

Supplementary Information for

Estimation of the volatility distribution of organic aerosol combining thermodenuder and isothermal dilution measurements

5 **Evangelos E. Louvaris et al.**

Correspondence to: Spyros N. Pandis (spyros@andrew.cmu.edu)

1. Estimated COA volatility distributions and properties

10 **Table S1:** Estimated volatility distributions for the COA along with estimated properties of the two experiments using both TD and isothermal dilution measurements and only TD measurements using the approach of Karnezi et al. (2014).

Saturation concentration C^* ($\mu\text{g m}^{-3}$)	Experiment 1		Experiment 2	
	TD + Dilution	TD - only	TD + Dilution	TD - only
10^{-3}	0.27	0.18	0.23	0.18
10^{-2}	0.33	0.24	0.31	0.27
10^{-1}	-	-	0.23	0.22
10^0	0.19	0.25	0.09	0.18
10^1	0.06	0.11	0.07	0.09
10^2	0.06	0.10	0.07	0.06
10^3	0.09	0.12	-	-
ΔH_{vap} (kJ mol^{-1})	100 ± 14	83 ± 19	85 ± 9	79 ± 17
Accommodation coefficient	0.06 (0.01-0.3) ^a	0.14 (0.03-0.59) ^a	0.07 (0.01-0.34) ^a	0.07 (0.01-0.36) ^a
Average volatility $\log_{10}C^*$ ($\mu\text{g m}^{-3}$)	0.1	0.44	0.047	0.08

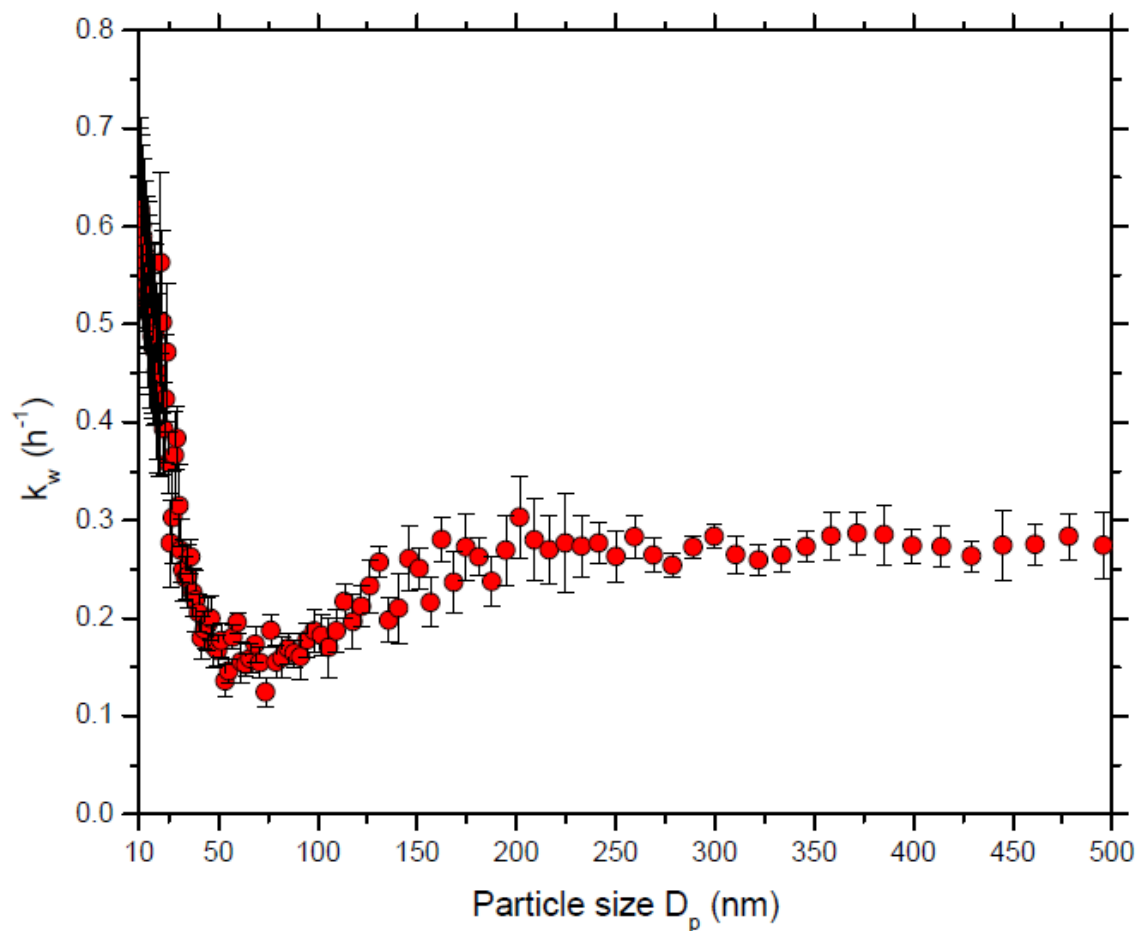
^a The values in parenthesis represent the corresponding uncertainties for the estimated accommodation coefficients

