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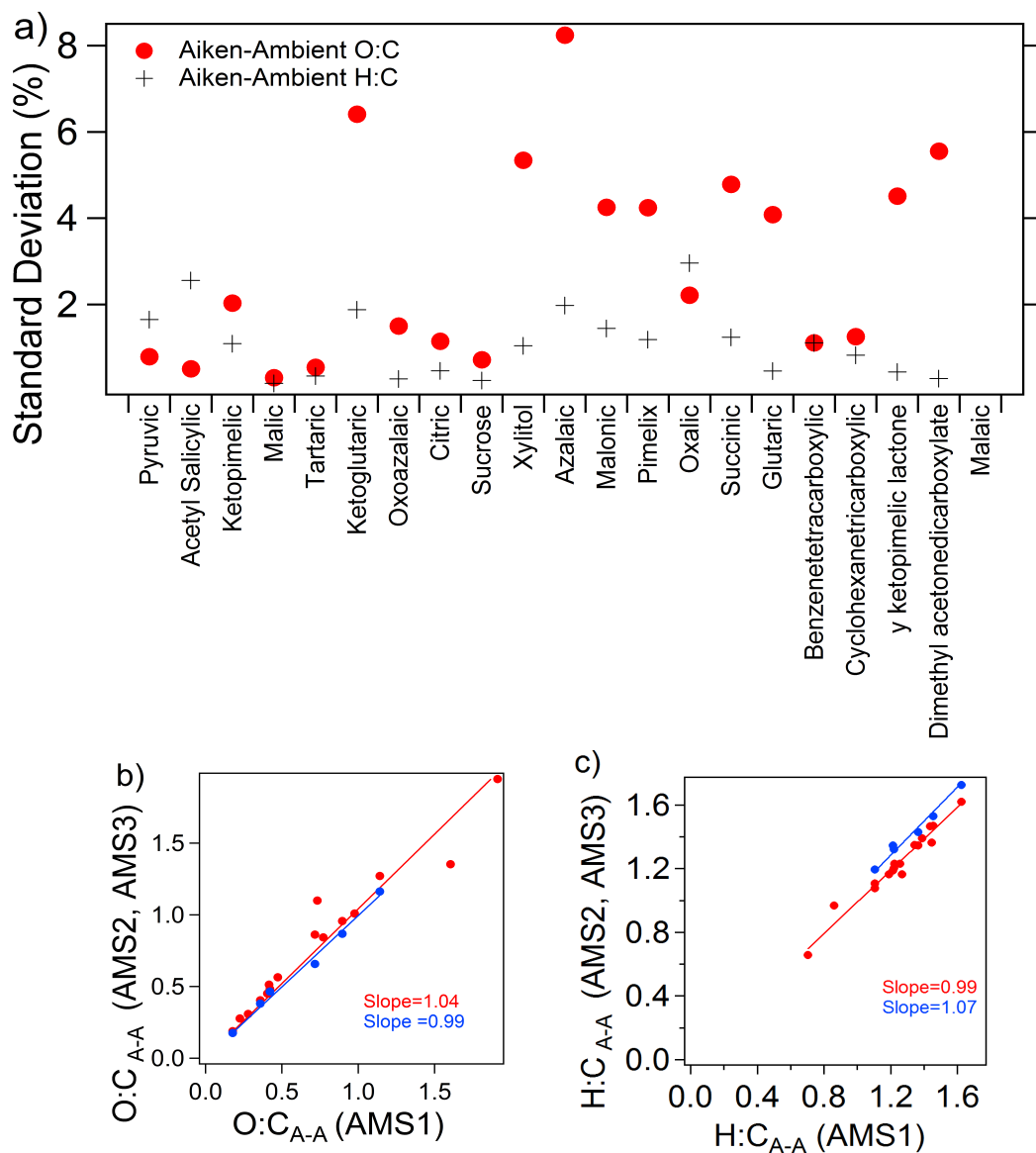


*Supplement of*

**Elemental ratio measurements of organic compounds  
using aerosol mass spectrometry: characterization,  
improved calibration, and implications**

