# Effect of Rain Scavenging on Altitudinal Distribution of Soluble Gaseous Pollutants in the Atmosphere

T. Elperin, A. Fominykh and B. Krasovitov

*Department of Mechanical Engineering*
*The Pearlstone Center for Aeronautical Engineering Studies*
*Ben-Gurion University of the Negev P.O.B. 653, Beer Sheva 84105*
*ISRAEL*

A. Vikhansky

*School of Engineering and Material Science*
*Queen Mary University of London, London, UK*

# Outline of the presentation

- **Motivation and goals**
- **Fundamentals**
- **Description of the model**
- **Results and discussion**
- **Conclusions**



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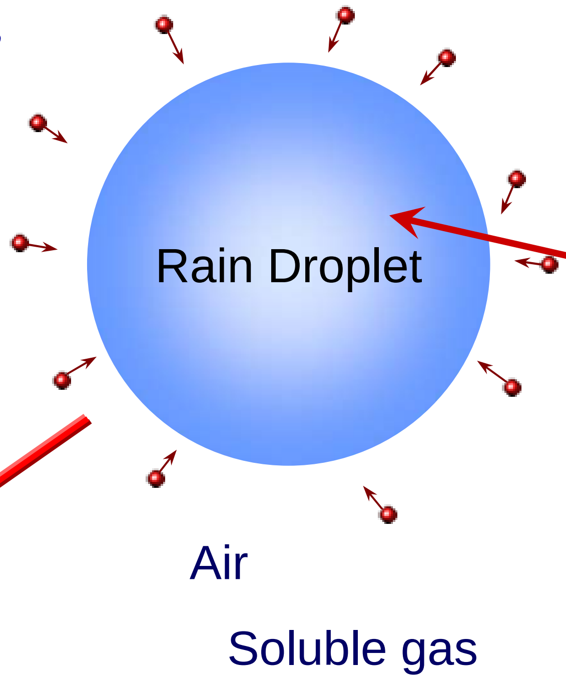
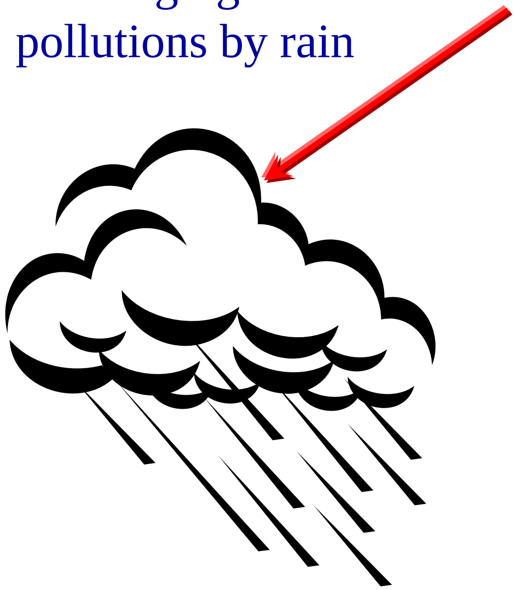
# Precipitation scavenging of gaseous pollutants by rain

Atmospheric polluted gases ($SO_2$, $CO_2$, CO, NOx, $NH_3$):

- In-cloud scavenging of polluted gases
- Scavenging of air pollutions by rain







Henry's Law:

$$A(g) + H_2O \rightleftarrows A \cdot H_2O$$

$A \cdot H_2O$ is the species in dissolved state

$$[A \cdot H_2O] = K_H p_A$$



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# Vertical concentration gradient of soluble gases

## Gaseous pollutants in atmosphere

- $SO_2$ and $NH_3$ – anthropogenic emission
- $CO_2$ – competition between photosynthesis, respiration and thermally driven buoyant

