with v_g the velocity of gravitational settling that depends on particle size.

An approximation is given by the Stokes velocity, given in equation (17). Rigorously, we should take into account the deviation from Stokes formula for non submicronic particles. This leads to solve the following non-linear system with respect to the sedimentation velocity:

$$v_g = \sqrt{\frac{4g\, d_p\, C_c\, \rho_p}{3\, C_d v_g\, \rho_{air}}} \tag{2}$$

with g the gravity constant, d_p the particle diameter, ρ_p its density and ρ_{air} the air density. The Cunningham coefficient C_c is given by (18) and the drag coefficient C_d is a function of the particle Reynolds number and then of the gravitational velocity (the expression is not detailed here).

The resolution of this algebraic equation is done with a Newton algorithm.

2.2 Below-cloud wet scavenging

In this section, the rain intensity p_0 is given in $\mathrm{mm\,h^{-1}}$. Moreover, the aerosol radius (for a monodisperse distribution) is r_p, given in μm.

D_r (in meters) is the diameter of a rain droplet (eventually in a polydisperse distribution or as a representative diameter for a population which is assumed monodisperse). The aerosol diameter is noted d_p (also in meters).

2.2.1 Theoretical model

We quote here the theoretical modelling of the scavenging coefficient for the washout process (below-cloud scavenging), that corresponds to the scavenging of aerosols by falling raindrops.

Representation of the raindrops distribution A key point is the representation of the raindrops distribution and of the falling velocity.

Droplet distribution The raindrops distribution is usually described by a Gamma distribution with 4 parameters:

$$n_r(D_r) = \alpha_0 \, D_r^{\alpha} \exp(-\beta \, D_r^{\gamma}) \tag{3}$$

The two classic cases correspond to the Marshall-Palmer ($\alpha = 0$; $\gamma = 1$) and to the Khrigian-Mazin ($\alpha = 2$, $\gamma = 1$) distribution.

Representative diameter Numerous parameterizations exist to define a *representative* diameter, in order to treat only one monodisperse population of raindrops:

1. In Pruppacher and Klett [1998] (page 34):

$$D_r = 0.976 \times 10^{-3} \, {p_0}^{0.21} \tag{4}$$

2. From a Marshall-Palmer distribution:

$$D_r = 0.243 \, 10^{-3} \, {p_0}^{0.21} \tag{5}$$

3. In a quasi-identic way to the previous parameterization (Andronache [2004]):

$$D_r = 0.24364 \, 10^{-3} \, {p_0}^{0.214} \tag{6}$$

4. In Loosmore and Cederwall [2004]:

$$D_r = 0.97 \, 10^{-3} \, {p_0}^{0.158} \tag{7}$$

5. In Mircea et al. [2000], parameterizations from measures taken in eastern Mediterranean sea give:

$$D_r = [0.63 - 0.72] \, 10^{-3} \, p_0^{0.23} \tag{8}$$

to compare with the first and the fourth equations.

6. At last, in Underwood [2001] (page 35), it is quoted that the original article of Slinn recommends:

$$D_r = 0.7 \, 10^{-3} \, p_0^{0.25} \tag{9}$$

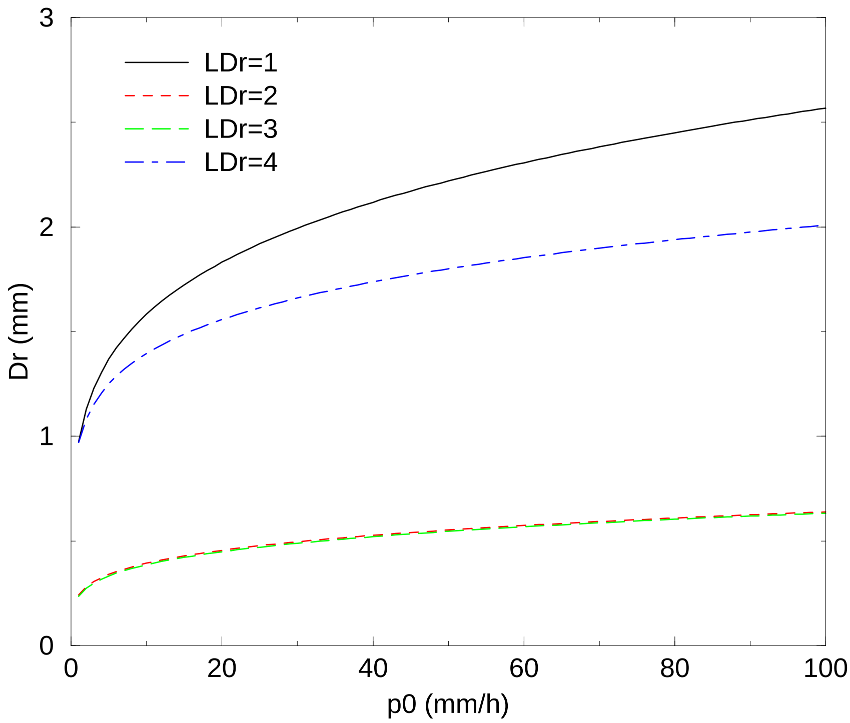

Figure 1: Evolution of the representative diameter wrt the rain intensity, for some parameterizations.

