Modeling of the Vertical Transport of Polydispersed Aerosol Particles in the Atmospheric Surface Layer

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ABSTRACT

The integral model equations for determining the dry deposition and resuspension of polydispersed aerosol particles in the atmospheric surface layer to and from aerodynamic smooth and rough surfaces are derived and discussed. These model equations are based on micrometeorological ideas of height-invariant vertical transfer of momentum, sensible heat and matter as well as a representative terminal settling velocity for the entire particle size distribution. The fluxes in the turbulent region of the atmospheric surface layer are parameterized by flux-gradient relationships. The calculation of molecular-turbulent fluxes in the underlying sublayer is based on flux-gradient relationships for aerodynamic smooth surfaces (where a representative Brownian diffusion coefficient for the entire particle size distribution is taken into account), and the sublayer Stanton number as well as Reynolds' analogy between concentration, temperature and wind velocity distributions for rough surfaces.

Model results which were derived from observation data of wind velocity, dry- and wet-bulb temperatures and NH_4^+-concentrations collected in the GREIV I experiment are presented and discussed.

1. INTRODUCTION

Improved methods for determining the dry deposition of atmospheric aerosol particles on vegetation, soil, and natural water systems are needed in the fields of environmental protection, nutrient ecology, and so on. The procedure presented here is based on the method for determining the dry deposition of monodispersed aerosol particles which was derived by Sehmel (1973). Most natural and anthropogenic generated aerosol particles, however, are not of same size. The size varies by several orders of magnitude, and the polydispersed structure of the aerosol may be described by different particle size distributions (Spurny, 1985). In this study, it is attempted to consider the size distribution of the aerosol particles in a model of the atmospheric surface layer.

2. MODEL DESCRIPTION

2.1 Basic Equations

By neglecting phoretic effects and assuming horizontal homogeneity, the vertical flux F_c of aerosol particles may be described by (Businger, 1986; Kramm, 1989a)

$$F_c = - (D_c + K_c) \frac{\partial c}{\partial z} - v_T c \quad . \tag{1}$$

Here, c is the mean concentration (e.g. in $\mu g/m^3$), z is the height, D_c is the Brownian diffusion coefficient, K_c is the eddy diffusion coefficient, and v_T is the terminal settling velocity. Both D_c and v_T depend on the particle size distribution and the shape of the particles.

For spherical particles, effective values for both quantities can be derived by (e.g., Kramm, 1991)

$$D_c = \frac{\int_0^\infty n(r)\, r^3\, D_c(r)\, dr}{\int_0^\infty n(r)\, r^3\, dr} \tag{2}$$

and

$$v_T = \frac{\int_0^\infty n(r)\, r^3\, v_T(r)\, dr}{\int_0^\infty n(r)\, r^3\, dr} \quad . \tag{3}$$

Here, n(r) is the particle size distribution, $D_c(r)$ is the Brownian diffusion coefficient for particles of radius r, and $v_T(r)$ is their terminal settling velocity.

The determination of particle fluxes on the basis of Eqs. (1) to (3) requires a detailed knowledge of D_c, v_T and K_c. In the following these individual components are illuminated in the frame of particle size distributions (to specify D_c and v_T) and a micrometeorological transfer and deposition module (to specify K_c and F_c in the turbulent region of the atmospheric surface layer). Note, that D_c is several orders of magnitude smaller than K_c in the turbulent layer, but is dominating in the viscous sublayer near the surface.

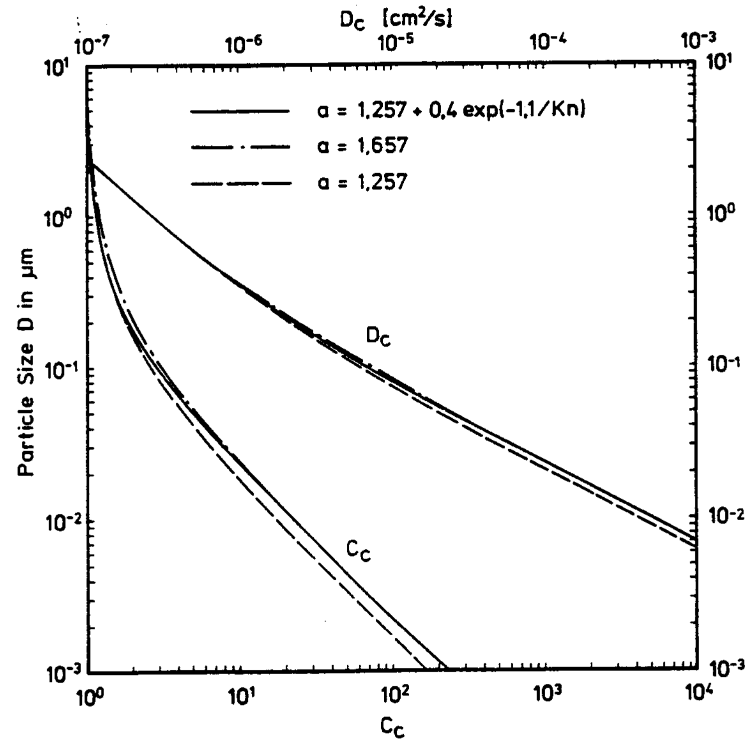

FIGURE 1: Diffusion coefficient D_c and slip flow correction factor C_c as functions of the particle size D (with reference to Seinfeld, 1986).

