ENVI-met implementation of the gas/ particle dispersion and deposition model PDDM

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1 Implementation of the gas/ particle dispersion and deposition model PDDM

1.1 Prognostic equations

The dispersion of a gaseous or particulate atmospheric component can be expressed using the standard advection-diffusion equation:

$$\frac{\partial \chi}{\partial t} + u\frac{\partial \chi}{\partial x} + v\frac{\partial \chi}{\partial y} + w\frac{\partial \chi}{\partial z} =$$
$$\frac{\partial}{\partial x}\left(K_\chi \frac{\partial \chi}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_\chi \frac{\partial \chi}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_\chi \frac{\partial \chi}{\partial z}\right)$$
$$+ Q_\chi(x,y,z) + S_\chi(x,y,z)$$

In the atmospheric equations of ENVI-met, the local concentration of a component χ is used in the unit $[mg(\chi)kg^{-1}(Air)]$. Any process that can lead to local de- or increase of χ such as sink/source processes, sedimentation of particles or chemical transformation of gases are included explicitly in the system by the source term Q_χ and/or sink term S_χ. Implicit dependencies between the local concentration and the source/sink rate of χ can lead to restrictions in the time step used and in the numerical treatment of the equation system (see also section 1.3)

1.1.1 Formal definition of source/sink processes

Local source/sink processes (only "sources" for simplicity hereafter) can be separated into four types with respect to their spatial structure (and consequently in the unit of their definition):

1. Point Sources: q_p, Unit $[mgs^{-1}]$
2. Line Sources: q_l, Unit $[mgs^{-1}m^{-1}]$
3. Area Sources: q_f, Unit $[mgs^{-1}m^{-2}]$
4. Volume Sources: q_v, Unit $[mgs^{-1}m^{-3}]$

Distinction between those four types is essential, when defining external sources (e.g. car exhausts) in the model area.

Before using sources in the prognostic model equations, their emission rate must be transformed into the correct formal unit for Q_χ $[mgkg^{-1}s^{-1}]$. First, all types of sources are transformed into q^* in $[mgs^{-1}]$:

$$\begin{aligned} q^* &= q_p \\ &= q_l \cdot \Delta x, y \\ &= q_f \cdot \Delta x \Delta y \\ &= q_v \cdot \Delta x \Delta y \Delta z \end{aligned}$$

Secondly, Q_χ is calculated with respect to the cell size in which the source is located:

$$Q_\chi = q^* \cdot (vol \cdot \rho)^{-1}$$

with cell volume $vol = \Delta x \Delta y \Delta z$. Note, that for the output of local concentrations, it is common to have the data related to the air volume in $[mgm^{-3}]$ rather than related to the air mass in $mgkg^{-1}$. Therefore, for output the local concentration is converted by

$$\chi^* = \chi \cdot \rho$$

ENVI-met allows to simulate sources with time dependent emission rates. To implement this, each source is defined by 24 values representing the emission rates $q(h)$ for each hour h [0-23] of the day. The actual emission rate for a hour h and a minute m [0-59] is then linearly interpolated as:

$$q(h,m) = \frac{60-m}{60}q(h) + \frac{m}{60}q(h+1)$$

After hour 23 the calculation restarts with h=0. The frequency of updating the emission rate can be selected by the user and should normally be around 10 min. Changes in emission rates only influence the local sources, no trend correction will be performed at the atmospheric grid points. In case of rapid changes in emissions, a smaller update interval is necessary to avoid unrealistic oscillations.

1.2 Treatment of sedimentation and deposition processes

The sedimentation of particles due to gravitational forces and the deposition of both, particles and gases, to surfaces are complex processes that influence the local concentration atmospheric to a large extend. Especially the absorption of gases at natural surfaces (leafs, soils) and the capturing and re-releasing of particles at rough surfaces are processes, that are not fully understood and lack usable numerical parameterizations. The model approach given here is based on classical laws for particle and gas dynamics in the atmosphere and at surfaces.