Table S2: Estimated volatility distributions for the various sensitivity tests.

	Saturation concentration ($\mu\text{g m}^{-3}$)							$\log_{10}C^*$ ($\mu\text{g m}^{-3}$)	ΔH_{vap} (kJ mol^{-1})	Accommodation coefficient
Experiment 1	10^{-3}	10^{-2}	10^{-1}	10^0	10^1	10^2	10^3			
Base case	0.27	0.33	-	0.19	0.06	0.06	0.09	0.1	100 ± 14	0.06 (0.01-0.30) ^a
Sensitivity to a_m										
$a_m=0.01$	0.20	0.25	-	0.30	0.09	0.07	0.09	0.25	104 ± 20	0.01
$a_m=0.1$	0.29	0.40	-	0.13	0.04	0.06	0.08	0.053	98 ± 6	0.1
$a_m=1.0$	0.35	0.40	-	0.06	0.03	0.06	0.10	0.04	95 ± 8	1.0
Sensitivity to ΔH_{vap}										
$\Delta H_{\text{vap}}=60 \text{ kJ mol}^{-1}$	0.10	0.39	-	0.32	0.05	0.05	0.09	0.25	60	0.07 (0.02-0.44) ^a
$\Delta H_{\text{vap}}=120 \text{ kJ mol}^{-1}$	0.50	0.20	-	0.10	0.04	0.06	0.10	0.036	120	0.01 (0.009-0.13) ^a
Experiment 2										
	10^{-3}	10^{-2}	10^{-1}	10^0	10^1	10^2	10^3			
Base case	0.23	0.31	0.23	0.09	0.07	0.07	-	0.047	85 ± 9	0.07 (0.01-0.34) ^a
Sensitivity to a_m										
$a_m=0.01$	0.14	0.24	0.30	0.14	0.09	0.09	-	0.12	89 ± 10	0.01
$a_m=0.1$	0.24	0.36	0.20	0.07	0.06	0.07	-	0.036	84 ± 8	0.1
$a_m=1.0$	0.33	0.35	0.16	0.04	0.05	0.07	-	0.022	82 ± 5	1.0
Sensitivity to ΔH_{vap}										
$\Delta H_{\text{vap}}=60 \text{ kJ mol}^{-1}$	0.16	0.32	0.27	0.11	0.07	0.07	-	0.066	60	0.09 (0.02-0.44) ^a
$\Delta H_{\text{vap}}=120 \text{ kJ mol}^{-1}$	0.35	0.33	0.12	0.05	0.06	0.09	-	0.026	120	0.025 (0.01-0.085) ^a

^a The values in parenthesis represent the corresponding uncertainties for the estimated accommodation coefficients

2. Wall loss corrections during isothermal dilution



15 **Figure S1:** Wall loss rate constants as a function of particle size (red circles) for the isothermal dilution chamber during Experiment 1. The error bars represent ± 1 standard deviation.

