

Supplemental Material

Appendix A: Calculation of Secondary Organic Aerosol Formation

Secondary organic aerosol formation is described as a two step process according to the procedure of Odum et al. [1]. Organic precursor species first are oxidized to form semi-volatile organic products according to reactions having the form:



where HC_a is the concentration of the organic vapor precursor, αx is the concentration of the relevant oxidizing species (e.g. OH , O_3 , NO_3); B_i and B_j are semi-volatile reaction products that are produced with stoichiometric coefficients α_i and α_j , respectively, relative to one molecule of HC_a . α_i and α_j are determined experimentally [2, 3]. Semi-volatile species then partition into an absorbing organic phase associated with all particles of a given type at a rate limited by the kinetic equation:

$$\frac{dA_{om,i}}{dt} = \frac{4\pi N D_i R_p}{\beta + 1} (B_{i,\infty} - B_{i,surf}) \quad (2)$$

where $A_{om,i}$ is the concentration of product i in the organic aerosol matrix, $B_{i,\infty}$ is the gas-phase concentration of semi-volatile species i far from the particle, $B_{i,surf}$ is the gas-phase concentration of semi-volatile species i at the particle surface, N is the particle number concentration, D_i is the diffusivity of the semi-volatile species of interest in air, R_p is the particle radius, $\beta = D_i/a_i c_i R_p$, a_i is the surface accommodation coefficient of species i (taken here to be 0.1 for all species), and c_i is the molecular velocity of species i [4]. The concentration of each semi-volatile gas-phase species at the particle surface is calculated using the equation:

$$B_{i,surf} = \frac{A_{om,i}}{K_{om,i} M_o} \quad (3)$$

where $K_{om,i}$ is a partitioning coefficient (measured experimentally), and M_o is the total organic aerosol mass concentration within the particle of interest [2, 3]. Table A1 lists the organic precursor species included in the lumped photochemical mechanism used by the air quality model, the symbols given to the semi-volatile oxidation products produced by that mechanism, the reaction product stoichiometric coefficient, and the value of the partitioning coefficients used to calculate the rate at which these species partition between the gas and particle phases.

Table A1: Parameters Used in the Calculation of Secondary Organic Aerosol Concentrations^a

Gas-phase Precursor ^b	First Semi-Volatile Oxidation Product B ₁			Second Semi-Volatile Oxidation Product B ₂			
	HC _a	Species Name	α_1^c	K_1 ($\mu\text{g}^{-1} \text{m}^3$) ^d	Species Name	α_2^c	K_2 ($\mu\text{g}^{-1} \text{m}^3$) ^d
AAR1	AEA1		0				
AAR2	AEA2		0				
AAR3	AEA3		0				
AAR4	AEA4a		0.083	0.093	AEA4b	0.22	0.001
AAR5	AEA5a		0.083	0.093	AEA5b	0.22	0.001
AAR6	AEA6a		0.038	0.042	AEA6b	0.167	0.0014
AAR7	AEA7a		0.083	0.093	AEA7b	0.22	0.001
OLE1	AEO1		9.2	1000			
OLE2	AEO2		0				
OLE3	AEO3a		0.048	0.171	AEO3b	0.595	0.004
APIN	AEAPa		0.048	0.171	AEAPb	0.595	0.004
BPIN	AEBPa		0.048	0.171	AEBPb	0.595	0.004
C7OL	AEC7		78	1000			
C8OL	AEC8		227	1000			
C9OL	AEC9		304	1000			
TOLU	AETLa		0.083	0.093	AETLb	0.22	0.001
BALD	AEBA		5	1000			
PBZN	AEPB		5	1000			
PHEN	AEPH		0				
CRES	AECR		0				
NPHE	AENP		0				

^a Secondary organic aerosol formation calculations follow the treatment of Odum et al. [1]. ^b Gas-phase precursor symbols correspond to the organic species used in the lumped photochemical mechanism of Pandis et al. [5]. ^c α_1 and α_2 are the stoichiometric coefficients that apply to the first and second semi-volatile reaction products, B_1 and B_2 respectively, from the reaction of one molecule of gaseous precursor HC_a [6]. ^d K_1 and K_2 are the gas / particle partitioning coefficients used in the calculation of equation (3). In those cases where there are no data on gas / particle partitioning of reaction products, the value of K_1 is set to a very high value and α_1 is set to the yield estimated by Pandis et al. [5] such that the model effectively reverts to the treatment of Pandis et al. [5] for that species.

References

- [1] Odum, J. R.; Dabdub, D.; Seinfeld, J. H. *Environ. Sci. Technol.*, 1998, *submitted for publication*.
- [2] Odum, J. R.; Jungkamp, T. P. W.; Griffin, R. J.; Forstner, H. J. L.; Flagan, R. C.; Seinfeld, J. H. *Environ. Sci. Technol.*, 1997, *31*(7),1890-1897.
- [3] Odum, J. R.; Jungkamp, T. P. W.; Griffin, R. J.; Flagan, R. C.; Seinfeld, J. H. *Science*, 1997, *276*,96-99.
- [4] Wexler, A. S.; Seinfeld, J. H. *Atmos. Environ.*, 1991, *25A*,2731-2748.
- [5] Pandis, S. N.; Harley, R. A.; Cass, G. R.; Seinfeld, J. H. *Atmos. Environ.*, 1992, *26A*,2269-2282.
- [6] Odum, J. R. personal communication to Michael J. Kleeman, 1997.