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Modeling dry deposition of S and N compounds to vegetation

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This paper deals with parameterization method based on meteorological parameters for calculation of dry deposition S and N compounds on natural surfaces (leaf of *Cassia siamea*) as direct measurement methods are cumbersome. A theoretical method for calculation of dry deposition of S and N compounds based on meteorological parameters has been outlined by which all the resistances responsible for deposition of gases and particles on vegetation could be determined. Since, numerous steps are involved in calculation; a computer program has been developed to make it fast, convenient and more useful. The deposition velocity of SO_2, HNO_3, SO_4^{2-} and NO_3^- obtained by current parameterization method on vegetation (leaf of *Cassia siamea*) at Dayalbagh, Agra in a semi-arid region of India is 0.32, 0.74, 1.16 and 1.07 cm s^{-1}. The obtained deposition velocities are in the reported range on vegetation.

Key words: Dry deposition, Parameterization, Sulphur, Nitrogen, Vegetation
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1 Introduction

Deposition of S and N compounds have attracted attention of scientific community worldwide due to their detrimental effects, including wide range of acidification of soil, ponds, lakes, corrosion of building materials, injury to vegetation and skin, etc. But, there is no widely accepted technique to measure the dry deposition. A broad range of techniques has been used for estimation of dry deposition[1], which has been categorized as direct and indirect methods. Indirect method includes gradient method and inferential techniques. In direct method, an explicit determination is made of the flux of material to the surface. It includes surrogate surface, natural surface, chamber method, micrometeorological viz., eddy correlation, eddy accumulation methods, surface analysis method, isotopic tracer, etc. Direct methods require considerably more effort and relatively sophisticated instrumentation.

Several efforts have been made to calculate the dry deposition theoretically by two distinct types of models: meteorological models and surface mass transfer models. Many meteorological models, which can be used to calculate the downwind transport and diffusion of gases and particles include or could be modified to include the dry deposition velocity boundary conditions. Some of these modified models include Sutton's equations[2], Pasquill's mode[3], Pasquill's equations[4], a Gausian plume model using surface depletion[5], Gaussian plume model by others[6-9], Slinn's theoretical model[10,11] and finite difference model[12]. Other models are based on surface mass transfer by dry deposition. These include Multibox resistance model[11,13-14], resistance model[15,16], four-canopy model[17], multibox k-theory model[18], long-range transport/deposition model[19,20] and multi-level Lagrangian atmospheric model[21].

Many dry deposition models have been developed during past ten years and efforts continue to improve their capabilities. The dry deposition module in the Acid Deposition and Oxidant Model (ADOM)[22], Regional Acid Deposition Model (RADM)[23-26] and Dutch Empirical Acid Deposition Model (DEADM)[27] have undergone testing and revisions and have appeared in several applications[28]. These models make extensive use of the resistance analogy in one form or another for deposition calculations. Newer models are expected to have improved capabilities that can reduce the dependency on empirically derived resistance values and provide means of coupling deposition and emission more closely[29]. These models provide estimates of soil moisture content and evapotranspiration, which can be a valuable input to dry deposition module[28].

Dry deposition can also be determined by parameterization method by calculating the three resistances (R_a, R_b, and R_c) governing dry deposition. Development of the parameterization and modeling of dry deposition rates of particle has been slowly advancing[17, 25, 30-33]. All these second generation models have been informative, but a comprehensive understanding of particle deposition has not been achieved. Models obtained using three resistances are second generation models. In the entire earlier reported second generation model, they have taken some parameters from the earlier reported studies but no one has calculated all the parameters. Recently third generation models have been given, but they are yet to be verified experimentally[28]. Innovative methods of measuring particle deposition need to be developed and applied to derive more universal parameterizations of depositions in natural settling outdoors. Hence, the present study was planned to determine aerodynamic resistance (R_a), quasi-laminar resistance (R_b) and surface resistance (R_c) using meteorological data by parameterization method.

In the present study all the three resistances have been calculated. So, this method is more realistic. As numerous parameters are involved in calculation of dry deposition velocity by the present method, a computer program has been developed to make this method fast, convenient and more useful. So, the present method is an improved second generation model.