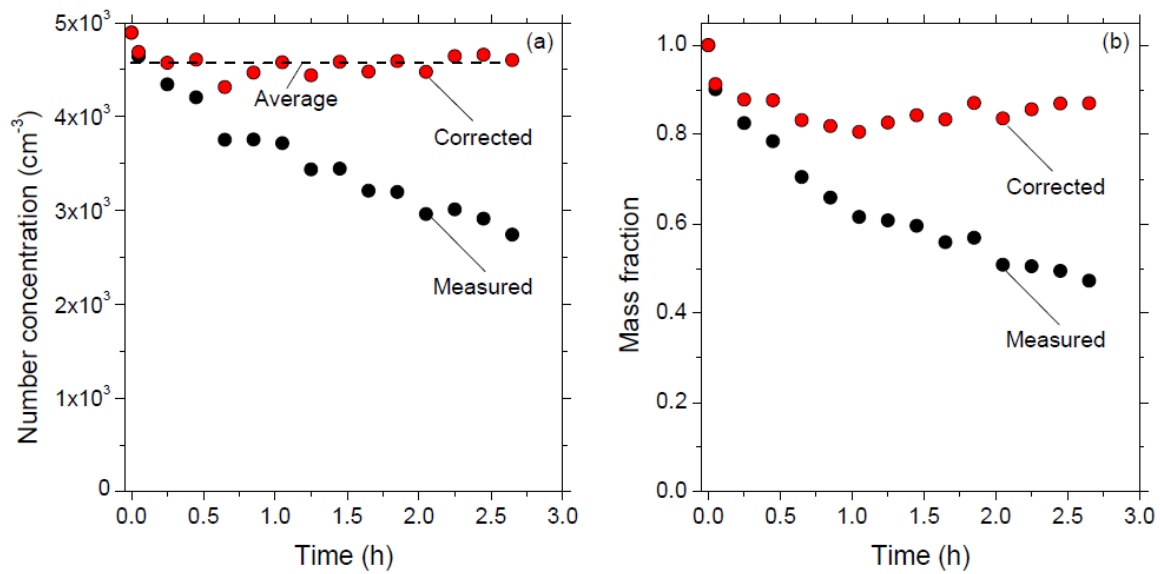
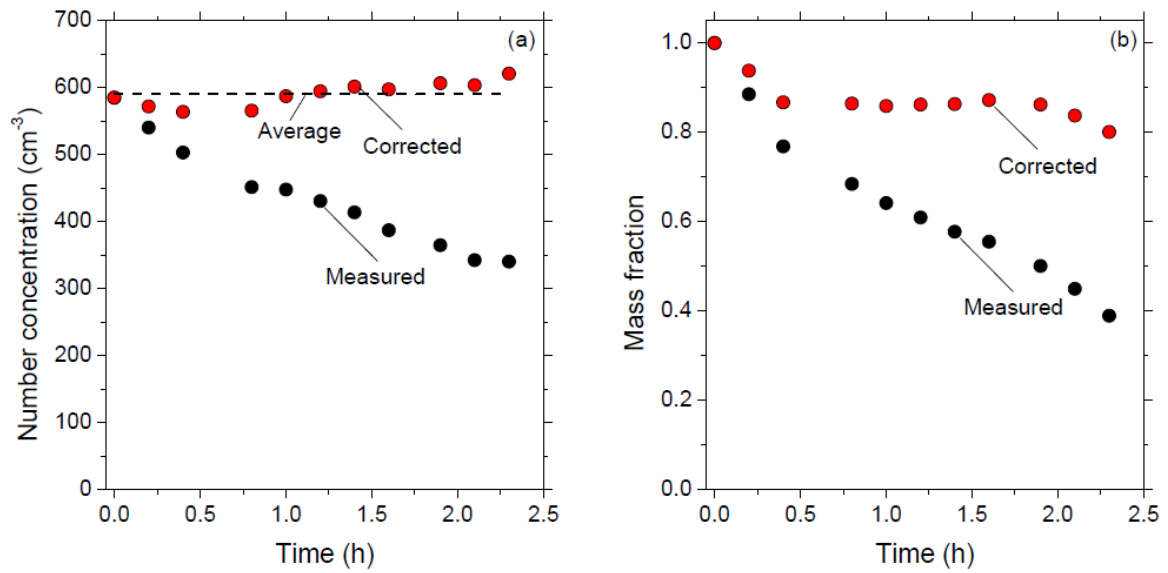


Figure S2: (a) Number concentration during isothermal dilution of Experiment 1 as a function of time measured by the SMPS. (b) Mass fraction as function of time during isothermal dilution of Experiment 1 measured by the SMPS.



20 **Figure S3:** (a) Number concentration during isothermal dilution of Experiment 2 as a function of time measured by the SMPS. (b) Mass fraction as function of time during isothermal dilution of Experiment 2 measured by the SMPS.