Globally, we have:

$$D_r = [0.2431 - 0.97]\, 10^{-3}\, D_r^{[0.158-0.25]} \tag{10}$$

The comparison of the four first parameterizations is given in the figure 1. We quote the high dispersion of the results: it is usually quoted that the Marshall-Palmer distribution overestimates the small droplets number, that leads to overestimating the collision efficiencies and then the scavenging.

Falling velocity Several parameterizations give an expression for the falling velocity U_{drop} (in $\mathrm{m\,s^{-1}}$) function of diameter:

1. Kessler's parameterization (Andronache [2003], page 143, and Mircea and Stefan [1998], table 2):

$$U_{drop} = 130\sqrt{D_r} \tag{11}$$

2. The parameterization cited in Seinfeld [1985] (page 632):

$$U_{drop} = 9.58 \left[1 - \exp\left(-\left(\frac{D_r}{0.171 \times 10^{-2}} \right)^{1.147} \right) \right] \quad (12)$$

3. The parameterization given in Seinfeld and Pandis [1998] or in Mircea et al. [2000] that uses the droplet final velocity (to be computed by an algorithm because outside the domain of the Stokes formula).

4. The parameterization in Andronache [2004]:

$$U_{drop} = 3.778\, 10^3\, D_r^{0.67} \quad (13)$$

5. Finally in Loosmore and Cederwall [2004]:

$$U_{drop} = 4.854\, D_r\, \exp(-195\, 10^{-3} D_r) \quad (14)$$

D_r is in meter in all these formulas. Comparisons are given in figure 2 with the parameterization $LDr = 1$ for the representative diameter.

Polair3D **configuration** Polair3D uses for parameterizations $LDr = 1$ and $LUdrop = 2$.

2.2.2 Expression for the scavenging coefficient

Monodisperse case The volume dragged by a raindrop with a diameter D_r is given by the following expression:

$$\frac{\pi}{4}\, {D_r}^2\, U_{drop}(D_r) \quad (15)$$

The collision volume or *effective volume*, that is to say the volume where the contact is efficient for one time unit, takes also into account the aerosol diameter d_p and its falling velocity u_{grav} (in $\mathrm{m\,s^{-1}}$):

$$\frac{\pi}{4}\, (D_r + d_p)^2\, (U_{drop}(D_r) - u_{grav}(d_p)) \quad (16)$$

where the gravitational sedimentation velocity u_{grav} is given by the Stokes formula:

$$u_{grav} = \frac{{d_p}^2\, (\rho_p - \rho_{air})\, g\, C_c}{18 \mu_{air}} \quad (17)$$

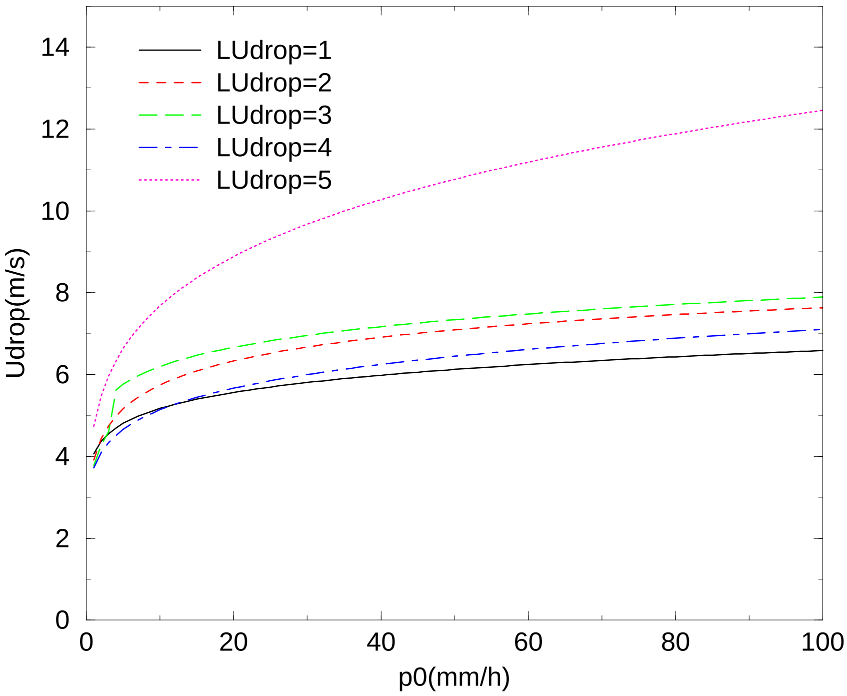

Figure 2: Evolution of the falling velocity wrt the rain intensity, for several parameterizations for velocity. The representative diameter is computed with $LDr = 1$.

with ρ_p (in kg·m^{-3}) the particle volumic mass, μ_{air} the air dynamic viscosity (in Pa s), g the gravity (in m $^{-2}$) and C_c the corrective Cunningham factor, meaning that slidings appear for small particles ($\simeq$ 1 μm). If we don't want to use tabulated values for C_c, the following expression could be used [Seinfeld and Pandis, 1998]:

$$C_c = 1 + \frac{2\,\lambda_{air}}{d_p}\left(1.257 + 0.4\,\exp\left(-0.55\frac{d_p}{\lambda_{air}}\right)\right) \tag{18}$$

with the air free mean path λ_{air} (in meters):

$$\lambda_{air} = \frac{2\,\mu_{air}}{P}\left(\frac{8}{\pi\,R_{air}\,T}\right)^{-1/2} \tag{19}$$

R_{air} is the gas constant for air (in J K^{-1} kg^{-1}) and T is the temperature (in K).