2.2 D_c and v_T Characteristics

2.2.1 Slip Flow Corrections

In the integral term of Eq. (2) the function $D_c(r)$ is given by the slip flow corrected Stokes-Einstein relation (e.g. Pruppacher and Klett, 1980)

$$D_c(r) = \frac{k\ T\ C_c}{6\ \pi\ \nu\ \ \rho_L\ r} \quad , \tag{4}$$

where k is the Boltzmann constant, T is the air temperature in K, ν is the kinematic viscosity of the air, and ρ_L is the density of air.

The terminal settling velocity $v_T(r)$ in Eq. (3) is given by the slip flow corrected Stokes' terminal velocity (e.g. Pruppacher and Klett, 1980)

$$v_T(r) = \frac{2}{9}\ \frac{g}{\nu}\ \frac{\rho_P - \rho_L}{\rho_L}\ r^2\ C_c \approx \frac{2}{9}\ \frac{g\ \rho_P}{\nu\ \rho_L}\ r^2\ C_c \quad , \tag{5}$$

where ρ_p is the density of the particle. In equations (4) and (5) the slip flow correction C_c is a function of the Knudsen number $Kn = \lambda/r$ (with λ being the mean free path). It is expressed by the Cunningham correction factor

$$C_c = 1 + \alpha\ Kn \qquad (6)$$

with (Pruppacher and Klett, 1980)

$$\alpha = 1.257 + 0.4\ \exp(-1.1/Kn) \quad , \qquad (7)$$

which can be approximated by

$$\alpha = \begin{cases} 1.257 \quad , & \text{if } Kn \ll 1 \quad , \\ 1.657 \quad , & \text{if } Kn \gg 1 \quad . \end{cases} \qquad (8)$$

Figure 1 shows that both approximations are enveloping the exact function. Note, that the approximation for large Knudsen numbers (small particles) is also an excellent approximation over the total particle size range.

2.2.2 SRA-Particle Size Distribution

Generally, the particle size distribution n(r) in Eqs. (2) and (3) is a result of time-dependent (and space-dependent) micro-physical and chemical interactions (e.g., Herbert and Beheng, 1986). However, in the frame of the constant flux approach implying steady-state conditions, time-invariant particle size distributions, such as the r^{-3}-distribution (Junge, 1963), the modified γ-distribution (Deirmendjian, 1963) and the log-normal distribution (e.g., Jaenicke, 1985), may be taken into consideration.

The International Radiation Commission (IRC) suggested a sum of three log-normal distributions as the SRA-distribution (SRA = Standard Radiation Atmosphere) for urban and industrial areas (Deepak and Gerber, 1983), i.e.:

$$\frac{dN}{dr} = n(r) = \frac{1}{r} \sum_{i=1}^{3} \frac{N_i}{(2\ \pi)^{1/2}\ \ln \sigma_i} \exp\{-\frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\} \qquad (9)$$

TABLE I: Characteristics of the SRA-distribution

Aerosol component	R_i (μm)	σ_i	C_i	Ω_i
water-soluble part. e.g. $(NH_4)_2SO_4$	$2.85 \cdot 10^{-2}$	2.239	0.61	$8.416 \cdot 10^{-2}$
dust-like particles e.g. SiO2	$4.71 \cdot 10^{-1}$	2.512	0.17	$2.125 \cdot 10^{-1}$
soot particles	$1.18 \cdot 10^{-2}$	2.000	0.22	$9.158 \cdot 10^{-1}$