2 Theoretical calculation

Temperature, wind direction, wind speed and solar radiation play a significant role in dry deposition processes[30]. These meteorological parameters were monitored using a self-contained battery operated WDL 1002 Data logger (Dynalab, Pune) system. The data logger acquires data from the sensors for wind speed, wind direction, air temperature and solar radiation, stores the data in its memory for later retrieval and prints the data. Dynalab-Data logger was stationed on the roof of the Science Faculty building of the Institute at Dayalbagh. Dayalbagh is located in Agra (North Central India, 27°10′N, 78°05′E), which is about 200 km south-east of Delhi. Agra is about 169 m above the mean sea level (msl), has semi-arid climate with atmospheric temperature ranging from 11-48°C (max) to 0.7-30°C (min), relative humidity 25-95%, light intensity 0.7-5.6 oktas (cloudiness) and annual rainfall 650 mm. Table 1 presents the sensors used along with resolution and accuracy of meteorological parameters. The data were collected for two years (July 1999 to June 2001). Annual and seasonal arithmetic mean, standard deviation, minimum and maximum values of meteorological parameters are presented in Table 2.

In the present study, dry deposition velocity was calculated by simulating the different processes that govern the dry deposition. The deposition velocity (V_d) for gases[34] is presented by Eq. (1), as

$$V_d = \frac{1}{R_a + R_b + R_c} \qquad \ldots (1)$$

and that for particles[34] by

$$V_d = \frac{1}{R_a + R_b + R_a R_b V_s} + V_s \qquad \ldots (2)$$

where, R_a = aerodynamic resistance, R_b = quasi-laminar resistance, R_c = surface resistance and V_s = settling velocity.

2.1 Calculation of aerodynamic resistance (R_a)

The aerodynamic resistance (R_a) for both gases and particles[30] in neutral and stable conditions is calculated by

$$R_a = 4(u\,\sigma_\theta^2)^{-1} \qquad \ldots (3)$$

and in unstable conditions by

$$R_a = 9(u\,\sigma_\theta^2)^{-1} \qquad \ldots (4)$$

where, u = mean wind speed and σ_θ = standard deviation of wind direction.

As the atmospheric conditions are stable in winter and unstable in summer and monsoon, the aerodynamic resistance for winter season was calculated by substituting the values of wind speed

Table 1 — Sensors used, resolution and accuracy of meteorological parameters

Measurement Parameters	Sensors	Resolution	Accuracy	Units
Wind speed	3 Cup Anemometer	-	± 2 %	m/s
Wind direction	Wind vane	1°	± 3°	deg
Relative humidity	Solid state capacitive type	0.1%	3% of full scale reading	% of full scale
Solar radiation	72 element thermopile	-	-	W m^{-2}
Ambient temperature	Platinum resistance	0.1 ºC	0.2ºC	ºC

Table 2 — Temperature, relative humidity, solar radiation, wind speed and wind direction of the study period

Statistics	Temperature, °C	Relative humidity, %	Solar Radiation, W m^{-2}	Wind Speed, m s^{-1}	Wind Direction
Winter					
Arithmetic mean	16.08	70.95	573.13	1.66	W, NW, NE
Standard deviation	3.24	9.88	154.29	0.33	45°
Minimum	11.1	52.3	365.0	1.14	
Maximum	23.0	87.8	732.5	2.83	
Summer					
Arithmetic mean	33.04	51.28	982.08	1.87	W, NW, SW
Standard deviation	3.10	16.31	238.86	0.68	45°
Minimum	25.1	24.1	620.0	0.91	
Maximum	39.2	83.0	1330.0	2.56	
Monsoon					
Arithmetic mean	30.43	71.49	847.94	1.96	W, SW, NW, NE
Standard deviation	1.66	12.20	250.54	0.53	67.5°
Minimum	28.1	45.19	370.0	1.11	
Maximum	34.25	86.3	1128.25	2.92	
Annual					
Arithmetic mean	25.79	63.98	872.73	1.88	NW, W
Standard deviation	8.36	16.09	266.7	0.54	67.5°
Minimum	11.1	24.10	365.0	0.91	
Maximum	39.2	87.8	1330.0	2.92	

Table 3—Aerodynamic resistance (R_a), quasi-laminar resistance (R_b), combined stomatal and mesophyll resistance (R_{sm}), cuticular resistance (R_c), foliar resistance (R_{cf}), settling velocity (V_s) and deposition velocity (V_d) for gaseous SO_2, HNO_3, and particulate SO_4^{2-} and NO_3^- as obtained by parameterization method for Cassia leaf