  mixing

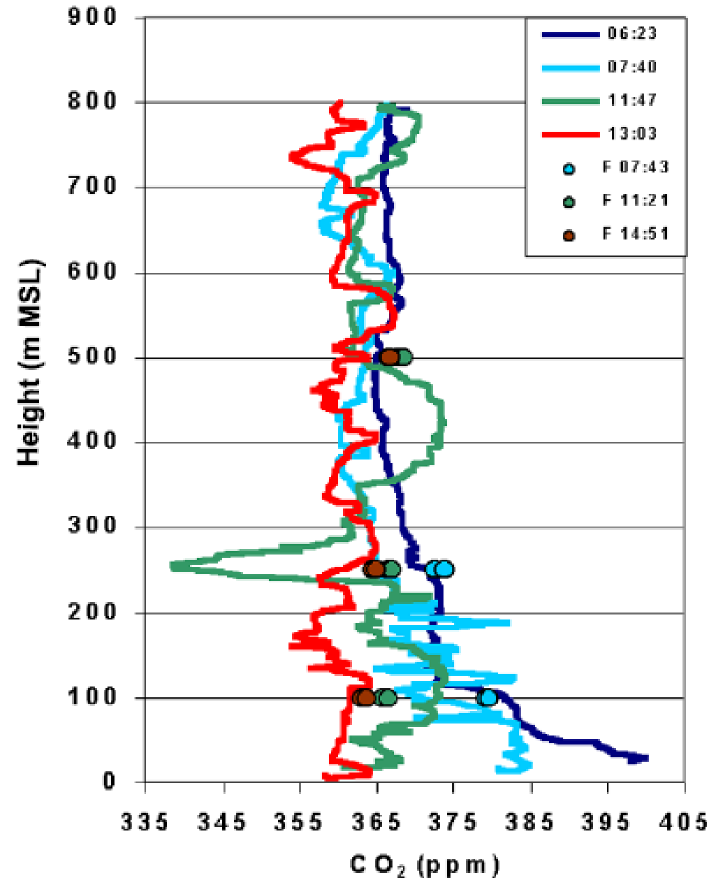


**Fig. 1a.** Aircraft observation of vertical profiles of $CO_2$ concentration (by Perez-Landa et al., 2007)

# Vertical concentration gradient of soluble gases

## Gaseous pollutants in atmosphere

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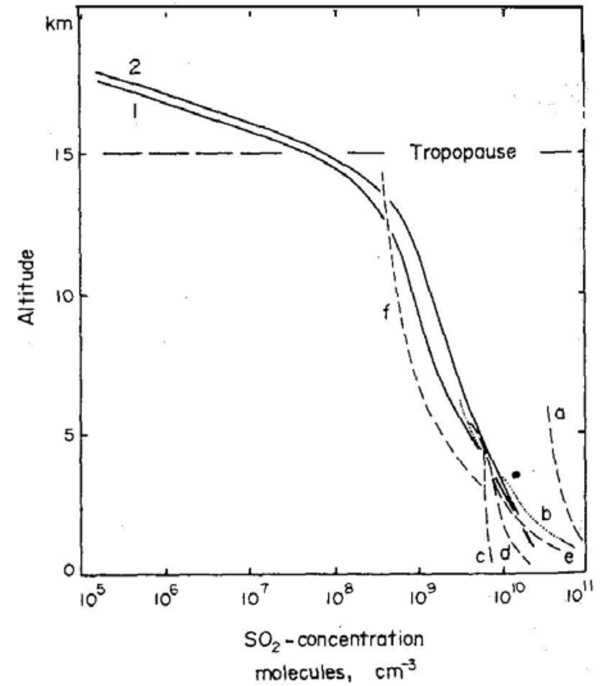



**Fig. 1b.** Vertical distribution of $SO_2$. Solid lines - results of calculations with (1) and without (2) wet chemical reaction (Gravenhorst et al. 1978); experimental values (dashed lines) – (a) Georgii & Jost (1964); (b) Jost (1974); (c) Gravenhorst (1975); Georgii (1970); Gravenhorst (1975); (f) Jaeschke et al., (1976)

# Scientific background

- **Gas absorption by rain:**
  - Asman, 1995 – uniformly distributed soluble pollutant gas
  - Slinn, 1974 – wash out of plums
  - Zhang, 2006 – wash out of soluble pollutants by drizzle
- **Measurements of vertical distribution of trace gases in the atmosphere:**
  - $SO_2$ – Gravenhorst et al., 1978
  - $NH_3$ – Georgii & Müller, 1974
  - $CO_2$ – Denning et al., 1995; Perez-Landa et al., 2007
- **Precipitation scavenging of gaseous pollutants by rain in inhomogeneous atmosphere:**
  - Elperin, Fominykh & Krasovitov 2009 – non-uniform temperature and concentration distribution in the atmosphere (single droplet)
  - Elperin et al. 2010 – Effect of Rain Scavenging on Altitudinal Distribution of Soluble Gaseous Pollutants in the Atmosphere

