Removal processes in SILAM (and in CTMs)

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Outline:

- Removal mechanisms
- Species properties
- Dry deposition
- Wet deposition

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Removal mechanisms

Stuff can get out of dispersion map:

- by decay and/or transformations
- through boundaries
- dry deposition
- wet deposition

Species properties

Gases:

- Diffusivity
- Solubility (care needed)
- Reactivity (much care needed)

Particles:

- Size (very approximate measure)
- Density
- Hygroscopicity, stickiness, etc.

These properties are not exactly those controlling removal.

Statements like “Gas has solubility X” or “particel of size Y” can be misleading. . .

Particle properties

“Particle of size X” in papers means (usually): “the particle has the same property Y as would spherical particle of diameter X and density Z”.

Currently in SILAM particles are considered spherical with specified material/density and material-dependent humidity growth.

Basic properties controlling removal:

- Inertial relaxation time τ_p or settling velocity $v_s = \tau_p g$
- Physical size d_p
- Diffusivity D or Schmidt number $\mathrm{Sc} = D/\nu$

Removal schemes are expressed in these terms. . .

Example: particle size

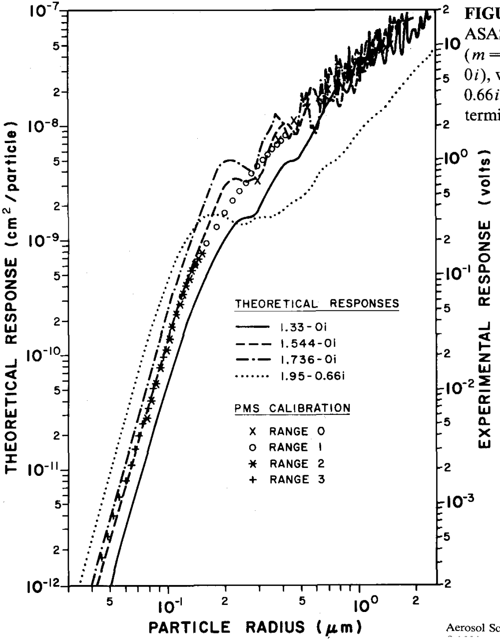


FIGURE 3. Comparison of the theoretical ASASP-X response curves for magnesium oxide ($m = 1.736 - 0i$), sodium chloride ($m = 1.544 - 0i$), water ($m = 1.33\text{-}0i$), and carbon ($m = 1.95 - 0.66i$) with the manufacturer's calibration determined as outlined in the Appendix.

D. M. Garvey and R. G. Pinnick

Aerosol Science and Technology 2:477–488 (1983)

Dry deposition

Problem:

Given concentration at some height above ground find the steady-state flux.

Deposition velocity:

$$v_d(z_1) = J/C(z_1)$$

Approaches:

- Fixed or size-dependent deposition velocity (particles)
- Landuse-dependent deposition velocity (gases)
- Resistance analogy
  (aerodynamic + quasi-laminar sub-layers etc. . .)
- Something more fancy. . .

Deposition pathway

- Turbulent (aerodynamic) layer
- Laminar layer
- Surface

For gases resistance analogy applies to layers:

$$J(z) = -K(z)\frac{\partial C}{\partial z}$$

results in:

$$J_{ij} = \frac{C(z_i) - C(z_j)}{r}, \quad r = \int_{z_i}^{z_j} \frac{dz}{K(z)}$$

r is the resistance of the layer.

“Exponential” scheme

For particles Resistance analogy does not apply.