1.2.1 Composition of sink term S_χ

The sink term S_χ is composed out of three components which, in their final balance, give the local sink rate of the component χ. These components are:

- concentration change due to gravitational settling including deposition at surfaces (composed of downward flux $\chi_\downarrow$ and flux received from grid boxes above $\chi^\downarrow$)
- deposition at leaf surfaces (χ_{plant})
- chemical transformations causing an decrease of components A and B and increase of product AB

(All fluxes in $[mg kg^{-1} s^{-1}]$). The concentration change due to gravitational settling only applies to particulate matter whereas the deposition at solid surfaces and plant leafs are calculated both for particles and gases. Finally, chemical transformations occur only in the case of reactive gaseous species. This reaction complex is not included in ENVI-met by now, so that the calculations are restricted to chemical inert gases.

1.2.2 Formal description of sedimentation and deposition

The changes in local concentration due to sedimentation and deposition can be written as an additional advective component using the sedimentation/ deposition velocity $v_{s/d}$ as the relevant transportation velocity:

$$\left.\frac{\partial \chi}{\partial t}\right|_{sed} = v_{s/d}\frac{\partial \chi}{\partial z} \qquad (1.1)$$

As only downward fluxes can occur in the case of gravitational settling, a simple upstream scheme can be used to transform (1.1) into the finite-difference form:

$$\left.\frac{\Delta \chi}{\Delta t}\right|_{sed} = v_{s/d}\frac{\chi(z+1) - \chi(z)}{\Delta z} \qquad (1.2)$$

where Δz is the vertical distance between the prognostic calculation points for level z and $z+1$. Splitting this equation into a loss and a gain term we get

$$\left.\frac{\Delta \chi}{\Delta t}\right|_{sed} = v_{s/d}\frac{\chi(z+1)}{\Delta z} - v_{s/d}\frac{\chi(z)}{\Delta z} \qquad (1.3)$$

where the receipt of particles from the z-level above is counted positive and the loss of particles is counted negative.

The downward flux of particles per time unit ($\chi_\downarrow$) due to gravitational settling can be written as

$$\chi_\downarrow(z) = -v_{s/d}\frac{\chi(z)}{\Delta z} \qquad (1.4)$$

and the gain of particles settling from the level above ($\chi^\downarrow$) is

$$\chi^\downarrow(z) = v_{s/d}\frac{\chi(z+1)}{\Delta z} \qquad (1.5)$$

Obviously and in order to keep the model mass-conserving, eq. (1.5) for level z must be balanced with eq. (1.4) for level $z+1$.

If the concerned grid box is the lowest box above the ground surface or the box below is covered by a building, the deposition velocity v_d (for calculation see section 1.2.4) is used instead of the settlement speed v_s in (1.4). In contrast to v_s, the deposition velocity is non-zero for gaseous components and depends on the type of component and the type of absorbing surface.

Finally, the deposition of particles and gaseous components at vegetation elements (χ_{plant}) has to be considered in the total balance. The total loss of particles due to deposition at the leaf surface is given by

$$\chi_{plant}(z) = LAD(x,y,z) \cdot v_{d,p} \cdot \chi(z) \qquad (1.6)$$

For the estimation of $v_{d,p}$ see next section.

The total balance of the local concentration change due to sedimentation and deposition can now be written as:

$$\frac{\partial \chi(z)}{\partial t} = \chi^\downarrow(z) + \chi_\downarrow(z) - \chi_{plant}(z) \qquad (1.7)$$

or, as sink term for the prognostic equation:

$$S_\chi(x,y,z) = \chi^\downarrow(z) + \chi_\downarrow(z) + \chi_{plant}(z) \qquad (1.8)$$

To obtain the total amount of deposed mass on a surface per second [mg m^{2}s^{-1}], for the ground surface and for roofs the equation

$$\frac{m_s}{\partial t} = \chi_\downarrow \cdot \rho \cdot \Delta z$$

is used. Here, the total mass deposed in the grid box volume is re-related to the horizontal $\Delta x \Delta y$ plane as the surface is the only place where deposition can take place.

For the leaf surfaces the equation

$$\frac{m_{plant}}{\partial t} = \chi_{plant}(z) \cdot \frac{1}{LAD(x,y,z)} \cdot \rho$$

is used in which the overall deposition inside the grid box is distribute over the available leaf area density in the box.