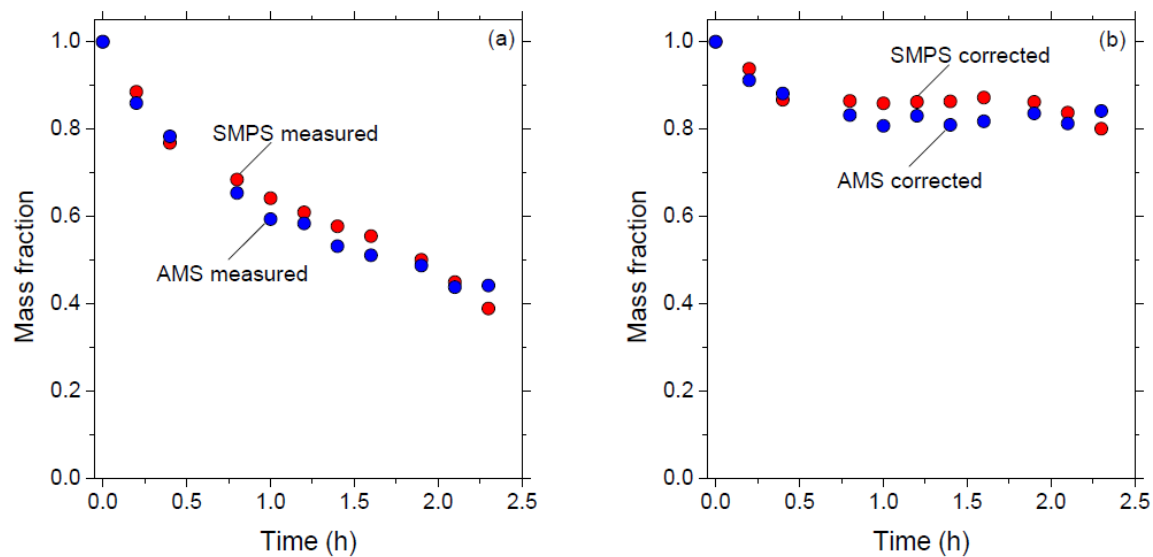


Figure S4: (a) Measured mass fractions as function of time during isothermal dilution of Experiment 2. (b) Corrected mass fractions as a function of time during isothermal dilution of Experiment 2.

