Appendix A – Aerosol deposition model

Introduction

The aerosol deposition model presented in chapter four is an implementation of the Slinn framework (Slinn, 1980) presented by Zhang *et. al.* (2002). In the interest of ease of repeatability, the major equations are reproduced here along with further definitions not presented in the above paper, but required in order to write the model. This is not intended as a review or critique of deposition models, but a record of the model used in chapter 4. Further details are available in the paper, or in other references used here. The general form of the model is presented first, followed by the required equations.

Model equations

As explained in chapters two and four, total deposition velocity is the sum of gravitational and turbulent deposition velocities. Also, the turbulent deposition velocity is controlled by the aerodynamic resistance and the surface resistance to uptake. The total is represented as:

$$v_d = v_g + 1 / r_a + r_s \tag{A1}$$

The aerodynamic resistance has been approximated by a number of researchers. Here, either Monteith and Unsworth's (1990) definition can be used, where:

$$r_a = U / u_*^2 \tag{A2}$$

or Zhang *et. al.* (2002) suggest:

$$r_a = \left(\ln\left(z / z_0 \right) - \psi_h \right) / k \cdot u_* \tag{A3}$$

where the only previously undefined quantity is the height integrated stability correction for heat (ψ_h; mentioned in chapter two) which is given by:

$$\psi_h = -5\left(z - d/L\right) \qquad \zeta < 0 \qquad \text{(A4a)}$$

$$\psi_h = 0 \qquad \zeta = 0 \qquad \text{(A4b)}$$

$$\psi_h = 2\ln\left(1/\Phi_h + 1\right) \qquad \zeta > 0 \qquad \text{(A4c)}$$

These are the stability corrections for heat and pollutant transport. For reference, those for momentum are also presented, although they are not used in this model.

$$\psi_m = -5\left(z - d/L\right) \qquad \zeta < 0 \qquad \text{(A5a)}$$

$$\psi_m = 0 \qquad \zeta = 0 \qquad \text{(A5b)}$$

$$\psi_m = 2\ln\left(1/\Phi_m + 1\right) + \ln\left(1/\Phi_m^2 + 1\right) - 2\tan^{-1}\left(1/\Phi_m\right) \qquad \zeta > 0 \qquad \text{(A5c)}$$

v_g (in equation A1) is the gravitational settling velocity, given by Stokes' law as follows.

$$v_g = \rho_p \cdot D_p^2 \cdot g \cdot C_c / 18 \cdot \eta \qquad \text{(A6)}$$

where ρ_p is particle density, g is the acceleration due to gravity (9.81 m s^{-1}), η is the viscosity coefficient of air (1.71×10^{-5} m^2 s^{-1}) and C_c is the Cunningham slip correction, – a correction for the small non-zero component of perpendicular flow at the surface of small particles given by:

$$C_c = 1 + 2\lambda \cdot \left(1.257 + 0.4e^{-0.55 D_p / \lambda} \right) \quad \text{(A7)}$$

where λ is the mean free path of air molecules (1×10^{-7} m). The final term in equation A1 is the surface resistance. This is the uncertain term, which gives rise to the variability in the predictions of different models, noted in chapter four. r_s is defined as:

$$r_s = 1 / \varepsilon_0 \cdot u_* \cdot (E_B + E_{IM} + E_{IN}) \cdot R_s \quad \text{(A8)}$$

here ε_0 is an empirical constant (set to 3 in the implementation used) and R_s is the size dependent sticking probability for aerosol (the probability that a particle will not bounce off the surface and be re-suspended). E_B, E_{IM} and E_{IN} are, respectively, the collection efficiencies of Brownian diffusion, impaction and interception. There are several formulations of each of these parameters, and it is from these that the variability in r_s stems. R_s is defined (Slinn, 1982) as:

$$R_s = e^{-St^{1/2}} \quad \text{(A9)}$$

The efficiency of particle interception in this model is treated as a function of particle diameter and the radius of collectors in this model:

$$E_{IN} = 1/2 \cdot \left(D_p / A \right)^2 \quad \text{(A10)}$$

with A being the radius of collectors. A value of 2 mm was used, to simulate blades of grass as the main collectors of aerosol. E_B is a similarly empirical function, given by:

$$E_B = Sc^{-\gamma} \quad \text{(A11)}$$

where γ is another empirical constant, in this case a value of 0.66 was used, following the recommendation of Slinn (1982) for vegetated surfaces. Sc is the Schmidt number,