Steady-state particle flux equation below z_1:

$$J(z) = -K(z)\frac{\partial C}{\partial z} + v(z)C = const \tag{1}$$

if $v(z) = v_s = const$ and $C(0) = 0$:

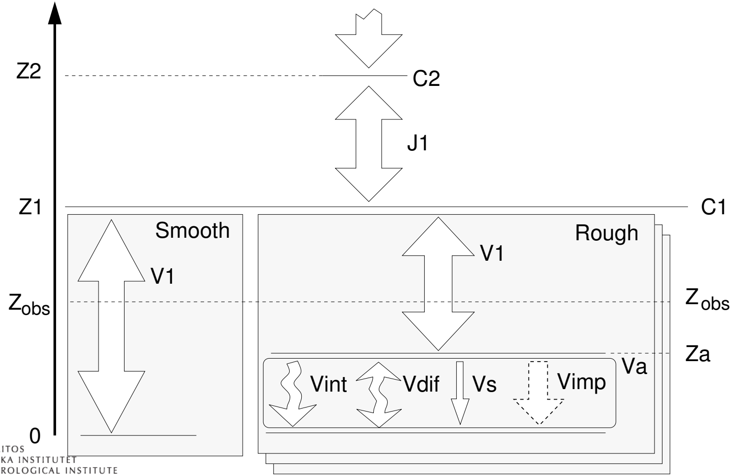
$$J(z_1) = \frac{C(z_1)}{1 - \exp(-v_s r)} v_s, \quad r = \int_0^{z_1} \frac{dz}{K(z)}$$

r is the resistance of the layer below z_1.

Eq. (1) can be also solved if $v(z) \neq const$ and for $C(0) \neq 0$.

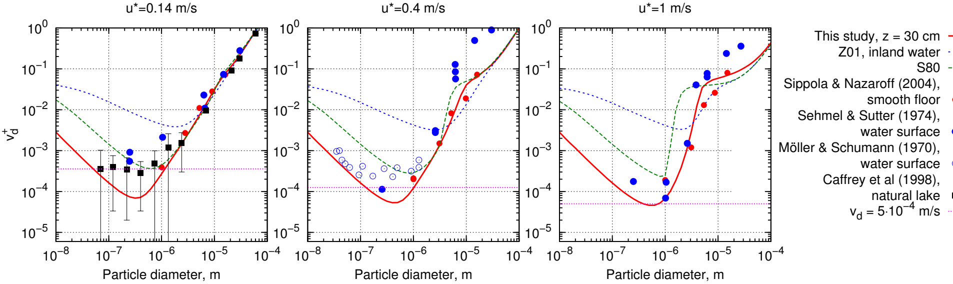
Dry deposition scheme in SILAM

- Uniform for particles and gases (surface resistances needed)
- Separate treatment of smooth and rough surfaces
- Smooth surfaces: no tuning parameters
- Rough surfaces – z_0 and “collection scale”



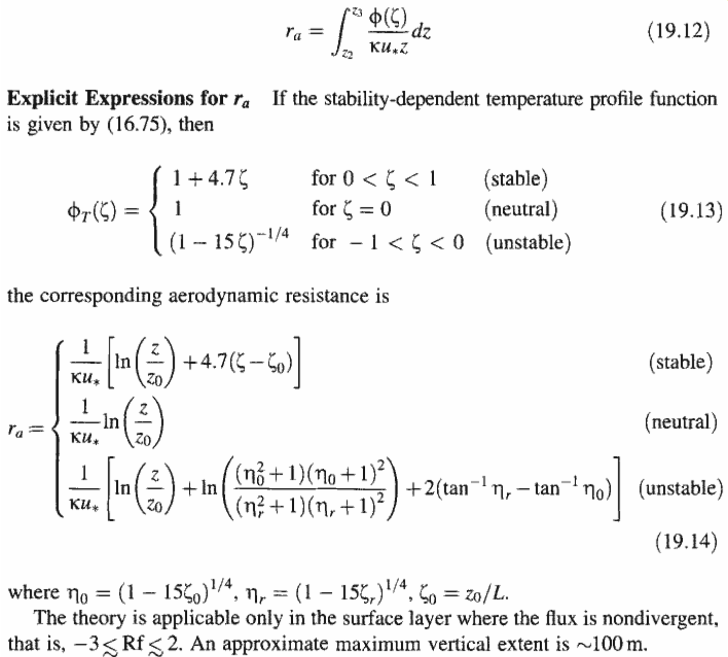
Smooth&water

Explicit solution of the steady-flux equation with turbophoresis term



- Deposition gap
- Small particles – diffusion (Sc)
- Large particles – settling ($v_s = \tau_p g$)
- Turbophoresis $v_{tf} \sim \tau_p u_*^3/\nu$
- Limit by diffusion in aerodynamic layer