In the recent version of ENVI-met, no re-suspension of particles is taken into account. The calculation of the sedimentation and deposition processes is realized explicitly, which means that the concentration of the last time step is used to calculate the flux through the horizontal cell walls and to the leaf surface. This leads to an additional stability condition to ensure that no negative concentration values can occur in the case of high sedimentation rates (see Section 1.3 for details).

1.2.3 Calculation of particle settlement speed v_s

The settling velocity of a particle depends on the particle diameter D, its density ρ_χ and the turbulence characteristics of the air flowing around the particle given by the Reynolds number Re. For any Re, the settling velocity v_s can be calculated following **?** with

$$v_s = \left(\frac{4\rho_\chi \cdot D \cdot g \cdot C_c}{3C_d \cdot \rho_\chi} \right)^{1/2} \tag{1.9}$$

where C_c is the so-called *Slip-Correction-Factor* given by (1.10) and C_d is the drag force on the particle calculated after (1.11).

$$C_c = \begin{cases} 1 + \frac{2\lambda}{D}\left(1.257 + 0.4\exp\left(-\frac{1.1D}{2\lambda}\right)\right) & ; D \leq 10\mu m \\ 1 & ; D > 10\mu m \end{cases} \tag{1.10}$$

The mean free path of air molecules (λ) used in the equation above is set to a constant value of 0.0651 μm which corresponds to an air temperature of 298 K, the small dependency of λ on the air temperature is hence neglected in the model. [1]

The drag force C_d on a particle is calculated with respect to the local Reynolds Number:

$$C_d = \begin{cases} \frac{24}{Re} & Re < 0.1 \\ \frac{24}{Re}\left(1 + \frac{3}{16}Re + \frac{9}{160}Re^2 \ln(2Re)\right) & 0.1 < Re < 1 \\ \frac{24}{Re}\left(1 + 0.15Re^{0.678}\right) & 1 \leq Re < 900 \\ 0.44 & 900 < Re \end{cases} \tag{1.11}$$

The boundary values for switching between the equations (Re =0.1, 1 and 900) have been changed from the original values given by Seinfeld and Pandis (1999)(0.1, 2 and 500) in order to allow a smoother transition between the definition intervals. For Reynolds numbers smaller then 0.1, the formulation of C_d is equivalent to the Stokes' law.

The Reynolds Number for a spherical particle is given by

$$Re = \frac{\rho_\chi \cdot v_s \cdot D}{\mu} \tag{1.12}$$

where μ is the viscosity of the air (=$1.8 \cdot 10^{-5}$ kgm^{-1}s^{-1} at 298K, kept constant).

Obviously, the equation (1.9) for v_s cannot be solved directly as C_d depends on v_s over Re. The equation set can be solved quickly in a few iteration steps, starting with v_s calculated using the linear expression valid for $Re < 0.1$ as a first estimate:

$$v_s = \frac{\rho_\chi \cdot D^2 \cdot g \cdot C_c}{18\mu} \tag{1.13}$$

The iteration is canceled after the change of v_s per iteration step falls below 0.1 mm/s.

The estimation of the settling velocity is calculated independently from other forcing effects (wind field, thermal effects). The movement of particles due to wind transport is taken into account by the advection term in the prognostic equation. This approach may lead to an underestimation of the Reynolds Number, but seems accurate enough, as the gravitational settling dominates only when the wind field is weak.

1.2.4 Calculation of deposition velocity v_d of particles and gases

The deposition velocity of both particles and gases to a surface or to a leaf is a complex process including aerodynamical aspects as well as possible chemical interactions with the interface. In general, the deposition speed v_d can be expressed as the inverse sum of three different resistances r_a,r_b and r_t :

$$v_d = \frac{1}{r_a + r_b + r_t}$$

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1. For the calculation, all variables must be in SI units (D in [m], ρ_χ in [kgm^{-3}], ...)

Here, r_a is the aerodynamic resistance of the surface, r_b is the sub-layer resistance and r_t is an additional surface resistance applied to gases which takes into account the absorption rate of the component at the surface. For **particles**, r_t does not apply, as they are assumed to be completely removed from the air as soon as they hit the surface. Here, the formulation for v_d becomes:

$$v_{d,part} = \frac{1}{r_a + r_b + r_a r_b v_{s,0}} + v_{s,0} \qquad (1.14)$$

where $v_{s,0}$ is the settlement speed close to the surface, set equal to v_s calculated after (1.9).