# Description of the model

Integral mass balance of the dissolved gas in a droplet:

$$\frac{dc^{(L)}}{dt} = \frac{1}{\tau_D}\left(mc^{(G)} - c^{(L)}\right) \qquad (1)$$

where

$\tau_D = \dfrac{a\,m}{3\beta}$ – characteristic diffusion time

$m$ – solubility parameter

$\beta$ – mass transfer coefficient in a gaseous phase

$c^{(G)}$ – concentration of a soluble gaseous pollutant in a gaseous phase

$c^{(L)}$ – mixed-average concentration of the dissolved gas in a droplet

# Description of the model

Dimensionless mass transfer coefficient for a falling droplet in a case of gaseous phase controlled mass transfer:

$$\beta = \frac{\mathrm{Sh}\, D_G}{d} \tag{1}$$

$$\mathrm{Sh} = 2 + 0.6 \cdot \mathrm{Re}^{1/2} \cdot \mathrm{Sc}^{1/3} \tag{2}$$

For small $\tau_D$ Eq. (1) yields:

$$c^{(L)} = m\left( c^{(G)} - \tau_D \frac{dc^{(G)}}{dt} \right) \tag{3}$$

Total concentration of soluble gaseous pollutant in gaseous and liquid phases reads:

$$c = (1-\phi)c^{(G)} + \phi c^{(L)} \tag{4}$$

# Description of the model

Since $$\frac{\tau_D}{c^{(G)}}\left|\frac{dc^{(G)}}{dt}\right| << 1$$

Eqs (3)-(4) yield:

$$c = c^{(G)}\left[(1-\phi)+m\phi\right] \tag{5}$$

where $\phi$ – volume fraction of droplets in the air.

The total flux of the dissolved gas transferred by rain droplets:

$$q_c = \phi \cdot u \cdot c^{(L)} \tag{6}$$

where $u$ – velocity of a droplet.

# Description of the model

Using Eqs. (3) and (6) we obtain:

$$q_c = m\phi u\left(c^{(G)} - \tau_D \frac{dc^{(G)}}{dt}\right) \tag{7}$$

Equation of mass balance for soluble trace gas in the gaseous and liquid phases:

$$\frac{\partial c}{\partial t} = -\frac{\partial q_c}{\partial z} \tag{8}$$

Combining Eqs. (3) – (8) we obtain:

$$\frac{\partial C^{(G)}}{\partial T} + \frac{\partial C^{(G)}}{\partial \eta} = \frac{1}{\mathrm{Pe}} \frac{\partial^2 C^{(G)}}{\partial \eta^2} \tag{9}$$

where $\mathrm{Pe} = UL/D,\quad T = tU/L,\quad U = \dfrac{m\phi u}{(1-\phi)+m\phi},\quad D = \dfrac{m\phi\tau \cdot u'^{\,2}}{(1-\phi)+m\phi},$

$$u' = u - U,\qquad m = H_A R_g T C_2 / C_1,\qquad C^{(G)} = c^{(G)} / c_{c,0}^{(G)}$$

# Description of the model

Peclet number:

$$\mathrm{Pe} = \frac{U \cdot L}{D} = \frac{6 D_G \cdot L}{m \cdot c_1 \cdot d^{5/2}} \cdot \left[ 2 + 0.6 \cdot \frac{c_1^{1/2} \cdot d^{3/4}}{\nu_G^{1/6} \cdot D_G^{1/3}} \right] \qquad (10)$$

where $c_1 = 130\,[\mathrm{m}^{\frac{1}{2}} \cdot \mathrm{s}^{-1}]$

***Boundary conditions:***

$$T = 0 \qquad C^{(G)} = f(\eta) \qquad (11)$$

$$\eta = 0 \qquad C^{(G)} = 1 \qquad (12)$$

$$\eta = 1 \qquad \frac{\partial C^{(G)}}{\partial \eta} = 0 \qquad (13)$$

where $\eta = z/L, \quad \eta \in [0,1]$

# Results and discussion

Feingold-Levin DSD:

$$n(d)=\frac{N_d}{\sqrt{2\pi}d\ln\sigma}\exp\left[-\frac{(\ln d-\ln d_r)^2}{2(\ln\sigma)^2}\right]$$