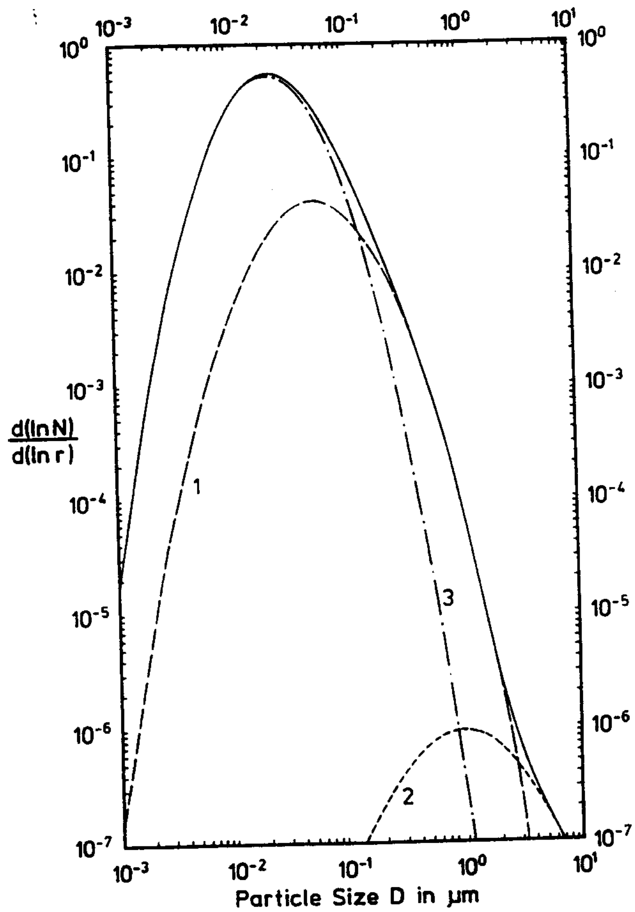

FIGURE 2: Normalized SRA-distribution of the urban-industrial aerosols (————) (Deepak and Gerber, 1983),
Mode 1 (—— ——): water-soluble particles (e.g. $(NH_4)_2SO_4$)
Mode 2 (--------): dust-like particles (e.g. SiO_2)
Mode 3 (——·——): soot particles.

or in normalized form (see Figure 2)

$$\frac{d(\ln N)}{d(\ln r)} = n^*(r) = \sum_{i=1}^{3} \frac{\Omega_i}{(2\ \pi)^{1/2} \ln \sigma_i} \exp\{- \frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\} \quad (10)$$

with

$$N_i = \Omega_i\ N = \frac{\frac{C_i}{v_i}}{\sum_{i=1}^{3} \frac{C_i}{v_i}} N \quad , \quad (11)$$

and

$$v_i = \frac{4}{3} \frac{\pi}{N_i} \int_0^\infty n_i(r)\ r^3\ dr = \frac{4}{3} \pi\ R_i^3\ \exp\{4.5\ (\ln \sigma_i)^2\} \quad . \qquad (12)$$

Here, $N = \Sigma\ N_i$ is the total number concentration of particles, N_i is the integral for the individual log-normal distribution, σ_i is the geometric standard deviation, R_i is the mean radius, and $C_i = N_i\ v_i/(\Sigma\ N_i\ v_i)$ is the individual volume normalized by the total volume. Table I summarizes the values proposed by the IRC.

For this sum of log-normal distributions one obtains

$$D_c = \frac{k\ T}{6\ \pi\ \nu\ \rho_L} \frac{\displaystyle\sum_{i=1}^{3} \frac{\Omega_i}{(2\ \pi)^{1/2}\ \ln \sigma_i} \int_0^\infty r\ (1 + \alpha\ \frac{\lambda}{r})\ \exp\{- \frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\}\ dr}{\displaystyle\sum_{i=1}^{3} \frac{\Omega_i}{(2\ \pi)^{1/2}\ \ln \sigma_i} \int_0^\infty r^2\ \exp\{- \frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\}\ dr} \qquad (13)$$

and

$$v_T = \frac{2}{9} \frac{g}{\nu\ \rho_L} \frac{\displaystyle\sum_{i=1}^{3} \frac{\rho_{P,i}\ \Omega_i}{(2\ \pi)^{1/2}\ \ln \sigma_i} \int_0^\infty r^4\ (1 + \alpha\ \frac{\lambda}{r})\ \exp\{- \frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\}\ dr}{\displaystyle\sum_{i=1}^{3} \frac{\Omega_i}{(2\ \pi)^{1/2}\ \ln \sigma_i} \int_0^\infty r^2\ \exp\{- \frac{(\ln(r/R_i))^2}{2\ (\ln \sigma_i)^2}\}\ dr} \qquad (14)$$

Approximative Solutions

The integrals which contain the slip flow corrections cannot be solved in an elementary manner. However, estimates for the upper and the lower limits of these integrals can be derived on the basis of the relationship (see Eqs. (7) and (8))

$$1.257\ Kn \le Kn\ (1.257 + 0.4\ \exp(-\ 1.1/Kn)) \le 1.657\ Kn$$

for which the following solutions exist:

$$D_c = \frac{k\ T}{6\ \pi\ \nu\ \rho_L} \frac{\sum_{i=1}^{3} \Omega_i\ R_i\ \{R_i\ \exp(2(\ln\ \sigma_i)^2)\ +\ \alpha\ \lambda\ \exp(0.5(\ln\ \sigma_i)^2)\}}{\sum_{i=1}^{3} \Omega_i\ R_i^3\ \exp(4.5(\ln\ \sigma_i)^2)} \tag{15}$$

and

$$v_T = \frac{2}{9} \frac{g}{\nu\ \rho_L} \frac{\sum_{i=1}^{3} \rho_{P,i}\ \Omega_i\ R_i^4\ \{R_i\ \exp(12.5(\ln\ \sigma_i)^2)\ +\ \alpha\ \lambda\ \exp(8(\ln\ \sigma_i)^2)\}}{\sum_{i=1}^{3} \Omega_i\ R_i^3\ \exp(4.5(\ln\ \sigma_i)^2)} \tag{16}$$