This representation implies that every particle in the effective volume is captured and then neglects the effects of the air movement resulting from the fall of the raindrop which alters the particles trajectory. This effect is parameterized by a collision efficiency $E(D_r, d_p)$, defined as the fraction of particles with diameter d_p, in the collision volume of a droplet with diameter D_r, that are collected:

$$\frac{\pi}{4}\,(D_r + d_p)^2\,(U_{drop}(D_r) - u_{grav}(d_p))\,E(D_r, d_p) \tag{20}$$

Simplifications Two classical approximations allow to simplify the previous expression:

- $U_{drop}(D_r) \gg u_{grav}(d_p)$
- $(D_r + d_p)^2 \simeq D_r^2$

If N_r is the total droplet density (in m^{-3}), assumed monodisperse, we finally have:

$$\Lambda(d_p) = \frac{\pi}{4}{D_r}^2\,U_{drop}(D_r)\,E(D_r, d_p)\,N_r \tag{21}$$

By definition, the rain intensity p_0 may be written as:

$$p_0 = \int_0^\infty \frac{\pi}{6}D_r^3\,U_{drop}(D_r)\,n_r(D_r)\,dD_r$$

that is to say for the monodisperse case:

$$p_0 = \frac{\pi}{6}{D_r}^3\,U_{drop}(D_r)\,N_r \tag{22}$$

We finally have the classical expression:

$$\Lambda(d_p) = \frac{3}{2}\,\frac{E(D_r, d_p)\,p_0}{D_r} \tag{23}$$

where p_0 is in ISU $(\mathrm{m\,s^{-1}})$.

Polydisperse case This framework can be applied to polydisperse populations of aerosol and droplets. Let us note respectively $n_p(d_p)$ and $n_r(D_r)$ (in $\mathrm{m^{-3}\,m^{-1}}$) the number distribution for aerosols and for raindrops.

The number of particles with a diameter in the range $[d_p, d_p + dd_p]$ collected in a time unit by raindrops is given by:

$$n_p(d_p)\,dd_p \int_0^{\infty} \frac{\pi}{4}(D_r + d_p)^2\,(U_{drop}(D_r) - u_{grav}(d_p))\,E(D_r, d_p)\,n_r(D_r)\,dD_r \tag{24}$$

On the basis of approximations, we directly obtain for the scavenging rate of the particles with diameter d_p, $\frac{dn_p(d_p)}{dt} = -\,\Lambda(d_p)\,n_p(d_p)$:

$$\Lambda(d_p) = \int_0^{\infty} \frac{\pi}{4} {D_r}^2\,U_{drop}(D_r)\,E(D_r, d_p)\,n_r(D_r)\,dD_r \tag{25}$$

2.2.3 Parameterization of the collision efficiency

A keypoint is the parameterization of the collision efficiency, defined as the ratio between the number of collisions between water droplets and particles, and the number of particles in the geometric volume covered.

E is equal to 1 if all particles are effectively captured but in practice $E \ll 1$. Measurements show that a collision results almost every time in capture, collisions are then rare.

It is necessary to take into account different phenomena to explain the eventual capture of a particle:

- *Brownian diffusion* might place a particle on a droplet trajectory.

  The hypothesis concerning the equivalence collision/capture and the fact that brownian diffusion is more important for fine particles justify that this process is in favor of the capture of small particles.

- Another phenomenon that favors capture of bigger particles is *inertia*. It could induce collision by preventing particles from following streamlines around droplets.

- The last phenomenon is interception that results from the contact of a particle following a streamline around the droplet because of its size. The considerations about inertia and interception could not strictly dissociate from considerations about particle density, inertia concerns in fact heavy particles and interception big ones.

Globally, both processes explain that scavenging is important for small aerosols (typically diameter less than 0.01 μm by brownian diffusion, and for the big aerosols (typically diameter higher than 2 μm) by inertial effect. Aerosols with intermediate diameters form what is called classically the *Greenfield Gap* or *scavenging gap*, in the range $[0.01;2]$ μm , weakly scavenged.

Note that experimentally, this scavenging default is less evident than predicted by theory (see below the neglected effects).

The expression proposed in Seinfeld and Pandis [1998] after a dimensional analysis and application of Buckingham's theorem gives E function of five adimensioned numbers:

- Reynolds' number of raindrop

$$Re = \frac{D_r \, U_{drop}}{2\nu_{air}} \tag{26}$$

  where $\nu_{air} = \mu_{air}/\rho_{air}$ is the air kinematic viscosity (in $\mathrm{m^2\,s^{-1}}$).

- Schmidt's number of the captured particle:

$$Sc = \frac{\nu_{air}}{D_B} \tag{27}$$

  with D_B the brownian diffusivity coefficient of the particle (in $\mathrm{m^2\,s^{-1}}$):

$$D_B = \frac{k\,T}{3\,\pi\,\mu_{air}\,d_p} C_c \tag{28}$$

  where k is the Boltzmann's constant (in $\mathrm{J\,K^{-1}}$).