$$N_d=172R^{0.22}\left(\mathrm{m}^{-3}\right)$$

$$\sigma=1.43-3\cdot10^{-4}R$$

$$d_r=0.72R^{0.23}\ (\mathrm{mm})$$

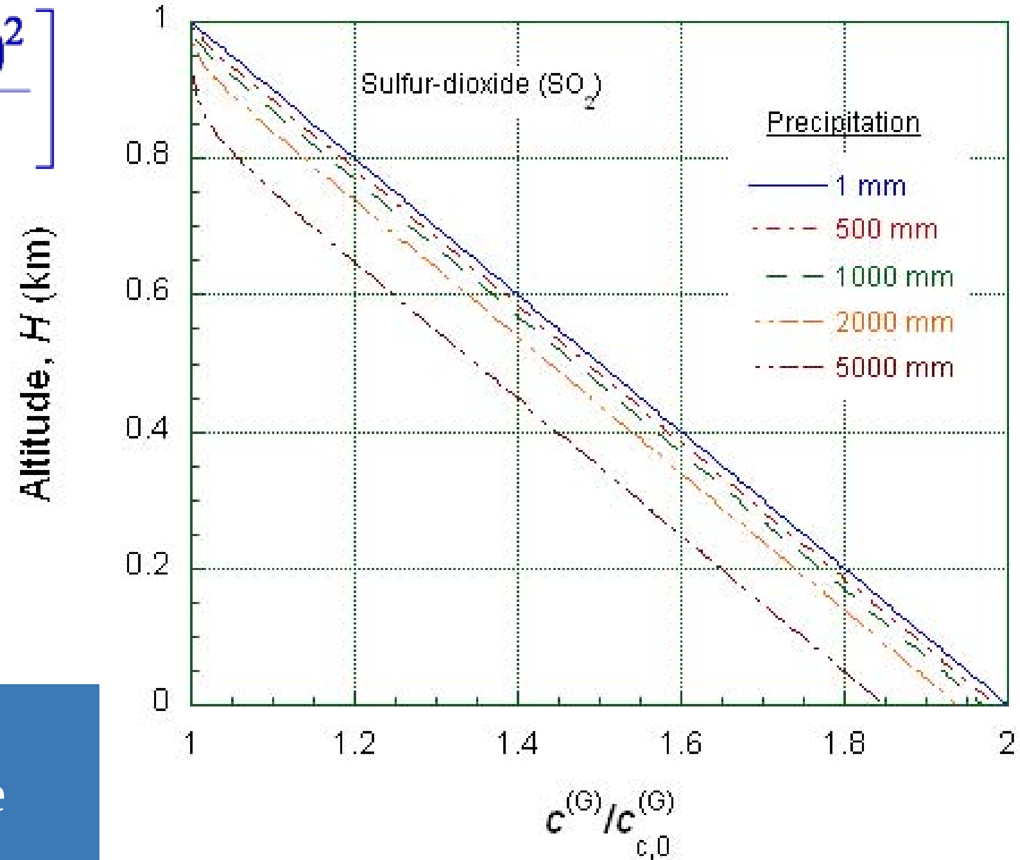
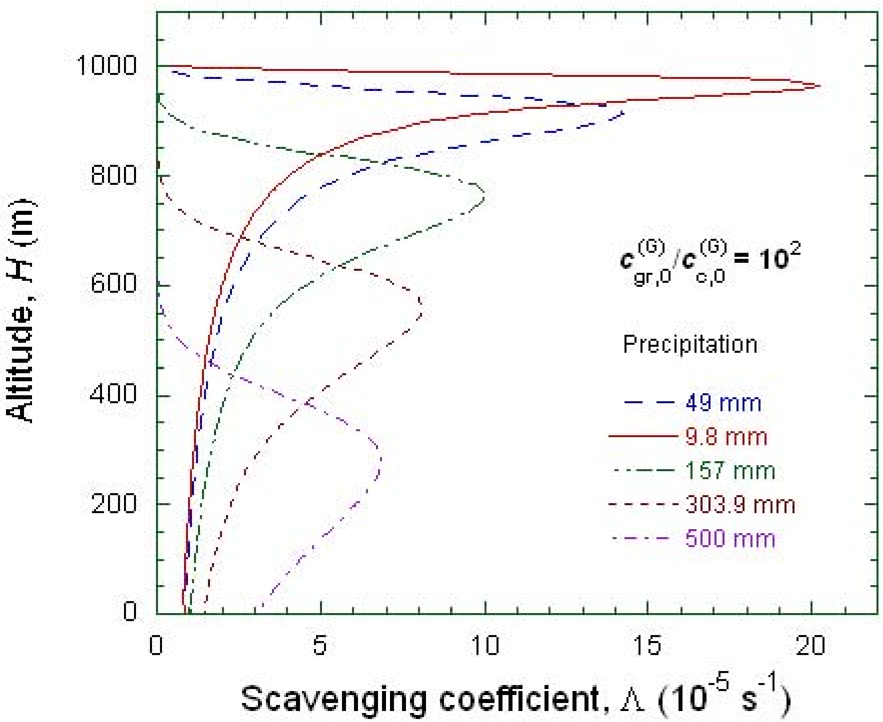