For the arbitrary values $T = 293.15$ K, $p_L = 1013.25$ hPa (p_L is the air pressure), and $\rho_{P,i} = 1\ g/cm^3$, $i = 1, \ldots, 3$, one obtains (Kramm, 1991)

$$D_c = \begin{cases} 4.87 \cdot 10^{-6}\ cm^2/s\ , & \text{for } \alpha = 1.657\ , \\ 3.97 \cdot 10^{-6}\ cm^2/s\ , & \text{for } \alpha = 1.257\ , \\ 1.17 \cdot 10^{-6}\ cm^2/s\ , & \text{for } \alpha = 0 \end{cases}$$

and

$$v_T = \begin{cases} 0.4068\ cm/s\ , & \text{for } \alpha = 1.657\ , \\ 0.4063\ cm/s\ , & \text{for } \alpha = 1.257\ , \\ 0.4045\ cm/s\ , & \text{for } \alpha = 0\ . \end{cases}$$

Comparison with the Jaenicke-Distribution

Jaenicke (1985) suggested another set of values for his sum of three log-normal distributions for urban aerosols (see Table II):

TABLE II: Characteristics of the Jaenicke-distribution

Aerosol component	R_i (μm)	σ_i	C_i	Ω_i
Mode 1	$6.51 \cdot 10^{-3}$	1.758	0.005	$7.258 \cdot 10^{-1}$
Mode 2	$7.14 \cdot 10^{-3}$	4.634	0.653	$8.113 \cdot 10^{-3}$
Mode 3	$2.48 \cdot 10^{-2}$	2.173	0.342	$2.661 \cdot 10^{-1}$

The same arbitrary values for T, p_L and $\rho_{P,i}$ yield the following results (Kramm, 1991):

$$D_c = \begin{cases} 1.49\cdot10^{-6}\ cm^2/s , & \text{for } \alpha = 1.657 , \\ 1.23\cdot10^{-6}\ cm^2/s , & \text{for } \alpha = 1.257 , \\ 4.33\cdot10^{-7}\ cm^2/s , & \text{for } \alpha = 0 \end{cases}$$

and

$$v_T = \begin{cases} 59.37\ cm/s , & \text{for } \alpha = 1.657 , \\ 59.36\ cm/s , & \text{for } \alpha = 1.257 , \\ 59.35\ cm/s , & \text{for } \alpha = 0 . \end{cases}$$

Results of Estimates

The results of these estimates show that 1) in the case of D_c the slip flow correction is important, and 2) in the case of v_T the slip flow correction is negligible, which can also be expressed by $\alpha = 0$. Note that for both distributions the results for the terminal settling velocities of the particle ensembles are nearly the same as the terminal settling velocities of the mode 2 of both particle ensembles, i. e. the contributions of Aitken nuclei and coarse particles to v_T are negligible. Since mode 2 of the Jaenicke distribution contains much more giant particles than mode 2 of the SRA-distribution, it is reasonable that its terminal settling velocity differs from that of the SRA-distribution by two orders of magnitude.

3. Micrometeorological Transfer and Flux Relations

3.1 Eddy Diffusivities

The eddy diffusivity K_c in Eq. (1) is related to the eddy diffusivities for heat (K_h) and momentum (K_m) by

$$K_c = K_h , \tag{17}$$

$$K_h = \begin{cases} K_m , & \text{if } Ri \geq 0 , \\ K_m\ (1 - 16\ Ri)^{1/4} , & \text{if } Ri < 0 , \end{cases} \tag{18}$$

$$K_m = \varkappa\ (z - d)\ u_* \begin{cases} (1 - 5\ Ri) , & \text{if } 0 \leq Ri < Ri_{cr} , \\ (1 - 16\ Ri)^{1/4} , & \text{if } Ri < 0 , \end{cases} \tag{19}$$

where Ri is the Richardson number, $Ri_{cr} = 0.2$ is the critical Richardson number, $\varkappa = 0.4$ is the von Kármán constant, d is the zero-plane displacement, and u_* is the friction velocity. Relations (18) and (19) were derived from the non-dimensional wind and temperature gradients empirically determined by Dyer and Hicks (1970) in unstable air and by Webb (1970) in stable air, where the dimensionless stability parameters Ri and $\varsigma = (z - d)/L$ (L is the Monin-Obukhov stability length) are related to each other by

$$Ri = \begin{cases} \varsigma/(1 + 5\ \varsigma) , & \text{if } L > 0 , \\ \varsigma , & \text{if } L < 0 , \end{cases} \tag{20}$$

with

$$L = \frac{u_*^2}{\varkappa \frac{g}{\Theta_m} (\Theta_* + 0.608\ \Theta_m\ q_*)} \quad . \tag{21}$$

The quantities Θ_* and q_* are the scaling parameters for heat and water vapour, and Θ_m is a characteristic temperature for the atmospheric surface layer.