the ratio of the kinematic viscosity of air (1.8×10^{-5} N m^2 s^{-1}) to the particle Brownian diffusivity. Brownian diffusivity is defined as:

$$D = {K \cdot T \cdot C_c}/{3\pi \cdot \eta \cdot D_p} \tag{A12}$$

where K is the Boltzmann constant (1.38×10^{-23} J K^{-1}), all other quantities as previously defined. Of the collection efficiencies, this leaves only E_{IM} undefined. Many approximations have been suggested for E_{IM}. Zhang et. al. (2002) present six of these, all functions of the Stokes number. Before presenting the E_{IM} equation, the Stokes number is defined, for vegetated surfaces (Slinn, 1982) as:

$$St = {v_g \cdot u_*}/{g \cdot A} \tag{A13}$$

There is an alternative approximation for Stokes number over smooth surfaces (Giorgi, 1988), but the Slinn form was used in this case as it was designed for use with vegetated surfaces. Of the available forms for E_{IM}, that presented by Giorgi (1988) was used, because it was derived for vegetated surfaces. It is similar to that reported by Peters and Eiden (1992) for forest, with slightly lower mean values, and significantly lower values for small particles.

$$E_{IM} = \left({St}/{0.6 + St}\right)^{3.2} \tag{A14}$$

Summary

The equations presented in this appendix are sufficient to recreate the model used for comparison with measurements in chapter four of this thesis. The model was written in Visual Basic (in a Microsoft Excel spreadsheet), which is available on request from the author (currently j.dorsey@umist.ac.uk).

This is *one possible* implementation of the Slinn type deposition model framework. For more detail on this type of model and the advantages / disadvantages of various

parameterisations of different terms, the reader is referred to Slinn (1980), Slinn (1982), Giorgi (1986), Zhang *et. al.* (2002) and especially to Ruijgrok *et. al.* (1997).

Appendix B – A note on notation

The Reynolds decomposition of time series outlined in chapter two is used widely throughout this thesis. It is not always convenient or desirable to use the full (over bar, prime) notation in every equation, and these have been omitted in certain cases, in the interest of removing unnecessary "clutter", where it is clear which component should be used. In almost all cases the quantity has instead been capitalised, and the averaged component of the decomposition should be used. As a rule of thumb, if the equation in question is unrelated to eddy covariance calculation or time series analysis, the averaged quantity should be used. An exception to the capitalisation "rule" above is that time series associated with power and co-spectral analyses are denoted using capital subscripts "W", and "T".

Because of the rather large number of variables involved in a study of this type, several letters and symbols are occasionally re-used in different sections of the thesis to denote different quantities. Where this done, the text accompanying the equations always clarifies what variable the symbols refer to.

Finally, although units are listed with variables in appendix C, different units are used at different times for many variables. An example is the vapour mass flux, j_v. In calculations j_v must be treated in MKS, or at least in units consistent with the rest of the calculation. However when tabulated or plotted, j_v appears in μg m^{-3} hr^{-1} because this is a more suitable unit for its magnitude in general. Similarly D_p is always presented in either μm or nm, but is always in m for calculation purposes. Wherever there could be any doubt, the required units are listed in the accompanying text with equations. However, care and common sense are required, as some equations taken from outside sources have mismatched units (*e.g.* equation 2.21, see k_w).

Appendix C – List of symbols

#	–	Dimensionless
*	–	Variable units

Roman alphabet

A	–	Empirical constant (chapter six)	–	#
A	–	Aerosol collector radius (chapter four)	–	m
b	–	Empirical constant (chapter six)	–	#
C	–	General constant	–	*
C_{AB}	–	Co-spectrum of time series A and B	–	*
C_c	–	Cunningham slip correction	–	#
c_d	–	Surface drag coefficient	–	#
c_p	–	Specific heat capacity of dry air at constant pressure (= 1004.67)	–	J kg^{-1} K^{-1}
c_v	–	Specific heat capacity of dry air at constant volume (= 717.02)	–	J kg^{-1} K^{-1}
d	–	Zero plane displacement	–	m
D	–	Gas diffusion coefficient	–	s^{-1}
D_3	–	Aerosol in range 11 nm < D_p < 3 µm, typically Representative of 11 nm < D_p < 100 nm.	–	Range
D_p	–	Particle diameter	–	µm *or* nm
E	–	Water vapour loading	–	kg m^{-3}
E_B	–	Brownian diffusion efficiency	–	#
E_f	–	Total anthropogenis energy input	–	W m^{-2}
E_{ff}	–	Fossil fuel derived anthropogenic energy input	–	W m^{-2}
E_{IM}	–	Impaction efficiency	–	#
E_{IN}	–	Interception efficiency	–	#
E_{nff}	–	Non-fossil fuel anthropogenic energy input	–	W m^{-2}
f	–	Nondimensional frequency	–	#
f_{min}	–	Minimum required logging frequency	–	Hz
f_{mA}	–	Peak in power spectrum S_A	–	*