25

3. Sensitivity tests to accommodation coefficient

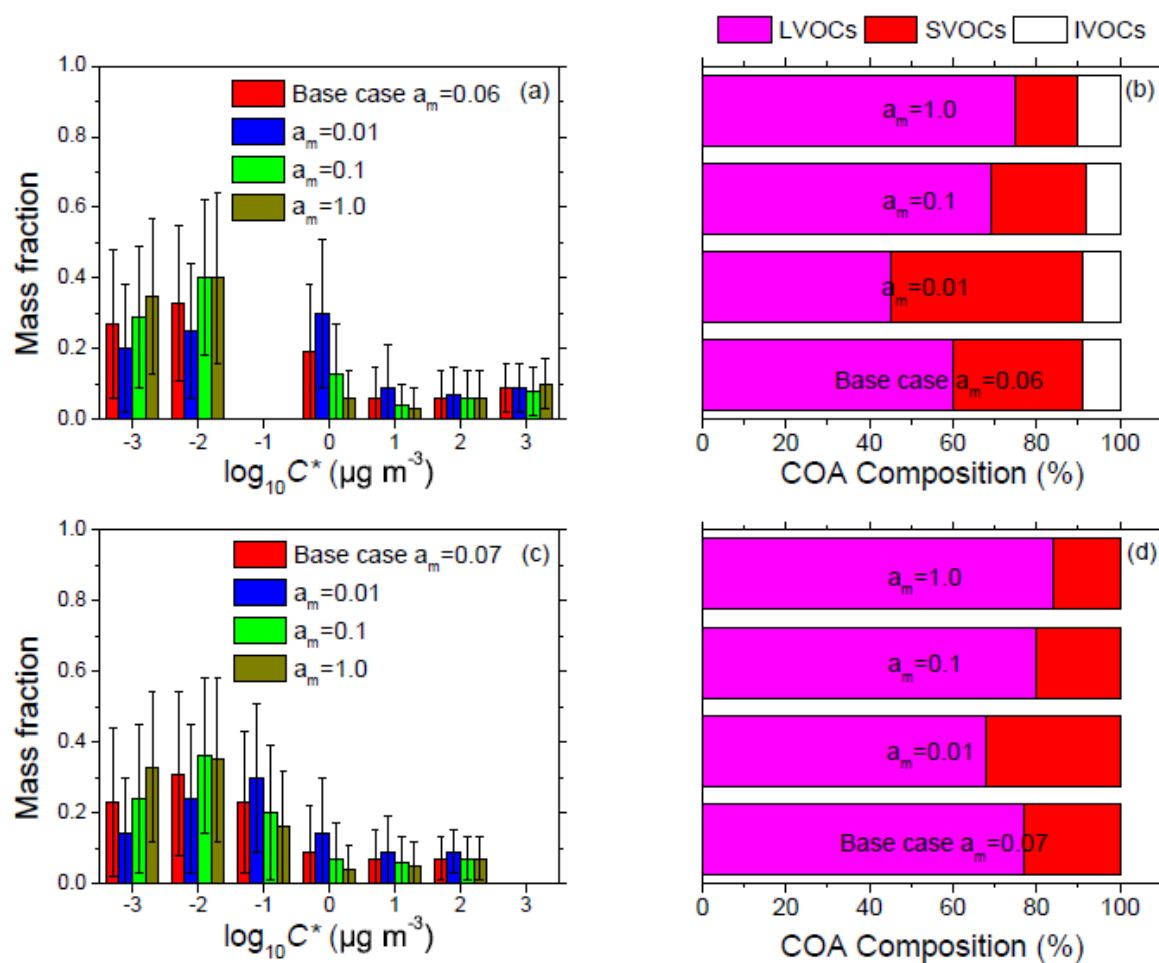


Figure S5: (a) Estimated volatility distributions assuming different accommodation coefficients for Experiment 1 using the approach of Karnezi et al. (2014). The error bars represent the corresponding uncertainty ranges. Red, blue, green, and dark yellow bars represent the estimated volatility distributions for the base case, $a_m=0.01$, $a_m = 0.1$, and $a_m=1.0$ respectively. (b) Corresponding COA composition of Experiment 1. LVOCs are represented in magenta, SVOCs in red, and IVOCs in white. (c) Estimated volatility distributions for different accommodation coefficients for Experiment 2 using the approach of Karnezi et al. (2014). The error bars represent the corresponding uncertainty ranges. Red, blue, green, and dark yellow bars represent the estimated volatility distributions for the base case, $a_m=0.01$, $a_m = 0.1$, and $a_m=1.0$ respectively. (d) Corresponding COA composition of Experiment 2. LVOCs are represented in magenta, SVOCs in red.