- Stokes' number of the captured particle:

$$St = 2\tau \, \frac{U_{drop} - u_{grav}}{D_r} \tag{29}$$

  with τ a characteristic relaxation time taken equal to u_{grav}/g, that is to say:

$$\tau = \frac{(\rho_p - \rho_{air})\, d_p^2 \, C_c}{18\mu_{air}} \tag{30}$$

- the ratios between diameters (ϕ) and viscosities (ω):

$$\phi = \frac{d_p}{D_r} \ , \quad \omega = \frac{\mu_w}{\mu_{air}} \tag{31}$$

with μ_w the viscosity of water.

The formula for E is then given by:

$$E = \frac{4}{Re\,Sc}\left(1 + 0.4\,Re^{1/2}\,Sc^{1/3} + 0.16\,Re^{1/2}\,Sc^{1/2}\right) + \\ 4\,\phi\left(\omega^{-1} + [1 + 2\,Re^{1/2}]\,\phi\right) + \left(\frac{St - S^{*}}{St - S^{*} + 2/3}\right)^{3/2}\left(\frac{\rho_p}{\rho_w}\right)^{1/2} \tag{32}$$

with the critical Schmidt's number $S^{\star}$:

$$S^{*} = \frac{1.2 + 1/12\,\ln(1 + Re)}{1 + \ln(1 + Re)} \tag{33}$$

The three terms respectively correspond to the terms of brownian diffusion, interception and impaction. The distribution $E(D_r, d_p)$ is given for a raindrop D_r =0.1 mm in figure 3, where the different contributions are verified.

The dependency of efficiency to the raindrop diameter is given in figure 4. The efficiency increases when diameter decreases. The amplitude of the differences has to be compared to the figure 1 about the dispersion of representative diameter parameterizations.

2.3 Wet scavenging of aerosols: *in-cloud*

When they fall, raindrops aggregate cloud droplets, and suspended gas and aerosols. This is the wet scavenging phenomenon. Inside a cloud, pollutants (gas and particles) are almost absorbed by cloud droplets, so that wet scavenging is reduced to their aggregation by raindrops.

Giving a concentration of pollutant c, its evolution due to wet scavenging in clouds is governed by the equation:

$$\frac{\partial c}{\partial t}(x, y, z, t) = -\Lambda(x, y, z)c(x, y, z, t) \tag{34}$$

where Λ is the wet scavenging coefficient, expressed in seconds, and whose expression depends on the collision process between cloud and rain droplets. For this reason it is independent from the considered pollutant.

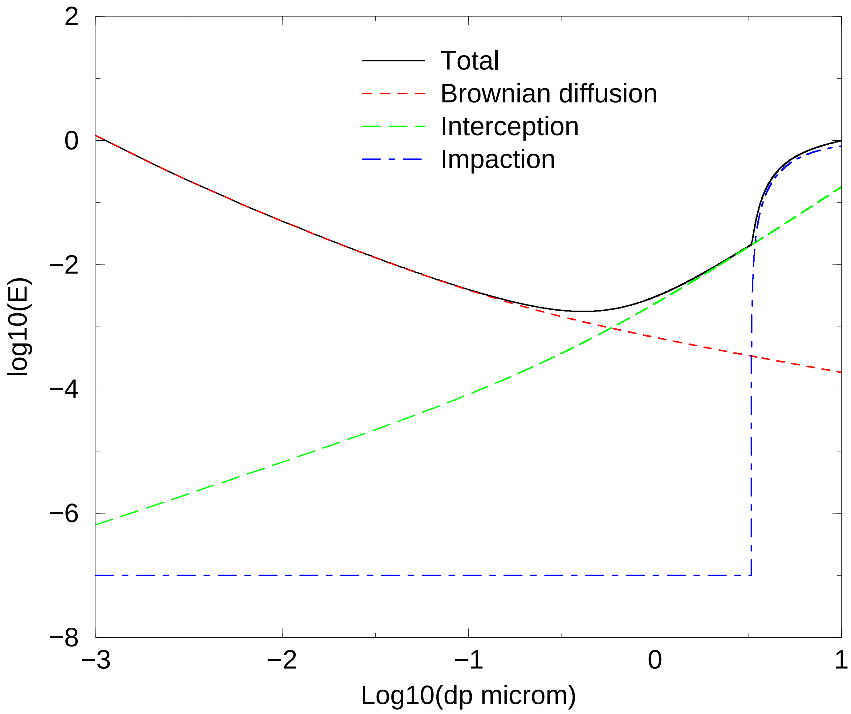

Figure 3: Contributions to the collision efficiency $E(D_r, d_p)$ for $D_r = 0.1\text{mm}$