1.2.4.1 Aerodynamic Resistance r_a The aerodynamic resistance of a surface to gases and particles is supposed to be the same as for the transport of heat. Using the logarithmic law, r_a can be written as:

$$r_a = \frac{(\ln(z_1/z_0))^2}{\kappa^2 \mathbf{u}(z_1)} \cdot \Phi_h \qquad (1.15)$$

where Φ_h is a scaling function to include thermal effects. In the ENVI-met model, r_a is directly derived from the turbulent exchange coefficient for heat at the ground surface or at the walls $K_{h,0}^{(w)}$ in which the effect of thermal stratification is already included. Hence, the formulation for r_a at the ground surfaces, at roofs and at walls becomes:

$$r_a = \frac{1}{c_{d,N} \cdot \mathbf{u}_{w+1}^{tang} \cdot \Phi_h} = \frac{1}{D_h} = \frac{\Delta_w}{K_{h,0}^{(w)}}$$

At plants, r_a is taken from the expression given by Braden (1982):

$$r_a = A \sqrt{\frac{D^*}{\max(\mathbf{u}, 0.05)}} \qquad (1.16)$$

where $\mathbf{u}$ is the wind speed at the leaf surface. A and D^* are plant specific parameters which are set to $A=87$ $\sqrt{s}m^{-1}$ for deciduous trees and grass and to $A=200$ $\sqrt{s}m^{-1}$ for conifers. D^* represents the typical leaf diameter and is 0.02 m for conifers and grass and 0.15 m for deciduous trees (Schilling, 1990; Naot and Mahrer, 1989).

1.2.4.2 Sublayer Resistance (quasi-laminar resistance) r_b The sublayer resistance r_b is an additional resistance caused by the quasi-laminar layer adjacent to the surface, across which the transfer velocity depends on the molecular properties of the transported substance as well as on the surface characteristics.

For **gaseous components**, the sublayer resistance can be expressed using the molecular diffusivity D_χ of the respective chemical species Wesley (1989):

$$r_{b,gas} = \frac{1}{u_*} \cdot 5 Sc^{2/3} \qquad (1.17)$$

where Sc is the Schmidt-Number

$$Sc = \nu/D_\chi$$

with the kinematic viscosity of the air ν (set to $\nu = 1.5 \cdot 10^{-5}$ at 298 K). For gases, the Brownian diffusion coefficient D_χ is fixed and given as input data.

In case of **particles**, the molecular diffusivity is replaced by the Brownian diffusivity which depends on the particle size. Smaller particles experience faster Brownian motion and therefore less sublayer resistance than bigger ones. The following expression includes the effects of Brownian motion (Sc-number) as well as the influence of inertial impaction (Stokes-number St)Seinfeld and Pandis (1999).

$$r_{b,part} = \frac{1}{u_* \left(Sc^{-2/3} + 10^{-3/St}\right)} \qquad (1.18)$$

The estimation of the Brownian diffusion coefficient is based on the Stokes-Einstein relationship :

$$D_\chi = \frac{k \cdot T \cdot C_c}{3\pi \cdot \mu \cdot D} \qquad (1.19)$$

where k is the Boltzmann constant (=$1.38 \cdot 10^{-23}$ JK^{-1}), and C_c is the slip correction factor given by (1.10). Figure 1 illustrates the relationship between D and D_c.
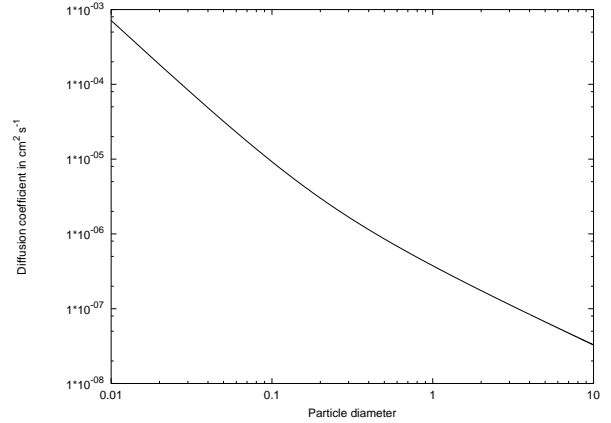