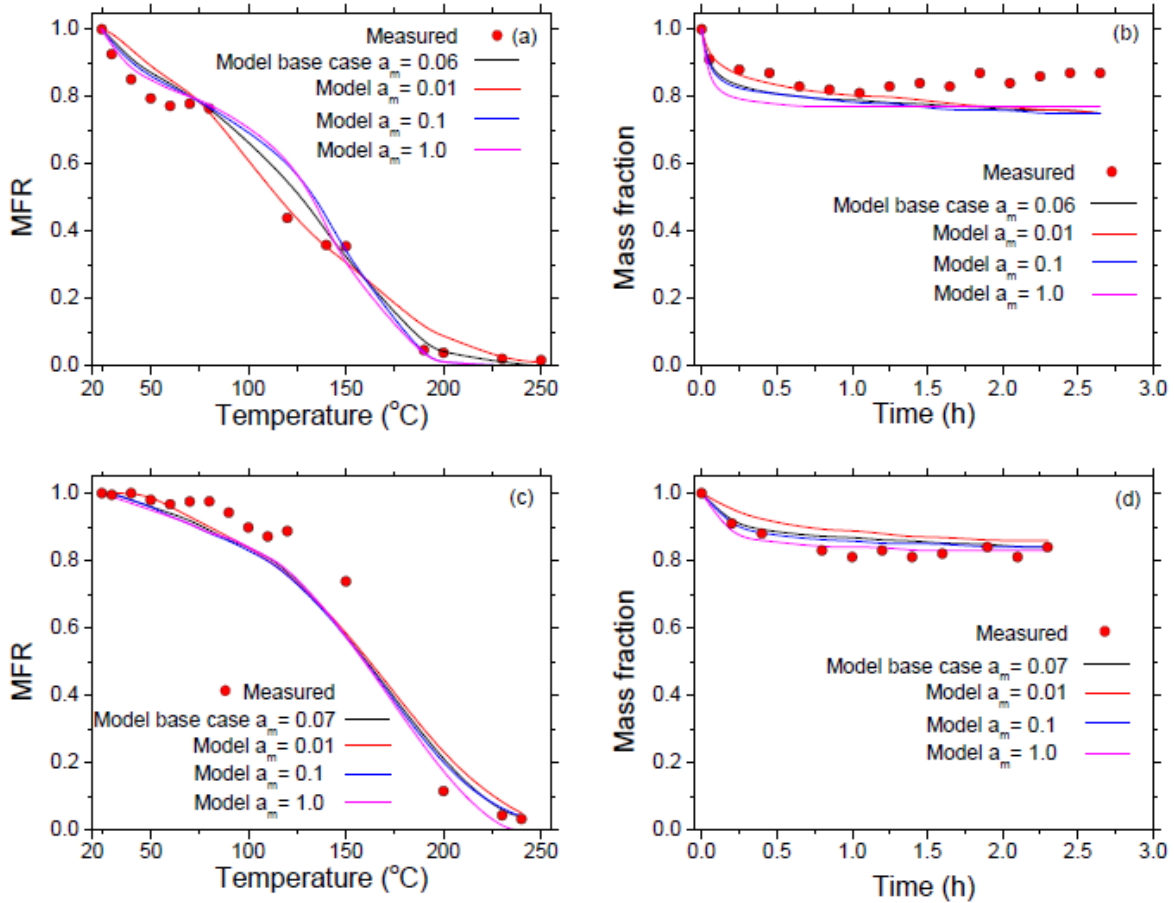
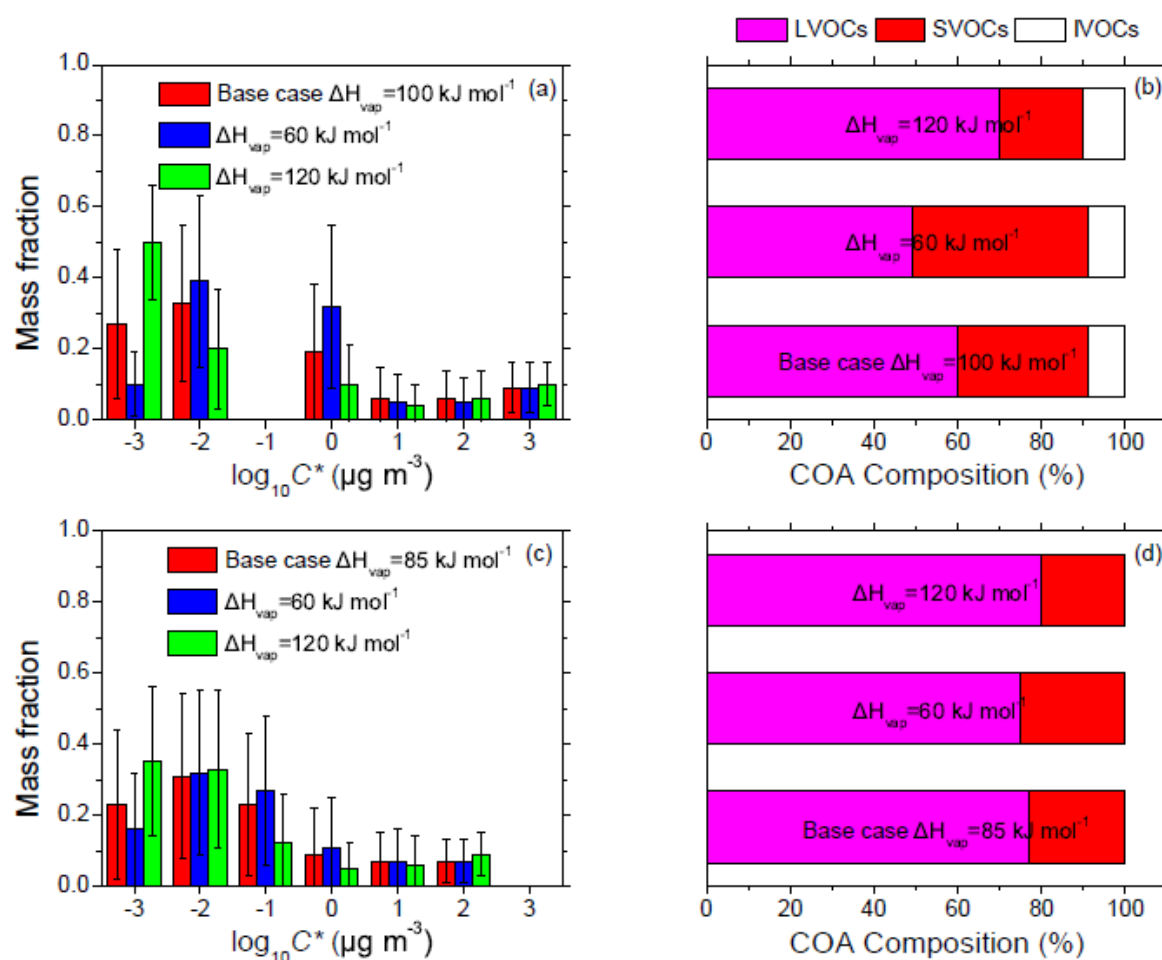