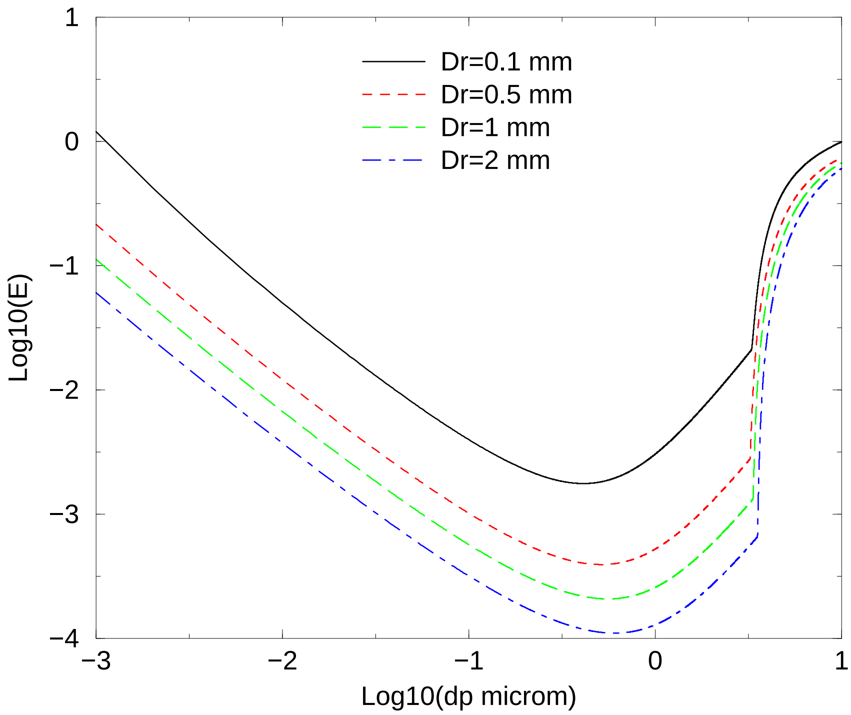

Figure 4: Collision efficiencies $E(D_r, d_p)$ for D_r =0.1 mm, 0.5 mm, 1 mm et 2 mm.

The integration of this equation between the initial t_0 and final t_1 time is done analytically:

$$c(x,y,z,t_1) = c(x,y,z,t_0)\exp[-\Lambda(x,y,z)(t_1 - t_0)] \tag{35}$$

Below we present the two parameterizations computed in POLAIR3D : one is from the model CAMx (Corporation [2005]), and the other one from the Multiscale Air Quality (CMAQ) (Roselle and Binkowski [1999]).

1. Parameterization from CAMx:

   The volume covered by a raindrop in its fall by time unit is equal to:

   $$V = \frac{\pi}{4}(d_d + d_c)^2 \nu_d \tag{36}$$

   where

   - d_d the raindrop diameter, in meters, is given by the empirical law (Scott [1978]) :

     $$d_d = 9.0 \times 10^{-4} p_0^{0.21} \tag{37}$$

     in which p_0 is the precipitation rate, expressed in $\mathrm{mm\,hr^{-1}}$.
   - d_c is the diameter of cloud droplets, in meters.
   - ν_d is the falling velocity of raindrops, in $\mathrm{m{\cdot}s^{-1}}$, given by the empirical law (Scott [1978]) :

     $$\nu_d = 3100\ d_d \tag{38}$$

   The wet scavenging coefficient in clouds can be written as:

   $$\Lambda = E\frac{\pi}{4}(d_c + d_d)^2 \nu_d N_d \tag{39}$$

   where

   - N_d is the numerical concentration ($\#.m^{-3}$) of raindrops, that could be computed from the precipitation rate:

     $$N_d = \frac{2.8 \times 10^{-7} p_0}{\pi (d_d)^3 \nu_d / 6} \tag{40}$$

     The number 2.8×10^{-7} takes into account the conversion of p_0 from $\mathrm{mm\,hr^{-1}}$ in m^{-1} ·

- E represents the probability that a cloud droplet on the trajectory of a raindrop is actually aggregated. The air fluxes created by the fall of the raindrop decrease this probability of the 0.9 order.

We generally admit that the diameter of a cloud droplet may be neglected as compared to raindrops:

$$d_c \ll d_d \Longrightarrow \Lambda = \frac{\pi}{4}(d_d)^2 \nu_d N_d \tag{41}$$

By replacing N_d by its expression (40) we obtain:

$$\Lambda = 4.2 \times 10^{-7} \frac{E\, p_0}{d_d} \tag{42}$$

then d_d by its empirical expression (37) :

$$\Lambda = 4.2 \times 10^{-4} p_0^{0.79} \tag{43}$$

2. Parameterization from CMAQ:

In this parameterization, the expression for the scavenging coefficient is:

$$\Lambda = -\frac{1 - e^{-\frac{\tau_{\text{cld}}}{\tau_{\text{washout}}}}}{\tau_{\text{cld}}} \tag{44}$$

where

- τ_{cld}, expressed in seconds, is equal to the timestep of the dispersion model if the cloud size exceeds the mesh dimensions, and is equal to 1 hour otherwise,
- τ_{washout} represents the necessary time for all the volume of water to precipitate to the ground.

$$\tau_{\text{washout}} = \frac{W_T \Delta z_{\text{cld}}}{\rho_{\text{H}_2\text{O}} p_0} \tag{45}$$

p_0 is the precipitation rate , Δz_{cld} is the cloud depth, W_T is the liquid water content of the cloud ($\text{kg}\,\text{m}^{-3}$), and $\rho_{\text{H}_2\text{O}}$ the density of the liquid water.