f_{Ny}	–	Nyquist frequency	–	Hz
f_{χ}	–	Pollutant (aerosol) flux	–	$cm^{-2}\ s^{-1}$
g	–	Acceleration due to gravity (= 9.81)	–	$m\ s^{-2}$
H	–	Sensible heat flux	–	$W\ m^{-2}$
h_c	–	Canopy height	–	m
$[i]$	–	Concentration of species "i"	–	$\mu g\ m^{-3}$
j_v	–	Vapour mass flux	–	$\mu g\ m^{-3}\ hr^{-1}$
k	–	von Karman constant (= 0.4)	–	#
K	–	Bolzmann constant (= 1.38×10^{-23})	–	$J\ K^{-1}$
k_e	–	Theoretical dissociation constant for NH_4NO_3	–	hPa^2
K_E	–	Eddy diffusivity for water vapour	–	s^{-1}
K_H	–	Eddy diffusivity for heat	–	s^{-1}
k_m	–	Measured vapour concentration product	–	hPa^2
K_M	–	Eddy diffusivity for momentum	–	s^{-1}
Kn	–	Knudsen number	–	#
$k_{species}$	–	Rate coeff. for second order reaction of "species"	–	#
k_w	–	KH2O extinction coefficient	–	$mV\ m^3\ g^{-1}\ cm^{-1}$
K_{χ}	–	Eddy diffusivity for entrained pollutants	–	s^{-1}
l	–	Mixing length	–	m
L	–	Obukhov length	–	m
L_v	–	Latent heat of vaporisation for water	–	$J\ kg^{-1}$
m_v	–	Molecular mass of condensing vapour	–	AMU
n	–	Natural frequency	–	Hz
N_{air}	–	Molarity of air	–	m^{-3}
nh	–	Highest frequency in power spectrum (chapter four)	–	Hz
N_i	–	Molarity of species i	–	m^{-3}
n_l	–	Lowest frequency in power spectrum (chapter four)	–	Hz
n_r	–	Highest properly resolved frequency in power spectrum (chapter four)	–	Hz
n_r	–	Radial frequency	–	$rad\ s^{-1}$

N	–	Avogadro constant (chapter five) (= 6×10^{23})	–	#
N	–	Number of samples in time series	–	#
N_{min}	–	Minimum aerosol required for significant flux	–	#
N_{tot}	–	Total number of aerosol observed	–	#
p	–	Pressure	–	hPa
p_0	–	Reference pressure (normally 1013.25)	–	hPa
$p[i]$	–	Partial pressure of species "*i*"	–	hPa
q	–	Water vapour loading (KH2O calculations)	–	kg m^{-3}
Q	–	Measured (aerosol) flux (Schuepp model)	–	m^{-2} s^{-1}
Q_0	–	(Aerosol) Flux normalisation (Schuepp model)	–	m^{-2} s^{-1}
r	–	Particle radius	–	m
r_a	–	Aerodynamic resistance	–	s m^{-1}
r_b	–	Laminar boundary layer resistance	–	s m^{-1}
r_c	–	Total canopy resistance	–	s m^{-1}
r_s	–	Canopy surface resistance to deposition	–	s m^{-1}
r_f	–	Sample flow rate	–	cm^3 s^{-1}
r_g	–	Ground surface resistance	–	s m^{-1}
R_g	–	Diameter dependent aerosol growth rate ratio	–	#
R_i	–	Gradient Richardson number	–	#
r_L	–	Water vapour mass mixing ratio	–	#
R_{Lu}	–	Upward long wave radiation	–	W m^{-2}
R_{Ld}	–	Downward long wave radiation	–	W m^{-2}
R_n	–	Net radiation	–	W m^{-2}
r_p	–	Resistance to chemical uptake at surface	–	s m^{-1}
R_s	–	Particle sticking efficiency	–	#
r_{sat}	–	Saturation water vapour mass mixing ratio	–	#
r_t	–	Total resistance to deposition	–	s m^{-1}
r_{xy}	–	Covariance of time series *x* and *y*	–	*
S_A	–	Power spectrum of time series *A*	–	*
Sc	–	Schmidt number	–	#
St	–	Stokes number	–	#