Figure S6: (a) Thermogram of the OA TD measurements of Experiment 1. Red circles represent the loss corrected measurements and the black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, blue, and magenta lines represent the model predictions for $a_m = 0.01$, $a_m = 0.1$, and $a_m = 1.0$ respectively. (b) Mass fraction during isothermal dilution as a function of time of Experiment 1. Red circles represent the loss corrected measurements and the black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, blue, and magenta lines represent the model predictions for $a_m = 0.01$, $a_m = 0.1$, and $a_m = 1.0$ respectively. (c) Thermogram of the OA TD measurements of Experiment 2. Red circles represent the loss corrected measurements and the black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, blue, and magenta lines represent the model predictions for $a_m = 0.01$, $a_m = 0.1$, and $a_m = 1.0$ respectively. (d) Mass fraction during isothermal dilution as a function of time of Experiment 2. Red circles represent the loss corrected measurements and the black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, blue, and magenta lines represent the model predictions for $a_m = 0.01$, $a_m = 0.1$, and $a_m = 1.0$ respectively.

4. Sensitivity tests to vaporization enthalpy



55 **Figure S7:** (a) Estimated volatility distributions assuming different vaporization enthalpies
 for Experiment 1 using the approach of Karnezi et al. (2014). The error bars represent the
 corresponding uncertainty ranges. Red, blue, and green bars represent the estimated volatility
 distributions for the base case, $\Delta H_{\text{vap}}=60 \text{ kJ mol}^{-1}$, and $\Delta H_{\text{vap}}=120 \text{ kJ mol}^{-1}$ respectively. (b)
 60 Corresponding COA composition of Experiment 1. LVOCs are represented in magenta,
 SVOCs in red, and IVOCs in white. (c) Estimated volatility distributions assuming different
 vaporization enthalpies for Experiment 2 using the approach of Karnezi et al. (2014). The
 error bars represent the corresponding uncertainty ranges. Red, blue, and green bars represent
 the estimated volatility distributions for the base case, $\Delta H_{\text{vap}}=60 \text{ kJ mol}^{-1}$, and $\Delta H_{\text{vap}}=120 \text{ kJ}$
 65 mol^{-1} respectively. (d) Corresponding COA composition of Experiment 2. LVOCs are
 represented in magenta, SVOCs in red.