2.4 Dry deposition

2.4.1 Fluxes Modeling

The dry deposition flux is a boundary condition applied to the diffusion operator:

$$K_z \nabla c \cdot \mathbf{n} = E - v_{dep} c \tag{46}$$

with K_z the vertical turbulent coefficient, E the surfacic emission and v_{dep} the deposition velocity. $\mathbf{n}$ is the unit vector upward oriented.

Here, we do not take into account the resuspension terms.

2.4.2 Theoretical model for dry deposition velocity

The dry deposition velocity is expressed for particles in function of the dynamical and surface resistances to the deposition and the sedimentation velocity:

$$v_d = v_g + \frac{1}{R_a + R_s} \tag{47}$$

Resistance parameterizations to the deposition for particles are inspired by those proposed by Zhang Zhang et al. [2001].

Sedimentation velocity It gives the conjugated effects of gravitation and friction on a particle. The parameterization of the sedimentation velocity used here is limited to the Stokes velocity.

Aerodynamic resistance The expression for aerodynamic resistance used for particles is similar to the one used for gases.

Surface resistance The surface resistance R_s is representative of several phenomena traducing the captation ability of the surface in regard to particles.

$$R_s = \frac{1}{3u_*(E_B + E_{IM} + E_{INT})R_1} \tag{48}$$

with:

- u_*, the friction velocity (in $\mathrm{m\,s^{-1}}$).
- E_B represents the part of contact particle/surface induced by the brownian diffusion. The tendency of this phenomenon is to equalize the particle concentrations between reference height and surface.

$$E_B = \frac{\nu_{air}}{D_B}^{-\gamma} \tag{49}$$

with γ a parameter of the model Zhang et al. [2001].

- E_{IM} is the impact coefficient and traduces the deposition directly due to the particles inertia:

$$E_{IM} = \left(\frac{St}{\alpha + St}\right)^2 \tag{50}$$

with α a parameter of the model Zhang et al. [2001] and St the Stokes number defined in function of the terrain type:

$$St = \begin{cases} u_{grav} \frac{u_*}{g\, A} & \text{for "rough" surfaces} \\ u_{grav} \frac{{u_*}^2}{g\, \nu_{air}} & \text{for "smooth" surfaces} \end{cases} \tag{51}$$

A is a parameter of the model called "characteristic radius of receptors" (in meters).

- E_{INT} is the interception coefficient of the particles by the surface:

$$E_{INT} = \frac{1}{2}\left(\frac{d_p}{A}\right)^2 \tag{52}$$

.

- R_1 is the corrector coefficient for the rebound and describes the possible rebound of a particle on the surface:

$$R_1 = \exp\left(-\sqrt{St}\right) \tag{53}$$

with St the Stokes number defined above.

2.5 Wet scavenging for gases

For the gaseous phase, the wet below-cloud scavenging is parameterized by $L_{wet}(c_i) = -\Lambda_i\ c_i$ with c_i the concentration of the chemical species i. The coefficient Λ_i is detailed in Sportisse and Du Bois [2002].

3 Gaussian model

Gaussian diffusion models are widely used in regulatory applications because they are analytical and conceptually appealing, and computationally cheaper to use. Basically, it consists in assuming a Gaussian distribution of mean concentration in the horizontal and vertical directions at any downwind location from the source. The Gaussian plume model gives the analytical expression of the concentration in the case of a continuous emission from the source at a constant rate. the Gaussian puff model gives the concentration for an instantaneous point source.

3.1 Form of the Gaussian plume model

Many assumptions are implied when using that model (see Arya [1999]), particularly :

1. Continuous emission from the source, at least for a time equal to or greater than the time of travel to the location (receptor) of interest, so that the material is spread out in the form of a steady plume between the source and the farthest receptor.

2. Steady-state flow and constant meteorological conditions. That is, the time between two different meteorological conditions is greater than the time of travel between source and receptor, so that the steady plume has been obtained for each meteorological condition.

3. A constant mean transport wind in the horizontal direction.

4. No wind shear in the vertical direction.

5. Strong enough winds to make turbulent diffusion in the downwind direction negligible in comparison to advection.

In that context, the concentration C at a given point within the boundary layer is given by the formula:

$$C(y,z) = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}}\exp(-\frac{(y-y_s)^2}{2\sigma_y^2})\Big[\exp(-\frac{(z-H)^2}{2\sigma_z^2}) + \exp(-\frac{(z+H)^2}{2\sigma_z^2})\Big] \tag{54}$$

Here, Q is the source emission rate, given in mass per second, $\bar{u}$ is the mean wind velocity, and σ_y and σ_z are the Gaussian plume parameters. The coordinate y refers to horizontal direction "crosswind", that is, at right angles to the plume axis which is also the wind axis, and y_s is the source coordinate in that direction. z refers to the point coordinate in the vertical direction, and H is the source height above ground. The purpose of the last term is to take into account the reflection of the plume on the ground. Note that, in the case of inversion, the reflection at the inversion layer can similarly be taken into account. The modified concentration formula in the case of inversion is then:

$$C = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}} \exp(-\frac{(y-y_s)^2}{2\sigma_y^2}) \sum_{N=-5}^{+5} \left[\exp(-\frac{(z-H+2Nz_i)^2}{2\sigma_z^2}) + \exp(-\frac{(z+H+2Nz_i)^2}{2\sigma_z^2}) \right] \tag{55}$$

Here, z_i is the inversion height, and 5 reflections on it are considered. The values of the terms in the above sum are negligible for $|N| > 5$ (see [Arya, 1999]).