Species	Seasons	R_a	R_b	Combined R_{st} and R_m	R_{cut}*	R_{cf}	V_s	V_d
SO_2	S	0.078	0.96	2.66	100	2.6	NR	0.28
	M	0.033	0.96	5.11	100	1.86	NR	0.35
	W	0.039	0.96	8.54	100	2.99	NR	0.25
	A	0.058	0.96	5.81	100	2.1	NR	0.32
SO_4^{2-}	S	0.078	1.36	NR	NR	NR	0.38	1.14
	M	0.033	1.36	NR	NR	NR	0.38	1.18
	W	0.039	1.36	NR	NR	NR	0.38	1.03
	A	0.058	1.36	NR	NR	NR	0.38	1.16
HNO_3	S	0.078	0.96	2.60	0	0.276	NR	0.76
	M	0.033	0.96	5.03	0	0.318	NR	0.75
	W	0.039	0.96	8.42	0	0.341	NR	0.74
	A	0.058	0.96	5.72	0	0.325	NR	0.74
NO_3^-	S	0.078	1.36	NR	NR	NR	0.47	1.06
	M	0.033	1.36	NR	NR	NR	0.47	1.09
	W	0.039	1.36	NR	NR	NR	0.47	1.08
	A	0.058	1.36	NR	NR	NR	0.47	1.07

Note: NR = not required. *Source*: *Seinfeld and Pandis, 1998[35]

and deviation in wind direction in Eq. (3) and for summer and monsoon seasons by substituting these parameters in Eq. (4). The mean of the values obtained for aerodynamic resistance (R_a) is presented in Table 3.

2.2 Calculation of quasi-laminar resistance (R_b)

The quasi-laminar resistance (R_b) for gases[35] is calculated by

$$R_b = 5(\mathrm{Sc})^{2/3}/u_* \qquad \ldots (5)$$

and for particles[35] by

$$R_b = 1/u_*(\mathrm{Sc}^{-2/3} + 10^{-3/\mathrm{St}}) \qquad \ldots (6)$$

where, u_* = friction velocity (root mean covariance between horizontal and vertical velocity components)[34], Sc = Schmidt number of species and St = stokes number of species.

Schmidt number of species is represented[34] by

$$\mathrm{Sc} = \nu/D \qquad \ldots (7)$$

where, ν = viscosity of air (at 20°C, 0.15 cm^2 s^{-1} at sea level) and D = molecular (for gas) and Brownian (for particles) diffusivities. The values of diffusivities (D) (cm s^{-1}) and the values of Schmidt number (Sc) for various species are presented in Table 4.

Table 4 — Molecular (for gases) and Brownian (for particles) diffusivities (D; cm^2/s) for a range of pollutants and deduced values of Schmidt number (Sc).

	D	Sc
	Gaseous species	
Nitric acid	0.12	1.25
Sulphur dioxide	0.12	1.25
	Particles (unit density)	
10^{-3}	1.28×10^{-2}	1.17×10^{1}
10^{-2}	1.35×10^{-4}	1.11×10^{3}
10^{-1}	2.21×10^{-6}	6.79×10^{4}
1	1.27×10^{-7}	1.18×10^{6}
10	1.38×10^{-8}	1.09×10^{7}

Source: *Hicks et al.*[34] (NOAA Technical Memorandum), 1986

Stokes number is calculated[35] by the relation

$$\mathrm{St} = V_s u_*^2 / g\nu \qquad \ldots(8)$$

where, V_s = settling velocity for particles, u_* = friction velocity, g = gravitational acceleration and ν = viscosity of air.

Settling velocity is calculated[35] by

$$V_s = \rho_p D_p^2 g C_c / 18\nu \qquad \ldots (9)$$

where, ρ_p = density of the particle (assumed to be unity for particles), D_p = particle diameter, g = gravitational acceleration (9.8 m s^{-2} at sea level), C_c = slip correction factor (Given in Table 5) and ν = viscosity of air

The settling velocity (V_s) of particles is determined by substituting the values of particles diameter, and other factors (ρ_p, g, C_c and μ) and values obtained are presented in Table 3. The quasi-laminar resistance (R_b) of gaseous SO_2 and HNO_3 are calculated by substituting the values of respective Schmidt number and friction velocity and for particles by substituting the values of Schmidt number, friction velocity and Stokes number in Eqs (5) and (6), respectively. The mean of the values of quasi-laminar resistance (R_b) is given in Table 3.