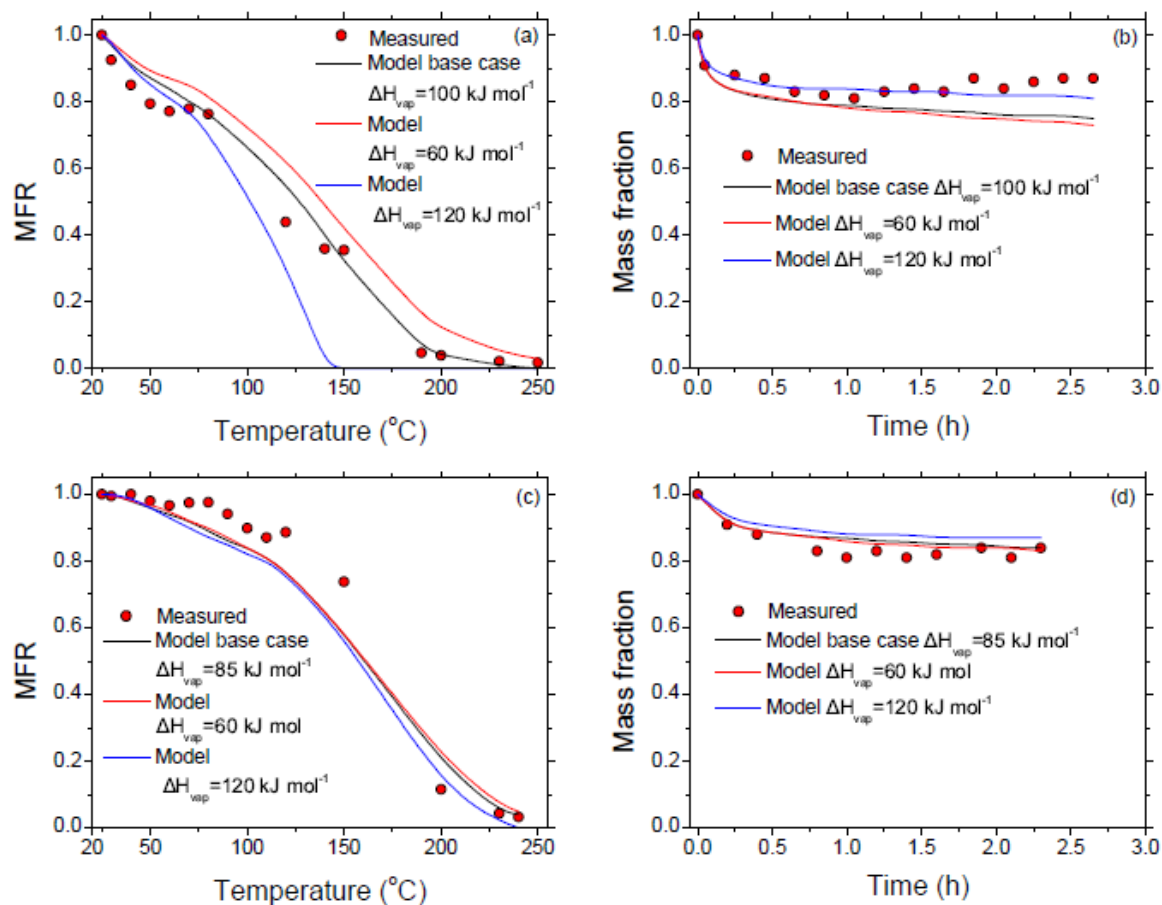


Figure S8: (a) Thermogram of the OA TD measurements of Experiment 1. Red circles represent the loss corrected measurements and black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, and blue lines represent the model predictions for $\Delta H_{\text{vap}} = 60$, and $\Delta H_{\text{vap}} = 120 \text{ kJ mol}^{-1}$ respectively. (b) Mass fraction during isothermal dilution as a function of time of Experiment 1. Red circles represent the loss corrected measurements and black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, and blue lines represent the model predictions for $\Delta H_{\text{vap}} = 60$, and $\Delta H_{\text{vap}} = 120 \text{ kJ mol}^{-1}$ respectively. (c) Thermogram of the OA TD measurements of Experiment 2. Red circles represent the loss corrected measurements and black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, and blue lines represent the model predictions for $\Delta H_{\text{vap}} = 60$, and $\Delta H_{\text{vap}} = 120 \text{ kJ mol}^{-1}$ respectively. (d) Mass fraction during isothermal dilution as a function of time of Experiment 2. Red circles represent the loss corrected measurements and black line represents the best fit for the base case estimated by the model of Karnezi et al. (2014). Red, and blue lines represent the model predictions for $\Delta H_{\text{vap}} = 60$, and $\Delta H_{\text{vap}} = 120 \text{ kJ mol}^{-1}$ respectively.

References

Karnezi, E., Riipinen, I. and Pandis, S. N.: Measuring the atmospheric organic aerosol volatility distribution: A theoretical analysis, *Atmos. Meas. Tech.*, 7, 2953–2965, 2014.