Note that the previous expressions do not take into account any loss process, it hence assumes mass conservation in the plume.

3.2 Form of the Gaussian puff model

The Gaussian puff model is based on the same assumptions as the plume model, except that the horizontal diffusion in the downwind direction is not negligible any more (see [Arya, 1999]).

The Gaussian puff formula for an instantaneous point source is given by:

$$C(x,y,z,t) = \frac{Q}{2\pi^{3/2}\sigma_x\sigma_y\sigma_z\bar{u}} \exp(-\frac{(x-x_c)^2}{2\sigma_x^2} - \frac{(y-y_c)^2}{2\sigma_y^2}) \left[\exp(-\frac{(z-H)^2}{2\sigma_z^2}) + \exp(-\frac{(z+H)^2}{2\sigma_z^2}) \right] \tag{56}$$

Here, Q is the total mass released by the instantaneous source, x is the coordinate in the downwind horizontal direction, x_c and y_c are the puff center coordinate in downwind and crosswind directions respectively. So, we can also write: $x_c = \bar{u}t$ and $y_c = y_s$, where t is the time since the puff has been released, and y_s is the initial puff coordinate along the crosswind direction. σ_x, σ_y and σ_z are the puff diffusion parameters, which are, in general, different from the plume dispersion parameters, although the current practice is to use the same parameterizations for both.

The effect of inversion can be taken into account in exactly the same way as for the plume model. However, it is not currently available in the Polyphemus puff model.

3.3 Dispersion parameterization schemes

For an estimate of the dispersion parameters σ_x, σ_y and σ_z, empirical parameterization schemes are widely used. Many schemes have been proposed, most of them giving the dispersion parameters as functions of the downwind distance and stability class, and based on a few diffusion experiments. In Polyphemus, the Pasquill stability scheme and the Briggs formula for dispersion parameters are used.

3.3.1 Pasquill stability scheme

Pasquill has defined six stability classes, from A to F, where:

- A corresponds to extremely unstable conditions
- B corresponds to moderately unstable conditions
- C corresponds to slightly unstable conditions
- D corresponds to neutral conditions
- E corresponds to slightly stable conditions
- F corresponds to moderately stable conditions

Currently, the Pasquill stability class is assumed to be known in the Polyphemus Gaussian model. However in the future a preprocessing program is planned, that will calculate the stability class depending on the surface wind speed, daytime incoming solar radiation and nighttime cloudiness.

3.3.2 Briggs interpolation formula

The Briggs formula are based on the Pasquill-Turner stability classes and on the Prairie Grass experiments. This parameterization is born from an attempt to synthesize several widely used parameterization schemes by interpolating them for open country and for urban areas. These formulas apply to a distance from the source up to 10 km and may be extended up to 30 km. They are particularly recommended for urban areas. The tables 1 and 2 give the formula for open country and urban area respectively, where x is the downwind distance from the source.

Pasquill Type	σ_y	σ_z
A	$0.22x(1+0.0001x)^{-1/2}$	$0.20x$
B	$0.16x(1+0.0001x)^{-1/2}$	$0.12x$
C	$0.11x(1+0.0001x)^{-1/2}$	$0.08x(1+0.0002x)^{-1/2}$
D	$0.08x(1+0.0001x)^{-1/2}$	$0.06x(1+0.0015x)^{-1/2}$
E	$0.06x(1+0.0001x)^{-1/2}$	$0.03x(1+0.0003x)^{-1}$
F	$0.04x(1+0.0001x)^{-1/2}$	$0.016x(1+0.0003x)^{-1}$

Table 1: Briggs Interpolation formula for open country

Pasquill Type	σ_y	σ_z
A-B	$0.32x(1+0.0004x)^{-1/2}$	$0.24x(1+0.001x)^{1/2}$
C	$0.22x(1+0.0004x)^{-1/2}$	$0.20x$
D	$0.16x(1+0.0004x)^{-1/2}$	$0.14x(1+0.0003x)^{-1/2}$
E-F	$0.11x(1+0.0004x)^{-1/2}$	$0.08x(1+0.0015x)^{-1}$

Table 2: Briggs Interpolation formula for urban areas

These formula are given for the plume model. However, the Gaussian model in Polyphemus uses them for the puff model as well as for the plume model,with $\sigma_x = \sigma_y$. The value taken for x is the distance from the puff to the release point (that is, at time t, $\bar{u}t$).

3.4 Plume rise and loss processes

3.4.1 Plume rise

This process will be taken into account in the next release of the system.