**Fig. 2.** Evolution of sulfud dioxide ($SO_2$) distribution in the atmosphere due to scavenging by rain

# Results and discussion

**Fig. 3. Dependence of scavenging coefficient vs. altitude for ammonia wash out (linear initial distribution of ammonia in the atmosphere ( $c^{(G)}_{gr,0} \big/ c^{(G)}_{c,0} = 10^2$).**



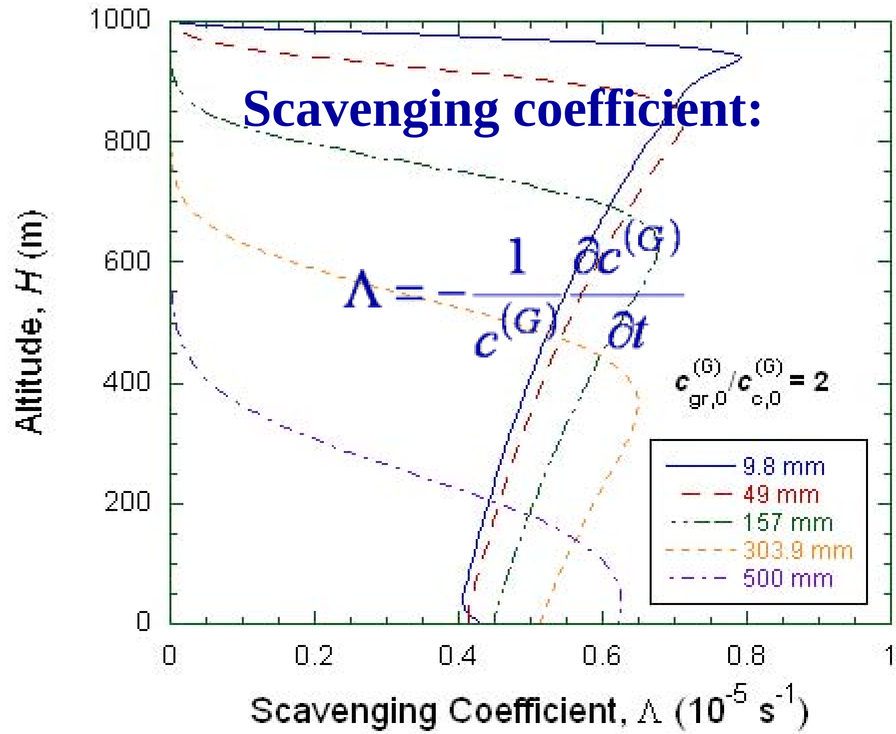




**Fig. 4. Dependence of scavenging coefficient vs. altitude for ammonia wash out (linear initial distribution of ammonia in the atmosphere ( $c^{(G)}_{gr,0} \big/ c^{(G)}_{c,0} = 2$ ).**