3.2 Deposition and Resuspension Relations

Integrating Eq. (1) over the constant flux layer $z_G \le z \le z_R$ provides (Kramm, 1991)

$$F_c = - \frac{v_T\ c_R}{1 - \exp(-\ v_T\ r_a)} \left(1 - \frac{c_G}{c_R} \exp(-\ v_T\ r_a)\right) \quad . \tag{22}$$

Here, c_R is the concentration at the reference height z_R and c_G is the concentration at the height z_G very close to the surface. The quantity r_a is the aerodynamic resistance which is more closely discussed in section 3.3. Note that Eq. (22) leads to the flux equation for long-lived trace gases when v_T decreases to zero ($v_T \rightarrow 0$):

$$F_c = - \frac{1}{r_a} (c_R - c_G) \quad . \tag{23}$$

The expression $\mu_p = c_G/c_R \exp(-v_T\ r_a)$ in Eq. (22) determines the direction of the vertical particle flux, i.e.

$$F_c \begin{cases} < 0 \quad , & \text{if } \mu_p < 1 \text{ (Deposition)} \quad , \\ = 0 \quad , & \text{if } \mu_p = 1 \text{ (Compensation)} \quad , \\ > 0 \quad , & \text{if } \mu_p > 1 \text{ (Resuspension)} \quad . \end{cases} \tag{24}$$

If $\mu_p \ll 1$ ($c_G \ll c_R \exp(v_T\ r_a)$), the deposition flux can be calculated approximately by

$$F_c = - \frac{v_T\ c_R}{1 - \exp(-\ v_T\ r_a)} \quad . \tag{25}$$

Equation (25) was derived by Sehmel (1973) for monodispersed aerosols.

The resuspension factor $f_c = c/M_c$ and the resuspension rate $\Lambda_c = R_c/M_c$ are used to describe the resuspension ($\mu_p > 1$) of particulate matter. Here, M_c is the contamination of the surface (in $\mu g/m^2$), and R_c is the resuspension flux which is equal to F_c in the case of $\mu_p > 1$. Resuspension factor and resuspension rate can be used to formulate a resuspension velocity $v_r = \Lambda_c/f_c$. For $\mu_p > 1$, one obtains

$$F_c = \frac{v_T\ c_R}{(1 + \frac{f_{c,G}}{\Lambda_c})\ \exp(-\ v_T\ r_a)\ -\ 1} \quad , \tag{26}$$

where $f_{c,G}$ is the resuspension factor at the height z_G which can be estimated by

$$f_{c,G} = \frac{F_c\ (1\ -\ \exp(-\ v_T\ r_a))\ +\ v_T\ c_R}{M_c\ v_T\ \exp(-\ v_T\ r_a)} \quad . \tag{27}$$

3.3 Layer Resistances

In the particle flux relations (section 3.2) the aerodynamic resistance r_a proves to be of fundamental importance. It is defined by $r_a = r_t + r_{mt}$, where r_t is the resistance of the turbulent region of the surface layer against mass transfer, and r_{mt} is the resistance of the underlying thin molecular-turbulent sublayer. r_t is given by (Kramm, 1989b)

$$r_t = \int_{\delta}^{z_R} K_h^{-1}\ dz = \frac{1}{u_*\ \varkappa}\ (\ln \frac{z_R\ -\ d}{\delta\ -\ d}\ -\ \Psi_h(\zeta_R,\ \zeta_\delta)) \quad . \tag{28}$$

In the frame of the constant flux approach r_t is related to the turbulent height-invariant scaling parameters u_*, θ_*, and q_* (see section 3.1) via

$$u_* = \frac{\varkappa\ (U_R\ -\ U_\delta)}{\ln \frac{z_R\ -\ d}{\delta\ -\ d}\ -\ \Psi_m(\zeta_R,\ \zeta_\delta)} \quad , \tag{29}$$

and

$$\begin{Bmatrix} \theta_* \\ \\ q_* \end{Bmatrix} = \frac{1}{u_*\ r_t} \begin{Bmatrix} \theta_R\ -\ \theta_\delta \\ \\ q_R\ -\ q_\delta \end{Bmatrix} \quad , \tag{30}$$

where u_*, θ_* and q_* can be determined on the basis of wind speed, U, potential temperature, θ, and specific humidity, q, at the heights δ and z_R. $\Psi_m(\zeta_R,\ \zeta_\delta)$ and $\Psi_h(\zeta_R,\ \zeta_\delta)$ are the dimensionless integral stability functions for momentum and heat, respectively (Kramm and Herbert, 1984; Kramm, 1989b).