2.3 Calculation of surface resistance (R_c)

The surface resistance (R_c) is assumed to be zero for particles as it mainly depends[35] on settling velocity (V_s). Thus, R_c for foliage is calculated for gases only. Value of R_c depends on cuticular resistance (R_{cut}), stomatal resistance (R_{st}), mesophyll resistance (R_m) and leaf area index (LAI) and is expressed by the relation[35]

$$R_c = (1/R_{cut} + 1/R_{st}+R_m)^{-1}\,(\mathrm{LAI})^{-1} \qquad \ldots (10)$$

Table 5 — Slip correction factor C_c for spherical particles in air at 298 K and 1 atm

D_p, µm	C_c
0.001	216
0.002	108
0.005	43.6
0.01	22.2
0.02	11.4
0.05	4.95
0.1	2.85
0.2	1.865
0.5	1.326
1.0	1.164
2.0	1.082
5.0	1.032
10.0	1.016
20.0	1.008
50.0	1.003
100.0	1.0016

Source: Seinfeld and Pandis, 1998[35]

where, R_c is the foliar resistance, R_{cut} the cuticular resistance, R_{st} the stomatal resistance, R_m the mesophyll resistance and LAI the leaf area index.

Hence, to calculate the surface resistance for foliar (R_c), the above mentioned parameters were calculated and leaf area index (LAI) was determined. Stomatal resistance (R_{st}) depends on solar radiation (G in Wm^{-2}) and surface air temperature (T_s in °C) (between 0 and 40 °C). The following relation[35] is used to calculate R_{st}

$$R_{st} = rj\left[1 + (200/G + 0.1)^2 \left(\frac{400}{T_s(40 - T_s)}\right)\right] \qquad \ldots (11)$$

where, rj is input resistance (s m^{-1}) and its value in case of coniferous forest[25] was assumed to be 130, 250 and 400 s m^{-1} for summer, monsoon and winter season, respectively. As selected tree (Cassia) is also a coniferous tree, the same values of rj have been used for calculating R_{st}. The stomatal resistance was calculated by substituting the values of rj and solar radiation and temperature (Table 2) in Eq. (11). The mesophyll resistance (R_m) depends on the solubility of gases[36]. The combined stomatal and mesophyll resistance is calculated from the following relation given by Seinfeld and Pandis[35]

$$\begin{aligned} R_{sm} &= R_{st} + R_m \\ &= R_{st}\,\frac{D_{H_2O}}{D_i} + \frac{1}{3.3\times10^{-4} H_i^* + 100 f_0^i} \qquad \ldots (12) \end{aligned}$$