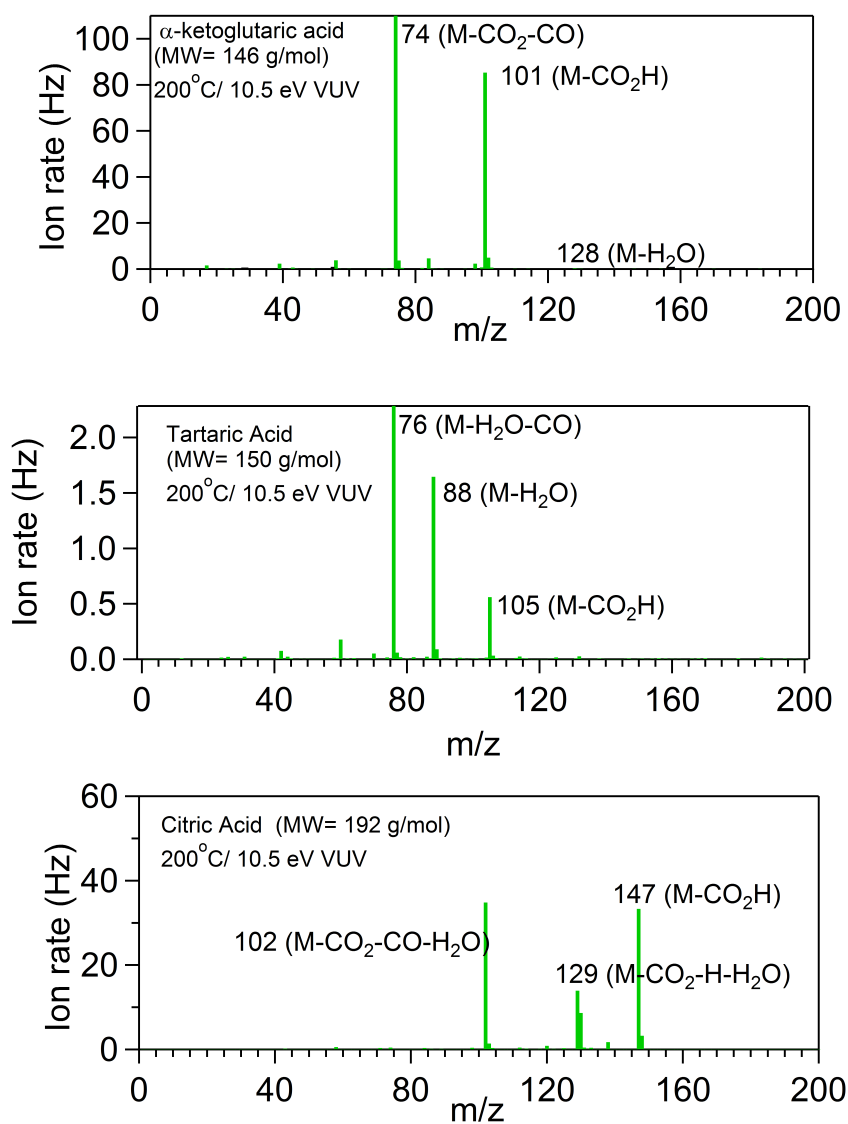
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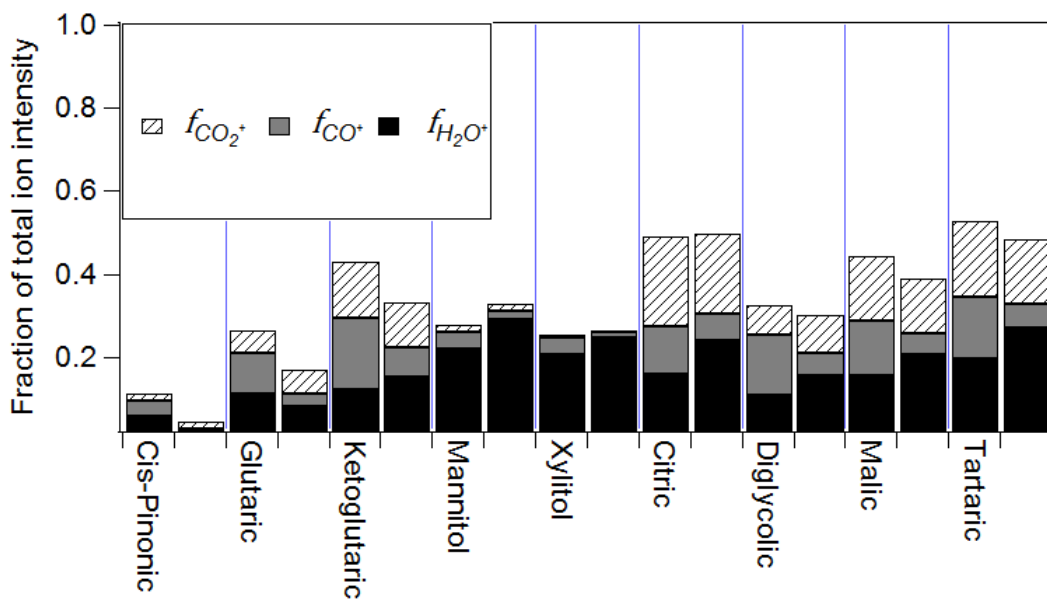
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2 Figure S1: a) Standard deviations in repeated elemental ratio measurements of several  
 3 standards obtained with a single instrument. Standard deviations are shown for O:C and  
 4 H:C ratios calculated with the Aiken-Ambient method. b) Scatter plots of Aiken-  
 5 Ambient O:C ratios calculated for several standards with three different AMS instrument  
 6 (AMS\_1, AMS\_2, and AMS\_3. c) Scatter plots of Aiken-Ambient H:C ratios calculated  
 7 for several standards with three different AMS instrument (AMS\_1, AMS\_2, and  
 8 AMS\_3).