The molecular-turbulent resistance r_{mt} is given by

$$r_{mt} = \int_{z_G}^{\delta} (D_c\ +\ K_h)^{-1}\ dz = \frac{1}{u_*}\ (\frac{U_\delta}{u_*}\ +\ B^{-1}) \quad , \tag{31}$$

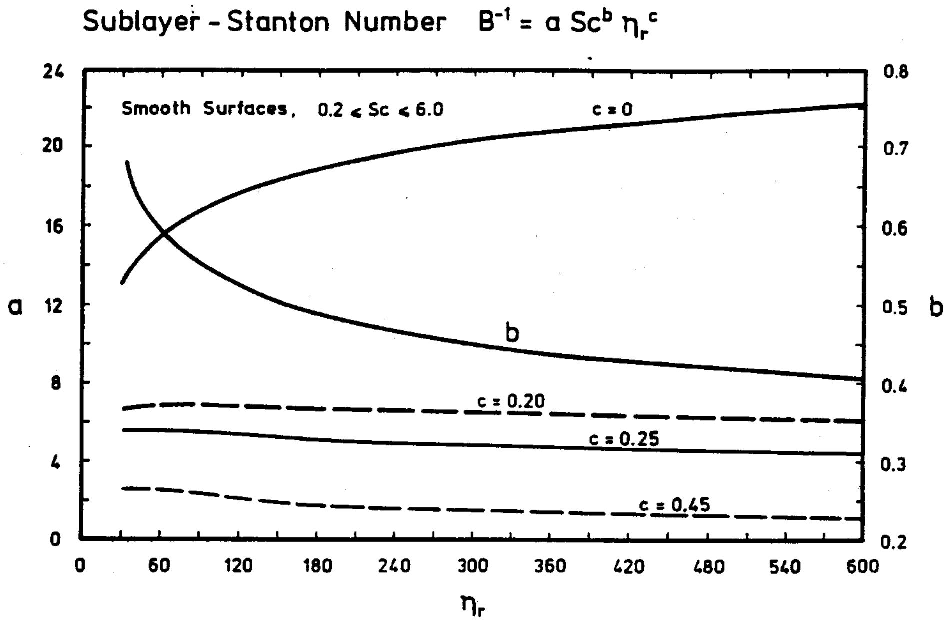

FIGURE 3: Coefficients a and b of the B^{-1} relationship for smooth surfaces as different functions of the dimensionless height $\eta_r = u_* \delta/\nu$. The results are based on the numerical solution of the integral expression in Eq. (32).

with (Müller *et al.*, 1991)

$$B^{-1} = Sc \int_{\eta_\delta}^{\eta_r} \frac{d\eta}{(1 + Sc\ K_m/\nu)} \quad . \tag{32}$$

Here, U_δ is the wind speed at the height δ (which separates the two regions of the atmospheric surface layer), $Sc = \nu/D_c$ is the Schmidt number, and B is the sublayer Stanton number.

For rough surfaces B may be calculated on the basis of the formula

$$B^{-1} = a\ Sc^b\ \eta_r^c + d \quad , \tag{33}$$

with $a = 0.52$, $b = 0.8$, $c = 0.45$, and $d = 0$, empirically derived by Owen and Thomson (1963). Other empirical relations of the same structure exist; Brutsaert (1982), for instance, suggested the following values: $a = 7.3$, $b = 0.5$, $c = 0.25$, and $d = -5$.

For smooth surfaces, the integral in Eq. (32) was numerically solved by Romberg integration techniques using Reichardt's (1951) K_m-approach

$$\frac{K_m}{\nu} = \varkappa \left(\eta - \eta_D \tanh \frac{\eta}{\eta_D}\right) \quad , \tag{34}$$

where $\eta = u_* z/\nu$ is a dimensionless height or a special Reynolds number, $\eta_D = 11$ is an empirical constant (which defines the thickness of the viscous sublayer) determined by Reichardt to fit his measured velocity profiles. Results of the numerical solutions are described in terms of $B^{-1} = a\, Sc^b\, \eta_r^c$, similar to the empirical formulae for rough surfaces. The dependence of the coefficients a and b on the upper integration limit η_r (= $u_* \delta/\nu$) for different c-values is shown in Figure 3. The exponent b does not depend on c, but varies considerably with η_r. The coefficient a evidences an interrelation with c and shows the largest variation for c = 0. Note, that the values suggested by Deacon (1977), i.e. a = 15.2, b = 0.61, and c = 0 (for $0.6 < Sc_c < 10$ and $\eta_r = 50$), are confirmed by our results. The same is true for the other special case ($\eta_r = 30$) published by Brutsaert (1982) with a = 13.6, b = 2/3, and c = 0 (d = -13.5, see Eq. (33)).

The upper limit of $\eta_r = 600$ in Figure 3 corresponds to δ = 4.5 cm when $u_* \approx 0.2$ m/s and $\nu \approx 1.5 \cdot 10^{-5}\ m^2\ s^{-1}$ is adopted.