Aerodynamic resistance



$$r_a = \int_{z_2}^{z_3} \frac{\phi(\zeta)}{\kappa u_* z} dz \qquad (19.12)$$

Explicit Expressions for r_a If the stability-dependent temperature profile function is given by (16.75), then

$$\phi_T(\zeta) = \begin{cases} 1 + 4.7\zeta & \text{for } 0 < \zeta < 1 & \text{(stable)} \\ 1 & \text{for } \zeta = 0 & \text{(neutral)} \\ (1 - 15\zeta)^{-1/4} & \text{for } -1 < \zeta < 0 & \text{(unstable)} \end{cases} \qquad (19.13)$$

the corresponding aerodynamic resistance is

$$r_a = \begin{cases} \frac{1}{\kappa u_*}\left[\ln\left(\frac{z}{z_0}\right) + 4.7(\zeta - \zeta_0)\right] & \text{(stable)} \\ \frac{1}{\kappa u_*}\ln\left(\frac{z}{z_0}\right) & \text{(neutral)} \\ \frac{1}{\kappa u_*}\left[\ln\left(\frac{z}{z_0}\right) + \ln\left(\frac{(\eta_0^2+1)(\eta_0+1)^2}{(\eta_r^2+1)(\eta_r+1)^2}\right) + 2(\tan^{-1}\eta_r - \tan^{-1}\eta_0)\right] & \text{(unstable)} \end{cases} \qquad (19.14)$$

where $\eta_0 = (1 - 15\zeta_0)^{1/4}$, $\eta_r = (1 - 15\zeta_r)^{1/4}$, $\zeta_0 = z_0/L$.

The theory is applicable only in the surface layer where the flux is nondivergent, that is, $-3 \lesssim \mathrm{Rf} \lesssim 2$. An approximate maximum vertical extent is ~100 m.

Rough surfaces

Simple thoughts:

- Air moves in a canopy consisting of *collectors*
- Same collectors absorb momentum and matter
- Momentum flux is (more or less) well studied
- Ratio of corresponding cross-sections gives ratio of deposition velocities

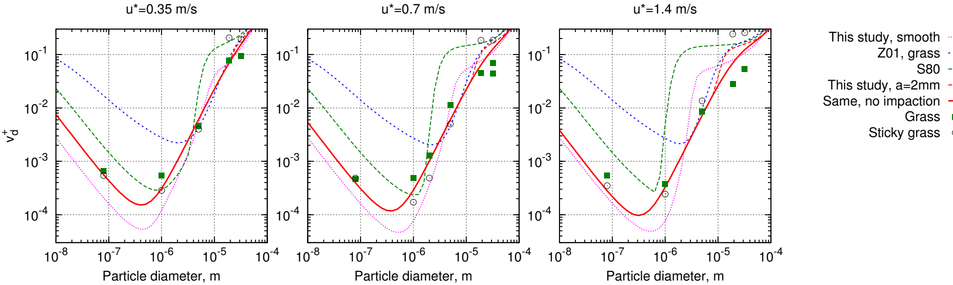
Flow-collector interaction:

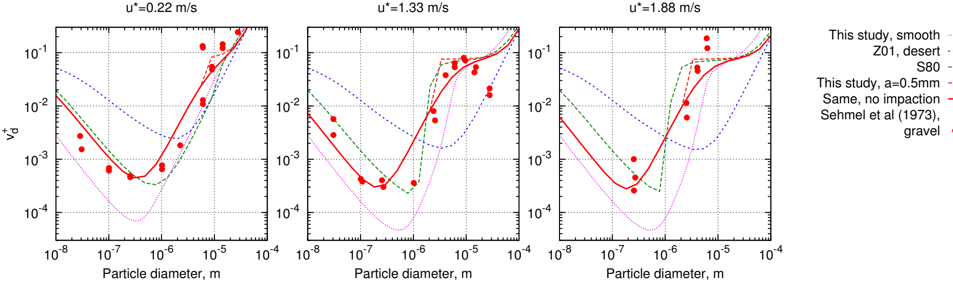
- $\mathrm{Re}_c = Ud_c/\nu$
- Relevant velocity scale is $U_{top} \simeq 3u_*$

Particle-collector interaction:

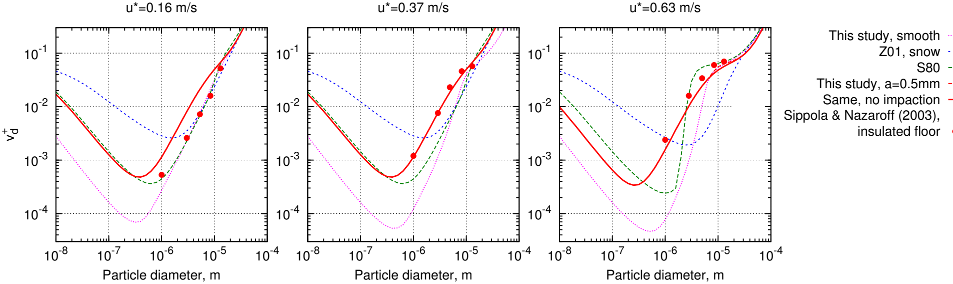
- Diffusion $\mathrm{Sc} = \nu/D$
- Interception d_p/d_c
- Impaction $\mathrm{St} = \dfrac{2\tau_p U_{top}}{d_c}$

Illustration: grass and gravel





Smooth to rough: Snow



Smooth or rough is decided from roughness Reynolds number. Transition occurs:

$$2 < u_* z_0 / \nu < 4$$

Deposition of gases

As it is seen to gurus:

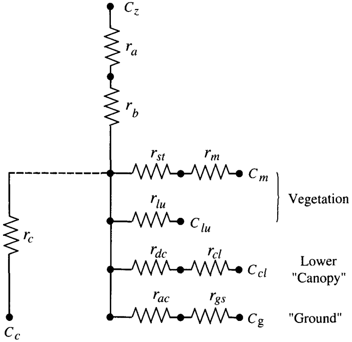


FIGURE 19.7 Resistance schematic for dry deposition model of Wesely (1989).

Weseley (1970)

As it is now:

- Above surface – as particles
- Surface resistances for each species for water and ground
- Water resistance for wet surfaces

Dry deposition Summary

- General features:
  - Deposition gap
  - Small particles – diffusion ($v_{diff} \sim \mathrm{Sc}^{-2/3} \sim d_p^{-1}$)
  - Large particles – settling ($v_s = \tau_p g \sim d_p^2$)
  - Limit by diffusion in aerodynamic layer
  - Turbophoresis $v_{tf} \sim \tau_p u_*^3/\nu$ (for smooth)
  - Interception $v_{int} \sim u_*(d_p/d_c)^2$ (for rough)
  - Impaction $v_{imp} \sim u_* f(\mathrm{St})$ (for rough)

- Open issues in SILAM:
  - Land uses (sea/land currently)
  - Surface resistances (wet/dry) currently
  - Collection scales for high vegetation