3.4.2 Loss processes

The loss processes modify the expressions of the concentration given in equations (54) and (56) by multiplying them by a loss factor. The new concentration is then given by:

$$c' = f_{decay} * f_{scavenging} * f_{deposition} * c \tag{57}$$

Where:

- f_{decay} is the loss factor due to radioactive or biological decay.

$$f_{decay} = \exp(-\lambda \frac{x}{\bar{u}})$$

in the plume model (x is the downwind distance from source) and

$$f_{decay} = \exp(-\lambda t)$$

for the puff model (t is the travel time from release point). One can define the constant λ as: $\lambda = 1/t_{1/2}$, where $t_{1/2}$ is the half-life time of the species.

- $f_{scavenging}$ is the loss factor due to scavenging. It is likewise defined as

$$f_{scavenging} = \exp(-\Lambda \frac{x}{\bar{u}})$$

in the plume model and

$$f_{decay} = \exp(-\Lambda t)$$

in the puff model. Here, Λ is the scavenging coefficient, which depends on the species, rain intensity and diameter for aerosol species (see section 4).

- $f_{deposition}$ is the loss factor due to dry deposition. The expression of this factor is more complicated. The model used in Polyphemus is the source-depletion model proposed by Chamberlain (1953). In this model, the deposition of the airborne material is taken into account by reducing the source rate Q as a function of downwind distance from the source (or from release point in the puff model). This leads to the following expression:

$$f_{deposition} = \exp\left(-\frac{v_d}{\bar{u}}\sqrt{\frac{2}{\pi}}\int_0^x \frac{dx}{\sigma_z \exp(H^2/(2\sigma_z^2))}\right)$$

Here, x is the downwind distance from source in the plume model and the distance between the center of the puff and the release point for the puff model (same x as used in the Briggs formula). v_d is the deposition velocity, which depends on the ground type, meteorological conditions and physical properties of the species (see section 4).

Another model has been proposed by Overcamp (1976), and might be implemented later in Polyphemus, for it is simpler and does not require to evaluate an integral. The limitation of these models is that they assume a uniform depletion along the vertical direction in the plume, whereas the dry deposition occurs mostly near the ground.

4 Loss processes for Gaussian model

For both scavenging and dry deposition, we have made the choice to use simple robust models in the case of the gaussian model. This is in coherence with the crude assumptions related to the gaussian formulation and with the possible lack of data required as inputs for more complicated microphysical parameterizations.

4.1 Scavenging

For gaseous species, the scavenging rate Λ is either given by the user or defined as a parameterization of rain intensity (p_0 in $\mathrm{mm\,h^{-1}}$) through the so-called Belot model:

$$\Lambda = A\, p_0^B \tag{58}$$

with A and B provided by the user.

For particulate matter, the scavenging rate is either given by the user for a given aerosol diameter or parameterized as:

$$\Lambda = \frac{3}{2} \frac{E\, p_0}{D_r} \tag{59}$$

with E the scavenging efficiency and D_r the diameter or rain drops. The exact computation of E implies the description of many processes (impaction, brownian scavenging, aso): we have chosen, following [Underwood, 2001], to use the simple law below depending on the aerosol diameter D:

- $E = 0.1$ for $D \leq D_1 = 1\,\mu m$;
- $E(D) = E(D_1) + (1 - E(D_1)) \frac{D - D_1}{D_2 - D_1}$ for $D_1 \leq D \leq D_2 = 10\,\mu m$;
- $E(D) = 1.$ for $D \geq D_2$.

Moreover, the raindrop diameter is given by:

$$D_r = 7.10^{-4}\, p_0^{0.25} \tag{60}$$

Notice that we only take into account *below-cloud* scavenging (washout) and not *in-cloud* scavenging (rainout).

4.2 Dry deposition

For gaseous species, the dry deposition velocity v_d is given by the user (in $\mathrm{m\,s^{-1}}$).

For particulate matter, v_d is a function of the dry deposition velocity without gravitational settling ($v_d^{'}$) and of the gravitational settling velocity (v_s). The exact form of this function is still controversial and we have chosen to use a form that guarantees mass conservation in the surface layer:

$$v_d = \frac{v_s}{1 - \exp\left(-\frac{v_s}{v_d^{'}}\right)} \tag{61}$$

In the first release of the system, $v_d^{'}$ is provided by the user (in order not to use data from Land Use Coverage). The gravitational settling velocity is given as a function of the diameter as the Stokes terminal velocity (valid for diameters less than 20 μm):

$$v_d = \frac{D^2(\rho_p - \rho_{air}) g C_c}{18\mu_{air}} \tag{62}$$

with ρ_p (in $\mathrm{kg\,m^{-3}}$) the aerosol density, μ_{air} the air dynamic viscosity (in Pa s), g the gravity constant (in $\mathrm{m\,s^{-2}}$) and C_c the Cunningham coefficient given by:

$$C_c = 1 + \frac{2\lambda_{air}}{D}\left(1.257 + 0.4\exp\left(-0.55\frac{D}{\lambda_{air}}\right)\right) \tag{63}$$

λ_{air} (in meters) is the air mean free path:

$$\lambda_{air} = \frac{2\mu_{air}}{P}\left(\frac{8}{\pi R_{air} T}\right)^{-1/2} \tag{64}$$

with R_{air} the molar gas constant for air (in $\mathrm{J\,K^{-1}\,kg^{-1}}$), T the temperature (in K) and P the pressure (in Pa).

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