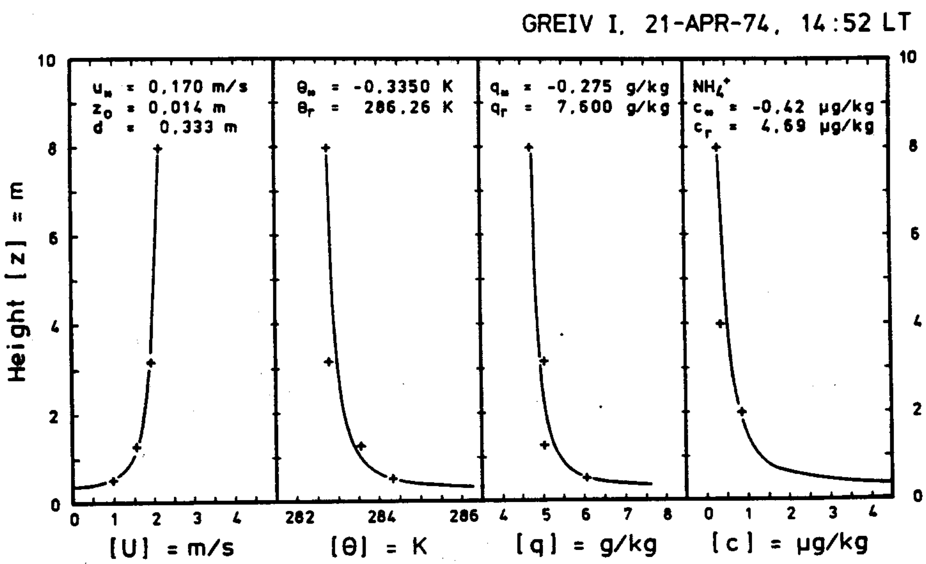

FIGURE 4: Vertical profiles of wind speed (U), potential temperature (θ) and specific humidity (q) as well as NH_4^+-concentrations (calculated with F_c = const. and v_T = 0) for unstable air ($\theta_* < 0$). The crosses represent the observed values and the lines the calculated profiles (Kramm, 1991).

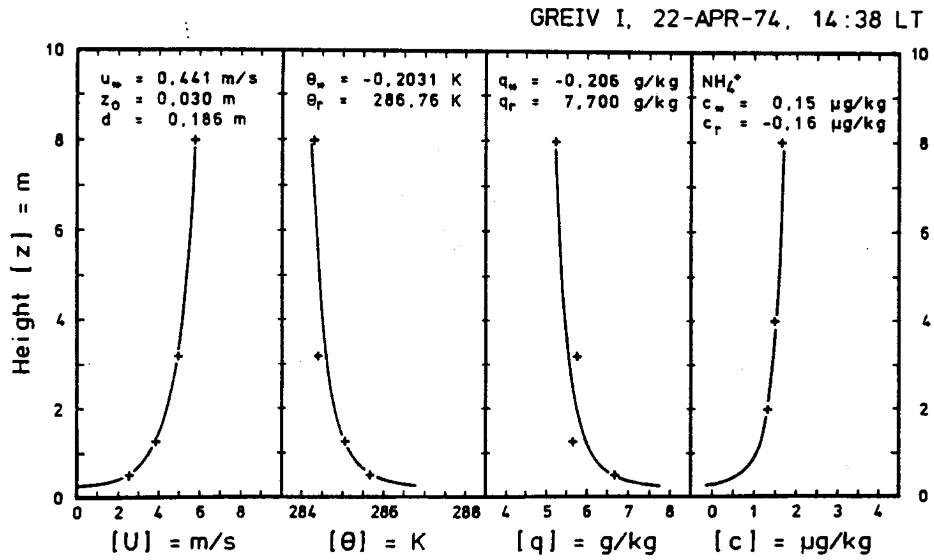

FIGURE 5: Vertical profiles of wind speed (U), potential temperature (θ) and specific humidity (q) as well as NH_4^+-concentrations (calculated with F_c = const. and $v_T = 0$) for unstable air ($\theta_* < 0$). The crosses represent the observed values and the lines the calculated profiles (Kramm, 1991).

3. TEST RESULTS AND CONCLUSIONS

Vertical profile data of wind speed, temperature, humidity, and particle concentrations as well as particle size distributions are required to determine the deposition (or resuspension) fluxes. Unfortunately, only a few profile data sets exist, which allow testing of numerical methods as described here. Moreover, a check of our method by comparison with simultaneously measured particle fluxes has to be postponed until such fluxes are available.