Figure 1: Particle diffusion coefficient D_χ calculated from (1.19) in dependency to diameter in μm

The Stokes Number is given by

$$St = \frac{v_s \cdot u_*^2}{g \cdot \nu} \qquad (1.20)$$

For the ground surfaces (including water) and for roofs, the friction velocity is re-calculated from the exchange

coefficient $K_{m,0}^{(w)}$ provided by the main model and the horizontal wind component with

$$u_* = \sqrt{K_{m,0}^{(w)} \frac{|\mathbf{u}_{hor}|}{\Delta^w}} \qquad (1.21)$$

where Δ^w is the vertical distance from the center of the grid cell to the solid surface, normally $0.5\Delta z$ (see also 3.4.6.3 of main documentation). In case of vegetation elements, u_* is recalculated from the aerodynamic resistance r_a (see above) with

$$u_* = \sqrt{\frac{1}{r_a} |\mathbf{u}|} \qquad (1.22)$$

where $\mathbf{u}$ is the three dimensional wind speed.

1.2.4.3 Additional surface resistance r_t for gases
The surface resistance for gases depends on the chemical properties of the substance and of the surface type. In ENVI-met, four different surfaces are distinguished:

- bare soil $r_{t,soil}$
- sealed/artificial soils including walls and roofs $r_{t,sealed}$
- water surfaces $r_{t,water}$
- plant leafs $r_{t,plant}$

Bare soils

For the calculation of the resistance of a **bare soil** ($r_{t,soil}$) the model needs the resistance of the surface to SO_2 ($r_{t,SO2}$) and O_3 ($r_{t,O3}$) as reference data for a non-reactive (SO_2) and a reactive (O_3) species. The values vary according to the structure of the surface but should normally be around 150 to 200 for a bare surface. When the surface is covered with old plant material or is extremely smooth, higher values are possible. Generally, the resistance for SO_2 is higher than the value for the reactive O_3, for example $r_{t,SO2} = 1000$ and $r_{t,O3} = 400$ for a sand surface. An overview of different resistances can be found in Seinfeld and Pandis (1999) p. 974 f. In the recent model version a constant value of 150 is used for both resistances $r_{t,SO2}$ and $r_{t,O3}$.

For other species than SO_2 or O_3 the surface resistance can be calculated from:

$$r_{t,soil} = \left(\frac{10^{-5} \cdot H_i^*}{r_{t,SO_2}} + \frac{f_0^i}{r_{t,O_3}} \right)^{-1} \qquad (1.23)$$

H_i^* is the Henry's Law constant in $[M \cdot atm^{-1}]$ and f_0^i is the normalized reactivity of the species. Values for common chemical components are listed in Table 1, more data can be found in Seinfeld and Pandis (1999). Note that in

Table 1: Henry's law constant H^*, Normalized reactivity f_0^i and Ratio of molecular diffusivity of different species after Seinfeld and Pandis (1999)

Species i	H^*	f_0^i	D_{h2O}/D_χ
SO_2	1×10^5	0	1.89
O_3	1×10^{-2}	1	1.63
NO_2	1×10^{-2}	0.1	1.60
NO	2×10^{-3}	0	1.29
CO	9.5×10^{-4}	0	1.20
H_2O_2	1×10^5	1	1.37
NH_3	2×10^4	0	0.97
APM	0	0	1

the version so far, the temperature dependency is not included in the equations. The values given by the equations are for 298 K surface temperature.

Sealed surfaces

For all **sealed surfaces**, it is assumed that no gases are absorbed and $r_{t,sealed}$ is set to 9999 (or another high value).

Water surfaces

In case of **water surfaces,** the approach presented by Sehmel (1980) is used to calculate $r_{t,water}$:

$$r_{t,water} = \frac{2.54 \cdot 10^4}{H_i^* \cdot T_o \cdot u_*} \qquad (1.24)$$

where T_0 is the local surface temperature of the water.