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# Results and discussion

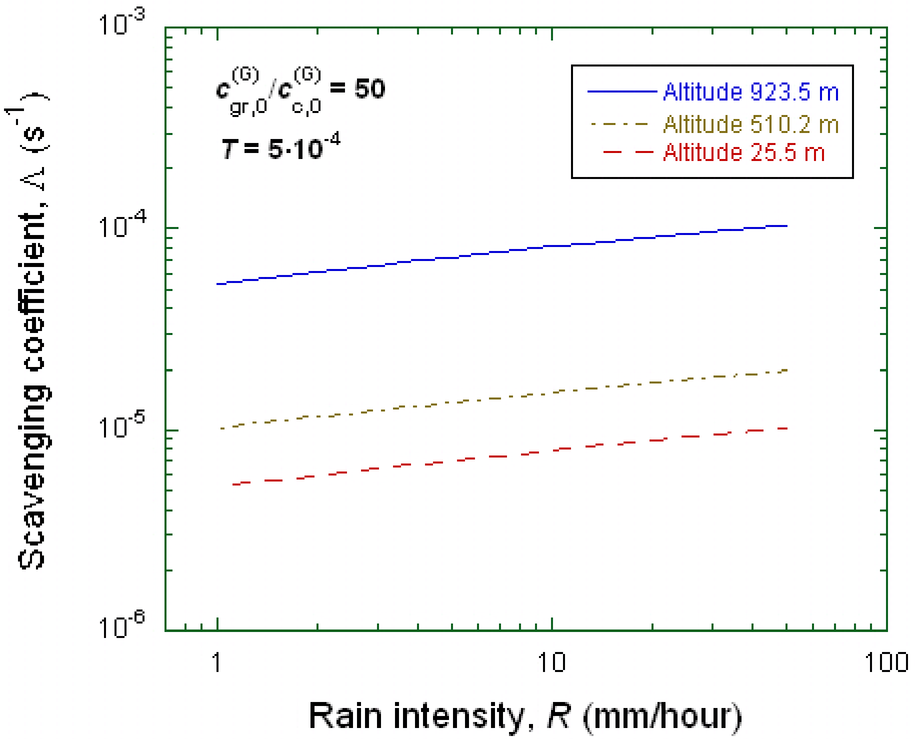
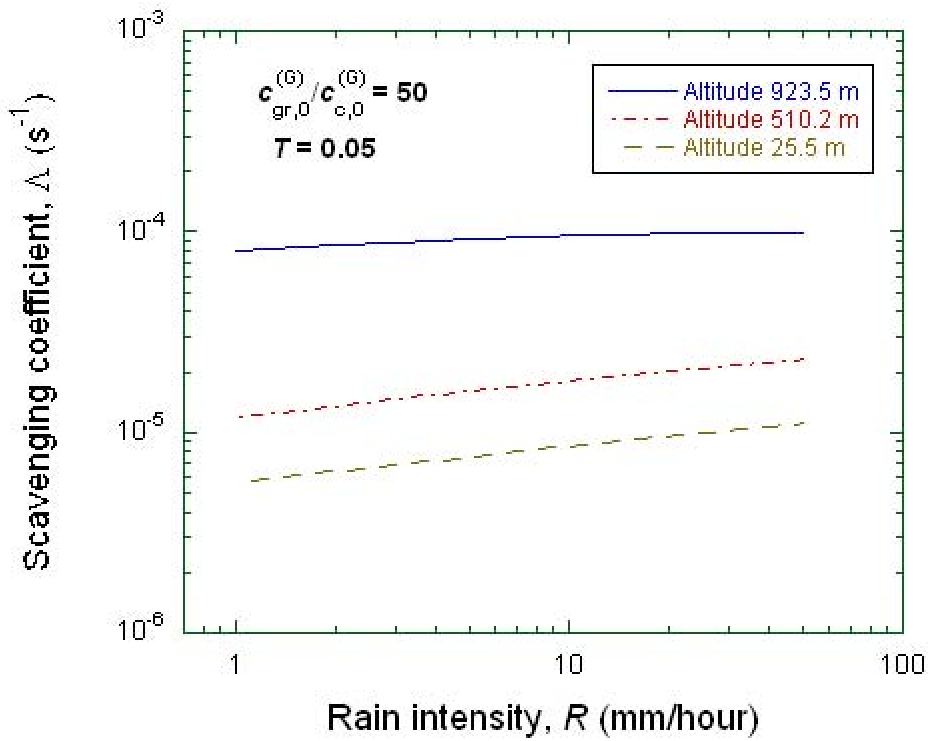



**Fig. 5. Dependence of scavenging coefficient vs. rain intensity for ammonia wash out at the early stage of rain (dimensionless time $T = 5 \cdot 10^{-4}$).**

**Fig. 6. Dependence of scavenging coefficient vs. rain intensity for ammonia wash out at the advanced stage of rain (dimensionless time $T = 0.05$).**



# Conclusions

- In this study we developed a model for scavenging of soluble trace gases in the atmosphere by rain. It is shown that gas scavenging is determined by non-stationary convective diffusion equation with the effective Peclet number that depends on droplet size distribution (DSD). The obtained equation was analyzed numerically in the case of log-normal DSD with Feingold-Levin parameterization**.**
- It is demonstrated that scavenging coefficient for the wash out of soluble atmospheric gases by rain is time-dependent.
- It is shown that scavenging coefficient in the atmosphere is height-dependent. Scavenging of soluble gas begins in the upper atmosphere and scavenging front propagates downwards with "wash down" velocity and is smeared by diffusion.
- It is found that scavenging coefficient strongly depends on the initial distribution of soluble trace gas concentration in the atmosphere. Calculations performed for linear distribution of the soluble gaseous species in the atmosphere show that the scavenging coefficient increases with the increase of soluble species gradient.