Test calculations with the deposition model were carried out with observation data from the GREIV I experiment (Beyer and Roth, 1976) which took place over a flat site, covered with winter barley (about 25 cm high) and rape (50-75 cm high), near Meppen/Emsland in northern Germany in April 1974. Data sets of wind speed, dry- and wet-bulb temperatures (simultaneously measured 30-min averages) obtained by the group from the University of Kiel were used for the calculation of the turbulent fluxes of momentum, sensible heat and water vapour (Kramm, 1989c). The measurements were carried out at heights of 0.5, 1.26, 3.18 and 8 m. Vertical particle fluxes were estimated on the basis of NH_4^+-concentrations measured (2 hours sampling intervals) at heights of 2, 4, 8 and 16 m. These measurements were performed by the group of the University of Frankfurt, where, however, the size distribution of the NH_4^+-particles was not determined. Since our estimates have shown that the contributions of water-soluble particles (mode 1) to the terminal settling velocities are small, sedimentation effects were neglected. Furthermore, since no data observed close to the surface are available, only the fully turbulent part of the atmospheric surface layer is taken into account. Consequently, molecular-turbulent transfer effects are not considered.

The results of our test calculations are shown in Figures 4 and 5. The calculated least-squares fits match the observed values very well. In the first case (21 April 1974), an upward flux (F_c = 0.088 $\mu g\ m^{-2}\ s^{-1}$) was calculated. In the second case (22 April 1974), the estimated deposition flux was F_c = -0.080 $\mu g\ m^{-2}\ s^{-1}$, where the deposition velocity at the height z = 1 m was 6.4 cm s^{-1}.

Upward particle fluxes were also observed by Wesely and Hicks (1979) and were determined by Garland and Cox (1982) and Nicholson and Davies (1987). Since during the GREIV I experiment the contamination of vegetation and the soil by NH_4^+-particles was not investigated, and since no measurements of NH_4^+-concentrations near the surface were carried out, the upward flux of NH_4^+-particles should not be considered as an example of resuspension. Note, that fluxes of NH_3 determined for the same measuring periods were of opposite directions. It may be suspected that these opposite flux directions were caused by chemical transformations, e.g., via

$$NH_3 + HNO_3 \leftrightarrow NH_4NO_3 \quad . \tag{35}$$

However, if such a transition is taken into consideration, then it is necessary to check whether the constant flux assumption is appropriate or not, because Brost *et al.* (1988) suggested that the transition between nitric acid and ammonium nitrate may produce slightly height-dependent fluxes when the lifetime of the dissociation equilibrium is less than or equal to 100 s. Since no concentrations of HNO_3 were measured during the GREIV I experiment, such a check could not be carried out.

Model estimates of the vertical transfer of aerosol particles in the atmospheric surface layer leads to qualitatively and quantitatively reasonable results. However, the verification of the overall physics of the model is still necessary. This would presuppose that directly measured particle fluxes can be used for the purpose of comparison. There is an urgent need for future field experiments to determine directly the vertical flux of particles, in addition to the vertical profiles of wind speed, dry- and wet-bulb temperatures, particle concentrations and size distributions.

ACKNOWLEDGEMENT

We should like to express our thanks to the Deutsche Forschungsgemeinschaft (SFB 73 "Atmosphärische Spurenstoffe" - Teilprojekt F9) and the Fraunhofer-Gesellschaft zur Förderung der angewandten Wissenschaften e.V. for their support of this study.

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DISCUSSION

V.M. VOLOSHCHUK. Why do you add the coefficient of Brownian diffusion of particles, D_c, to the coefficient of turbulent diffusion, K_c, if $D_c/K_c \approx 10^{-10}$?

G.KRAMM. The Brownian diffusion coefficient, of course, plays no role in the turbulent region of the atmospheric surface layer because K_c is much larger than D_c. However, in the viscous sublayer (the layer in the immediate vicinity of any receptor surface), which controls and limits the diffusion processes, D_c is of same order or larger than the decreasing K_c. Therefore, it is necessary to include D_c in Eq. (1) to describe the transition from turbulent to Brownian diffusion (see also Eq. (34)). Note, that Brutsaert's results on diffusive transfer to and from smooth and rough surfaces mentioned above (see Eqs. (33) and (34)) are in close agreement with experimental results reported by Chamberlain A.C., Garland J.A., and Wells A.C., 1984, Transport of gases and particles to surfaces with widely spaced roughness elements, *Boundary-Layer Meteorology* **29**, 343-360.

S.E. SCHWARTZ. It would be useful (to my thinking) to present a size-dependent deposition velocity (graph) and an overall deposition velocity = mass flux/mass concentration for the two cases you present, or do you consider this anathema?

G.KRAMM. We do not. However, the size distribution of the NH_4^+-particles was not determined during the GREIV I-experiment, but the profile data of particle concentration, wind speed, dry- and wet-bulb temperatures used here were the best data sets available for our model tests (we hope to receive additional data sets in the future). Therefore, we could not calculate any terminal settling velocity (neither in a size-dependent, nor in an overall form). Sedimentation effects were neglected, because our estimates have shown that the contributions of water-soluble particles (mode 1) to the terminal settling velocity are small (since relative humidity was less than 80 %, effects of water vapour adsorption, which may produce an increase of particle size, are negligible; see also G. Hänel, 1982, Influence of relative humidity on aerosol deposition by sedimentation, *Atmos. Environ.* **16**, 2703-2706).