Wet deposition

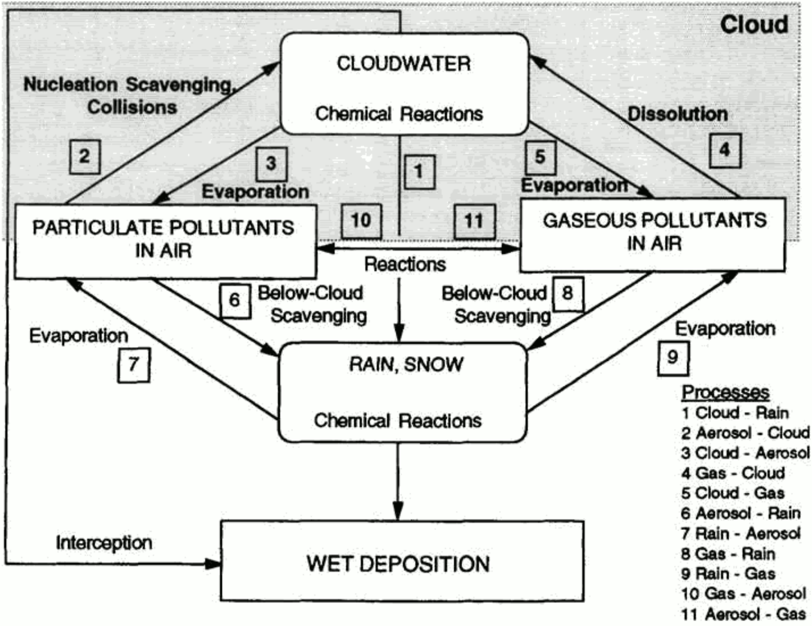


FIGURE 20.1 Conceptual framework of wet deposition processes.

Seinfeld & Pandis (2006)

Wet deposition

- In-cloud
    - Get into a droplet (ice crystal)
    - Fall out
- Sub-cloud
    - Snow
    - Rain
- Simple chemistry (dissociation)

Wet deposition in SILAM

Operational SILAM:

- Precipitation rate from meteo
- Prescribed cloud height
- Prescribed scavenging coefficients (in- below- cloud, rain or snow)
- Species-dependent scaling
- Saturation of $\mathrm{SO_2}$

In development:

- Meteo input:
  - Precipitation rates
  - Cloud water content
  - Cloud fraction
- Equilibrium in clouds
- Fraction of cloud precipitating
- Solubilities
- Simple dissociation chemistry ($\mathrm{SO_2}$)

Sub-cloud scavenging efficiency

- Diffusion D
- Interception d_p
- Impaction τ_p

Accumulation around 1 μm

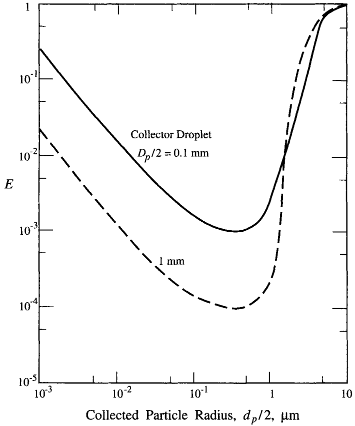


FIGURE 20.6 Semiempirical correlation for the collection efficiency E of two drops (Slinn 1983) as a function of the collected particle size. The collected particle is assumed to have unit density.

Seinfeld & Pandis (2006)

Sub-cloud scavenging rate

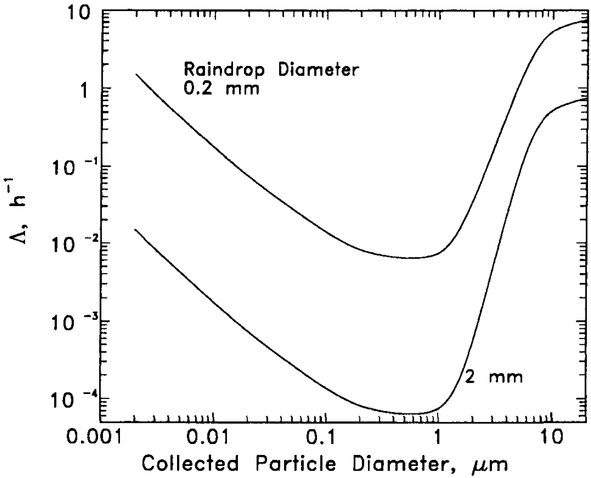


FIGURE 20.7 Scavenging coefficient for monodisperse particles as a function of their diameter collected by monodisperse raindrops with diameters 0.2 and 2 mm assuming a rainfall intensity of 1 mm h^{-1}.