Plant leafs

The estimation of the **surface resistance at plants** is quite more complex, as the plant stomata actively regulate the gas exchange with the ambient air. In addition, a certain amount of gas, depending on the reactivity, can directly pass through the mesophyll of the leaf surface or parts the plant might be covered by dew, so that the resistance of a water surface is effective there.

To summarize, the surface resistance for plants $r_{t,plant}$ can be composed as:

$$r_{t,plant} = (1 - f_w)r_s^* + f_w r_{t,water} \qquad (1.25)$$

in which r_s^* is the modified stomata resistance (including the mesophyll transport) and f_w is the fraction of leaf that is wet. The latter one is also directly provided by the plant model. The modified stomata resistance is

$$r_s^* = r_s(D_{H_2O}/D_\chi) + r_{meso} \qquad (1.26)$$

where D_{H_2O}/D_χ is the ratio of the molecular diffusivity of water and the observed species (see Table 1) and r_{meso} is the mesophyll resistance calculated after Seinfeld and Pandis (1999):

$$r_{meso} = \left(3.3 \cdot 10^{-4} H_i^* + 100 f_0^i\right)^{-1} \qquad (1.27)$$

Finally, r_s is the stomata resistance for vapour transfer as it is supplied by the plant model.

1.2.5 Special case CO_2

The treatment of CO_2 differs significantly from other gases. The local CO_2 concentration is normally calculated by the plant model and is used to estimate the stomata resistance of the plant. However, it is also possible to add CO_2 sources to the model. In this case, the equations presented before are not used and the additional mass flux is added to the plant CO_2 system. The absorption of CO_2 at the plant is also directly calculated by the plant model and for a bare soil it is assumed to be zero due to the lack of a soil respiration model.

1.3 Numerical aspects

The explicit calculation of the sedimentation and deposition process adds a new stability criteria for the solution of the prognostic equation. The time step used must be small enough to ensure, that no negative concentrations can occur due to an overestimated downward sedimentation flux or deposition.

For each grid cell, a critical time t_{zero} is calculated, after which the concentration would be below zero if all fluxes were kept constant:

$$t_{zero}(i,j,k) = \frac{\chi(i,j,k)}{-(\chi^\downarrow(i,j,k) - \chi_\downarrow(i,j,k) - \chi_{plant}(i,j,k)}$$
$$(1.28)$$

If t_{zero} is below 0, the concentration in the cell is increasing and no restriction for the time step apply. The maximum usable time step in the model can then be defined to be half of the time needed to reach the zero concentration condition:

$$\Delta t_{\max} = 0.5 t_{zero,\min} \qquad (1.29)$$

The determination of t_{zero} has to be performed after each solution of the prognostic equation. If $\Delta t_{\max}$ is below the main time step of the ADI system, the solution procedure is splitted into sub-loops using smaller time steps with $\Delta t = \Delta t_{\max}$ until the main time step is reached.

1.4 Example Calculations

Some example calculations are presented next to test the functionality of the presented system as a module in ENVI-met

1.4.1 Example A: Sedimentation and deposition of particles with different diameter

The first example aims to test the sedimentation and deposition of particles with diameter from 0.01 μm to 100 μm. The particle density ρ_χ is kept constant with 1 gcm^{-3}. The model area consists of a simple plane where the source is located in a height of 10 m, emitting as a point source with e=10 mgs^{-1}. A small band of average dense trees (12m width, 10 m height) have been placed at the grids x=18-21 to test the effect of vegetation on the particle distribution. In the simulation, the wind direction was set to 90° (from the right hand side of the model domain) and the initial wind speed at 10 m height was 3 m/s. The results presented in Figure 2 are taken from the model after 1 h of calculation time.

References

Braden, H. (1982). Simulationsmodell für den Wasser-, Energie- und Stoffhaushalt in Pflanzenbest"anden. *Rep. Inst. Meteorol. Uni. Hannover*, (23).

Naot, O. and Mahrer, Y. (1989). Modelling microclimate environments: A verification study. *Boundary Layer Meteorology*, 46:333–354.

Schilling, V. K. (1990). A paramerization for modelling the meteorological effects of tall forests– A case study of a large clearing. *Boundary Layer Meteorology*, 55:283–304.

Sehmel, G. A. (1980). Particle and Gas Deposition. A Review. *Atmospheric Environment*, 14:983–1011.