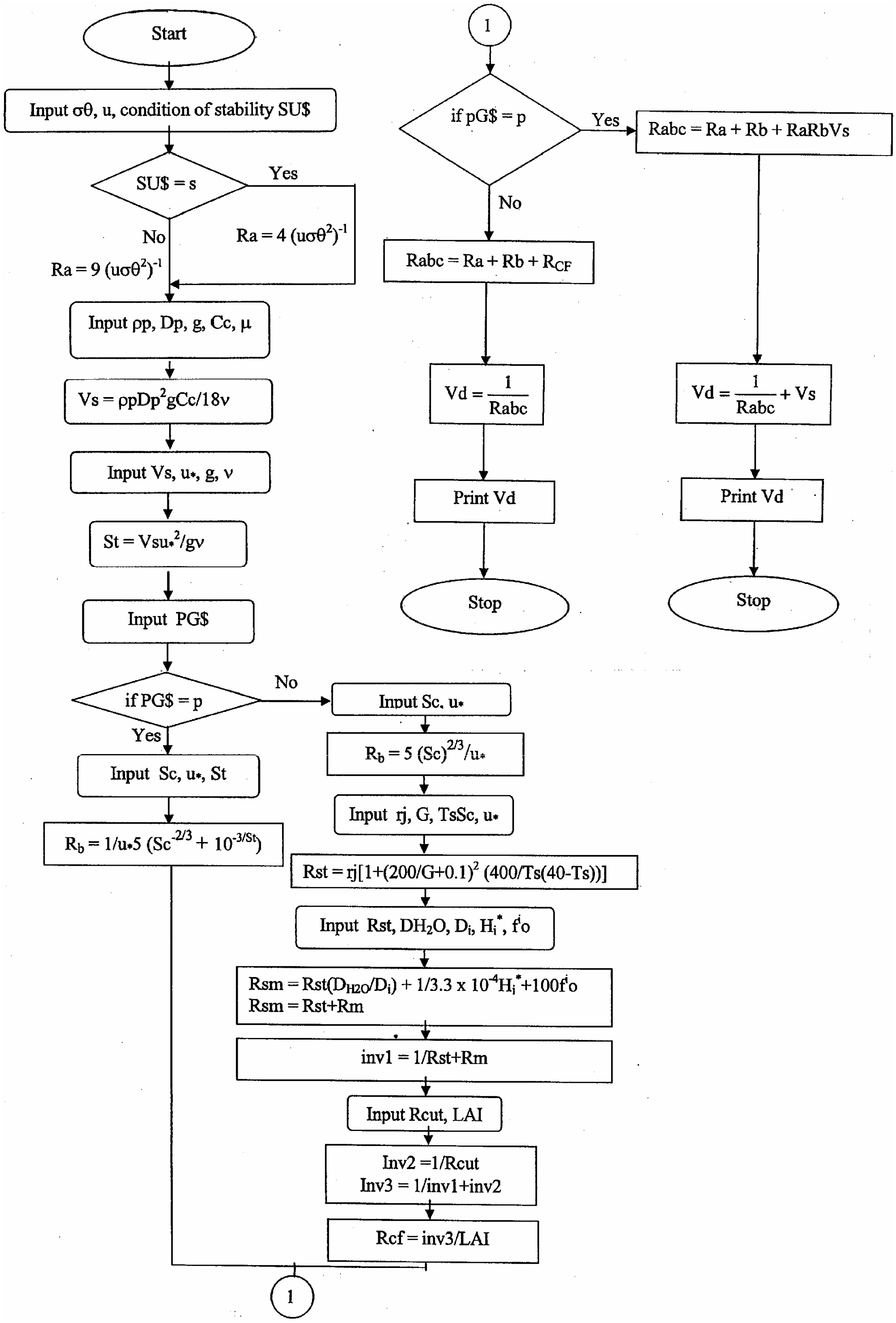

Fig. 1 — Algorithm of parameterization method to compute dry deposition

where, D_{H_2O}/D_i is the ratio of molecular diffusivity of water to that of the specific gas (SO_2 = 1.89; HNO_3 = 1.87), H_i* = Hennry' law constant (M atm^{-1}) ($SO_2 = 1\times10^5$; $HNO_3 = 1 \times10^{14}$), f_0^i = normalized reactivity factor (0 to 1) (SO_2 = 0; HNO_3 = 0). The calculated values of combined stomatal and mesophyll resistance is presented in Table 3.

Transfer of gases through the cuticle is generally less important than through the stomata and can be neglected[35]. Stomatal resistance decreases as relative humidity increases. In general, R_{cut} for HNO_3 has been considered[35] to be zero while for SO_2 to be 100 s cm^{-1}.

The leaf area index (LAI) is calculated[34] by the relation:

LAI = the total area of foliage/area of the earth's surface covered by canopy

The calculated LAI of Cassia leaf is 2.62. By putting the values of cuticular resistance (R_{cut}), combined stomatal and mesophyll resistance (R_{sm}) and leaf area index (LAI) in Eq. (10), surface resistance (R_c) is obtained and is presented in Table 5. The deposition velocities (Table 3) were obtained by substituting values of R_a, R_b, and R_c, for gaseous SO_2 and HNO_3 in Eq. (1) and deposition velocities of particulate SO_4^{2-} and NO_3^- were obtained by substituting the values of R_a, R_b and V_s in Eq. (2). The obtained deposition velocities of SO_2, HNO_3, SO_4^{2-} and NO_3^- are 0.32, 0.74, 1.16 and 1.07 cm s^{-1}, respectively. The reported deposition velocities are between 0.15 and 0.96 cm s^{-1} for SO_2 over Scots pine[37], 0.3 and 1.8 cm s^{-1} for HNO_3 over grass (pasture)[38], 1.2 ± 0.26 cm s^{-1} for SO_4^{2-} over wheat[39], and 0.7 and 1.1 cm s^{-1} for NO_3^- over *Querchus palustris*[40]. The deposition velocities determined by present parameterization method for SO_2, HNO_3, SO_4^{2-} and NO_3^- are in the reported range. As so many parameters and calculation steps are involved in computation of dry deposition velocity, a computer program has also been developed and used. Figure 1 shows the algorithm of computer program for calculation of dry deposition velocity and deposition flux by the present parameterization method.

3 Conclusion

A scheme for calculation of dry deposition velocity on natural surfaces (*Cassia siamea*) by parameterization method using meteorological data is proposed and a computer program is developed. The deposition velocities were determined for gaseous SO_2, HNO_3 and particulate SO_4^{2-} and NO_3^- to foliage (Cassia leaf) using meteorological data collected at Dayalbagh. The deposition velocities obtained by the current parameterization method are in the range of values determined by other methods. The present study is more accurate, realistic and rigorous.

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