S_t	–	Global short wave radiation	–	W m^{-2}
T_*	–	Surface layer scaling temperature	–	K
T	–	(Air) Temperature	–	K
T_a	–	Air temperature	–	K
T_e	–	Arbitrary turbulence time scale	–	s
T_s	–	Surface temperature	–	K
u_*	–	Friction velocity	–	m s^{-1}
u	–	Instantaneous horizontal wind velocity	–	m s^{-1}
$\bar{u}$	–	Mean horizontal wind velocity	–	m s^{-1}
u'	–	Perturbation in horizontal wind velocity	–	ms^{-1}
u_0	–	Rotated horizontal wind velocity	–	m s^{-1}
v	–	Instantaneous transverse wind velocity	–	m s^{-1}
$\bar{v}$	–	Mean transverse wind velocity	–	m s^{-1}
v'	–	Perturbation in transverse wind velocity	–	ms^{-1}
V	–	Voltage	–	V
v_0	–	Rotated transverse wind velocity	–	m s^{-1}
v_d	–	Deposition velocity	–	mm s^{-1}
v_{dm}	–	Measured deposition velocity	–	mm s^{-1}
v_e	–	Emission velocity	–	mm s^{-1}
v_g	–	Gravitational settling velocity	–	mm s^{-1}
V_q	–	KH2O signal voltage	–	V
w	–	Instantaneous vertical wind velocity	–	m s^{-1}
$\bar{w}$	–	Mean vertical wind velocity	–	m s^{-1}
w'	–	Perturbation in vertical wind velocity	–	ms^{-1}
w_0	–	Rotated vertical wind velocity	–	ms^{-1}
x	–	KH20 optical path length	–	cm
x	–	Upwind distance (Schuepp model)	–	m
z	–	Height	–	m
z_0	–	Roughness length	–	m
z_0'	–	Sink level for ammonia vapour	–	m
z_m	–	Measurement height	–	m

Greek alphabet

α	–	Horizontal wind angle	–	Deg
α	–	Vapour sticking coefficient	–	#
β_m	–	Transition regime vapour flux correction	–	#
χ	–	(Aerosol) Pollutant concentration	–	cm^{-3}
χ_c	–	Coincidence corrected aerosol concentration	–	cm^{-3}
χ_v	–	Condensable vapour concentration	–	$\mu g\ m^{-3}$
χ_{veff}	–	Effective condensable vapour concentration	–	$\mu g\ m^{-3}$
Δ	–	Proportional error	–	*
ε_0	–	Empirical constant (Zhang model) (= 3)	–	#
Φ_h	–	Empirical stability correction for heat	–	#
Φ_m	–	Empirical stability correction for momentum	–	#
Φ_χ	–	Empirical stability correction for pollutants	–	#
ϕ_h	–	Dissipation rate of temperature variance	–	#
ϕ_ε	–	Dissipation rate of turbulent kinetic energy	–	#
γ	–	c_p / c_v	–	#
η	–	Viscosity coefficient of air (= 1.71×10^{-5})	–	$m^2\ s^{-1}$
λ	–	Latent heat of vaporisation for water	–	$J\ kg^{-1}$
λ	–	Arbitrary turbulence length scale (chapter two)	–	m
λ	–	Mean free path of condensable vapour (= 1×10^{-7})	–	m
λE	–	Latent heat flux	–	$W\ m^{-2}$
θ	–	Wind azimuth (vertical) angle	–	Deg
θ	–	Potential temperature	–	K
θ_v	–	Virtual potential temperature	–	K
ρ	–	(Air) Density	–	$kg\ m^{-3}$
ρ_a	–	Density of dry air (= 1.225)	–	kg m-3

ρ_p	–	Particle density	–	kg m^{-3}
σ_A	–	Standard deviation of time series "*A*"	–	*
τ	–	Reynolds stress	–	kg m^{-1} s^{-2}
τ	–	Aerosol inlet lag time	–	s
τ_m	–	Time period of measurement	–	s
ψ_h	–	Height integrated stability correction for heat	–	#
ψ_m	–	Height integrated stability correction for momentum	–	#
ζ	–	Boundary layer stability parameter	–	#

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