Seinfeld, J. H. and Pandis, S. N. (1999). *Atmospheric chemistry and physics*. Wiley-Interscience, New York.

Wesley, M. L. (1989). Parameterizations of surface resistance to gaseous dry deposition in regional scale numerical models. *Atmospheric Environment*, 23:1293–1304.
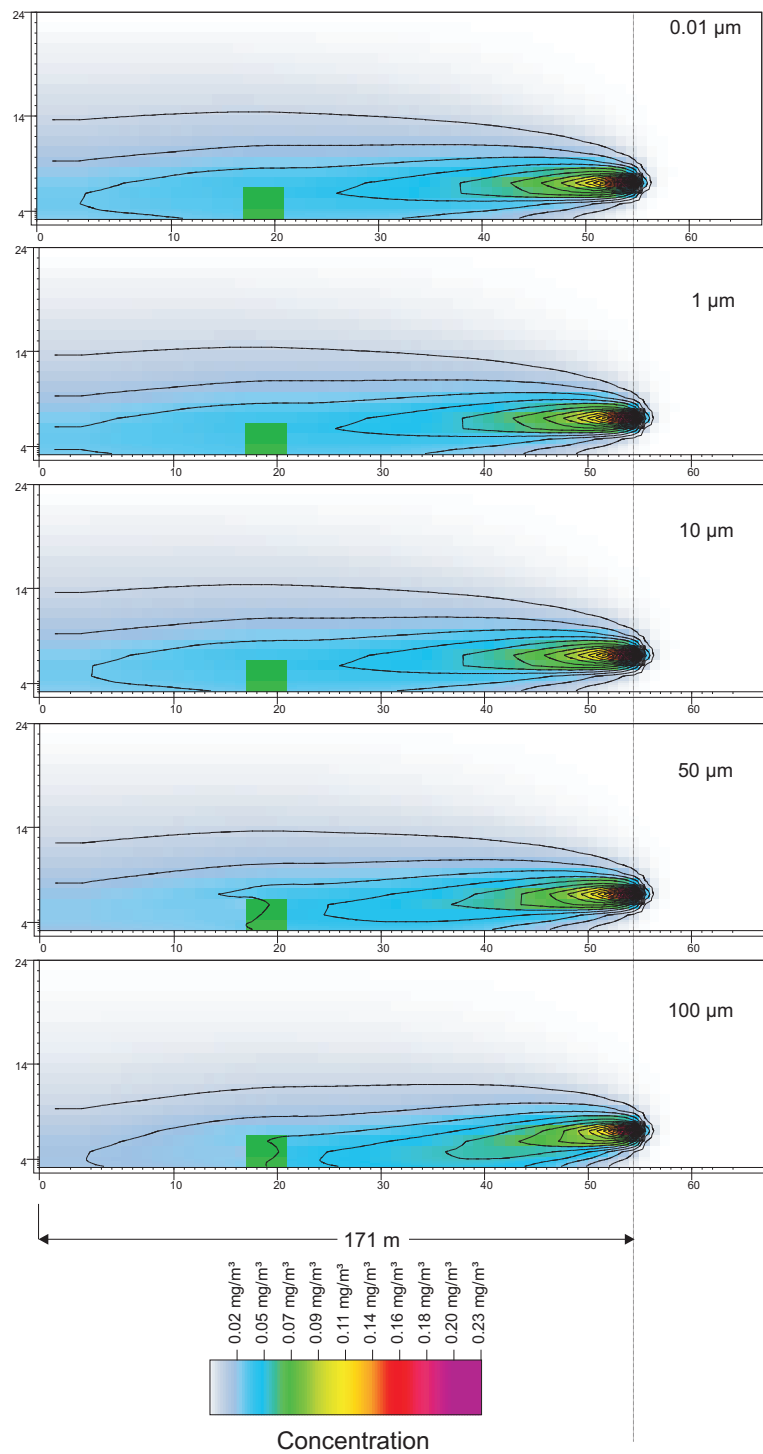
Figure 2: Concentration fields for different sized particles. X-Z cut at y=14, Source: Point-type, height 10 m, e= 10 mgs^{-1}, Isoline distance 0.01 mgm$^-$3