Seinfeld & Pandis (2006)

Sub-cloud scavenging for gases

Relaxation between in-drop and in-air concentration, but

- high solubility – scavenging works
- low solubility – do not scavenge much

Effect of resolution

Example:

- heavy rain in half of grid cell
- rain scavenges 99% per time step
- little wind

Question:

- How much stuff will be left in air after two time steps?

Answer:

- Do not know. . .

The rain/snow structure is accounted in scavenging coefficients. . .

Effect of scavenging rate

Does not affect much:

- long-term wet deposition much
- Does affect short-term deposition patterns

Does affect:

- Short-term deposition patterns
- In-water concentrations

Gas solubility

- Henry factor – mole/(l Pa) says how much of dissolved $\mathrm{SO_2}$ is in equilibrium with 1 Pa of partial pressure
- Temperature-dependent
- Henry factor does not say "how much stuff can get into droplet at given in-air concentration"
- Reason: dissociation
- Effective Henry helps helps sometimes

For $\mathrm{SO_2}$ situation even worse...

Gas solubility (cont.)

The effective Henry factor for $\mathrm{SO_2}$ depends on pH, in particular on the amount of dissolved $\mathrm{SO_2}$

$$[\mathrm{S(IV)}] = [\mathrm{SO_2}]\left(1 + \frac{K_{S1}}{[\mathrm{H^+}]}\right)$$

Result: Solubility concept does not apply.

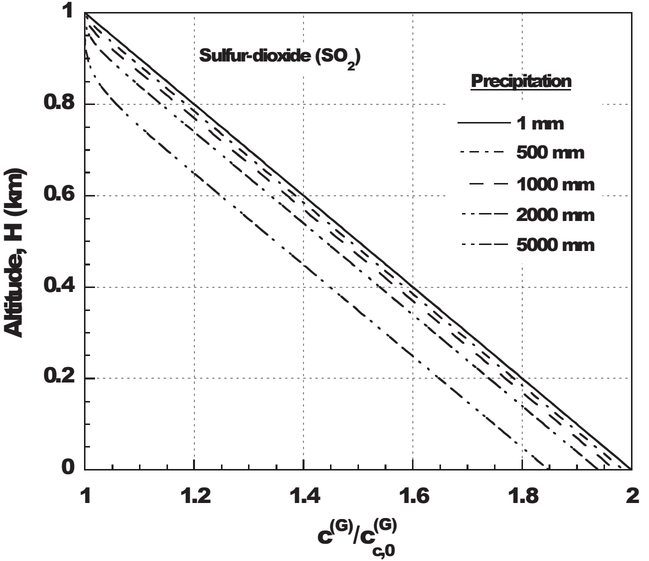
Operational SILAM: Saturation of SO2 in rain water.

New scheme – approximate electro-neutrality:

$$[\mathrm{H^+}] \simeq [\text{Strong Acids}] + [\mathrm{HSO_3^-}] - [\mathrm{NH_4^+}], \quad [\mathrm{HSO_3^-}] \simeq [\mathrm{S(IV)}].$$

Luckily, many species can be treated with “effective Henry constant”.

SO_2: how NOT to



(c) Respected authors in respected journal (IF=3.7), 2011

Summary

Dry deposition

- Not simply size
- Deposition gap (∼1μm)
- Surface resistance needed for gases: landuse and species-dependence

Wet depositoin

- Incloud and subcloud
- Deposition gap for subcloud
- Dissociation is important
- Subgrid effects have to be treated