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2 Figure S2: VUV-AMS spectra of three different organic standards obtained with  
 3 vaporizer set to 200°C and VUV energy of 10.5 eV. Ketoglutaric acid, tartaric acid, and  
 4 citric acid correspond to multifunctional organic acid species with carbonyl, hydroxyl,  
 5 and polycarboxylic functional groups, respectively.



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2 Figure S3: Vaporizer temperature dependence of the fractional AMS ion intensity  
 3 measured for  $CO_2^+$ ,  $CO^+$ , and  $H_2O^+$  for some of the standards measured in this study. For  
 4 each standard, fractional ion intensities measured at 600°C (first bar) and 200°C (second  
 5 bar) are shown.

6

Multifunctional, Esters  
Polyacids, Alcohols, Diacids

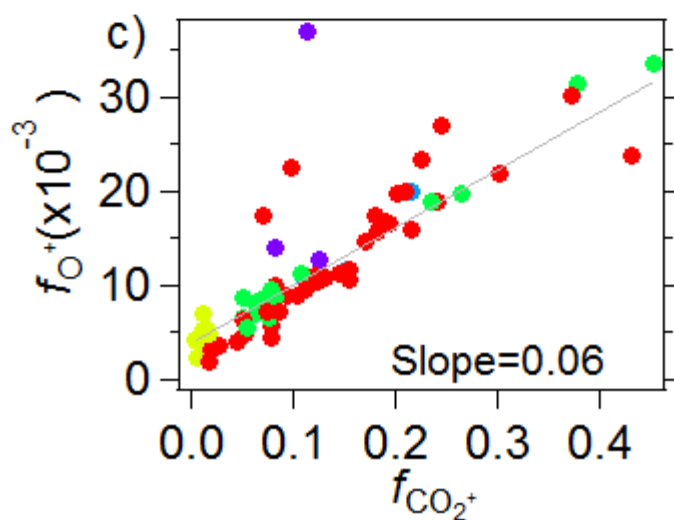
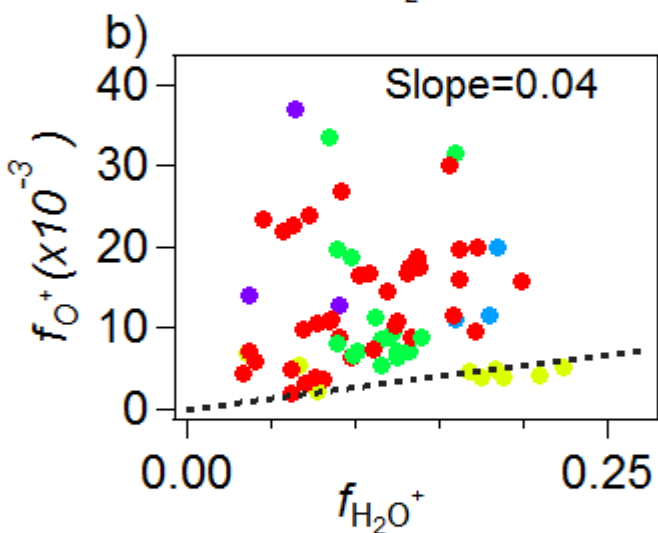
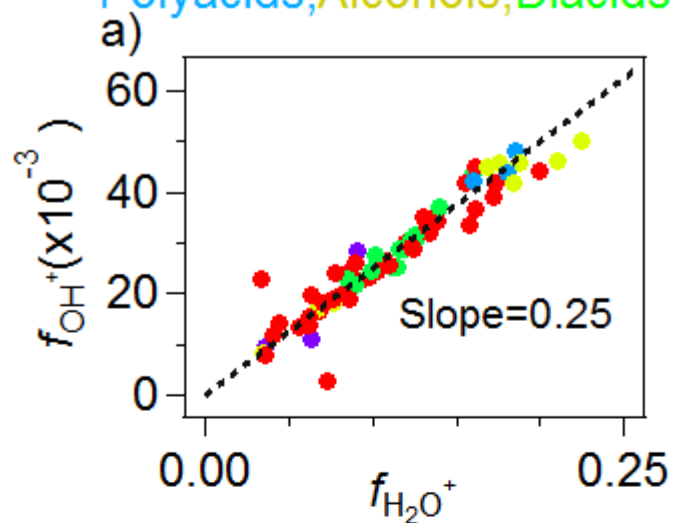
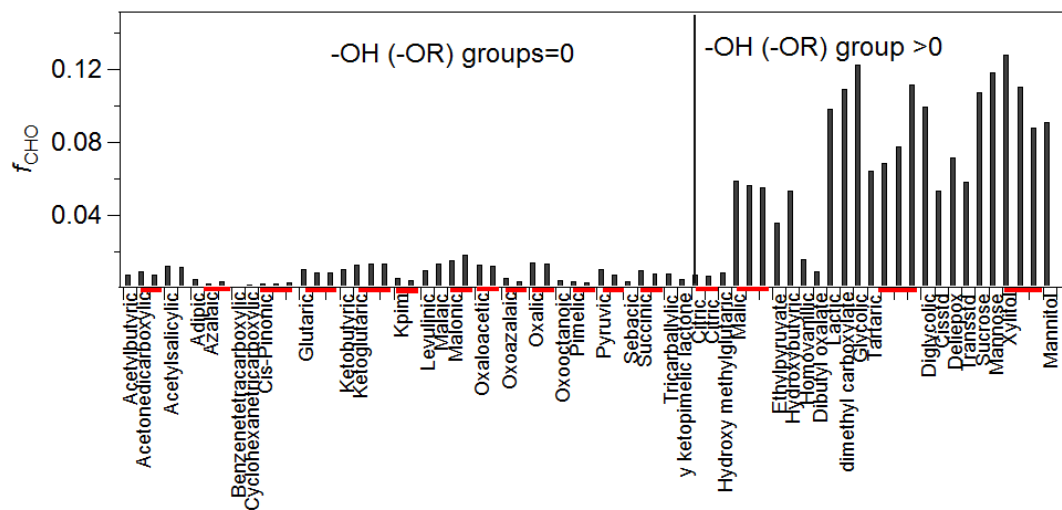
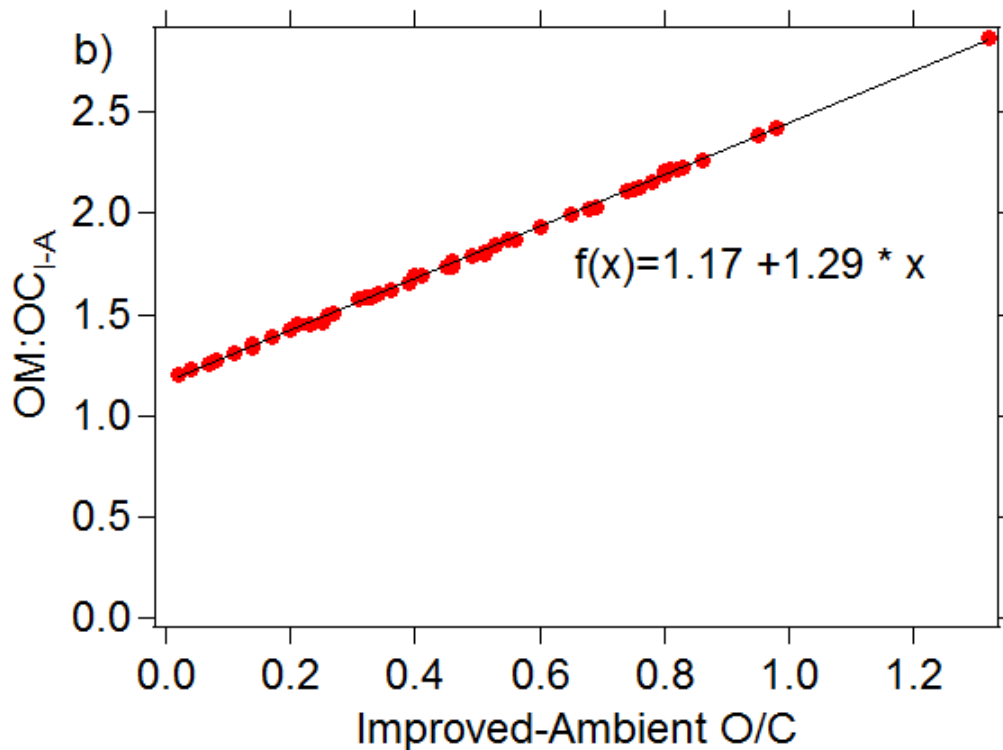
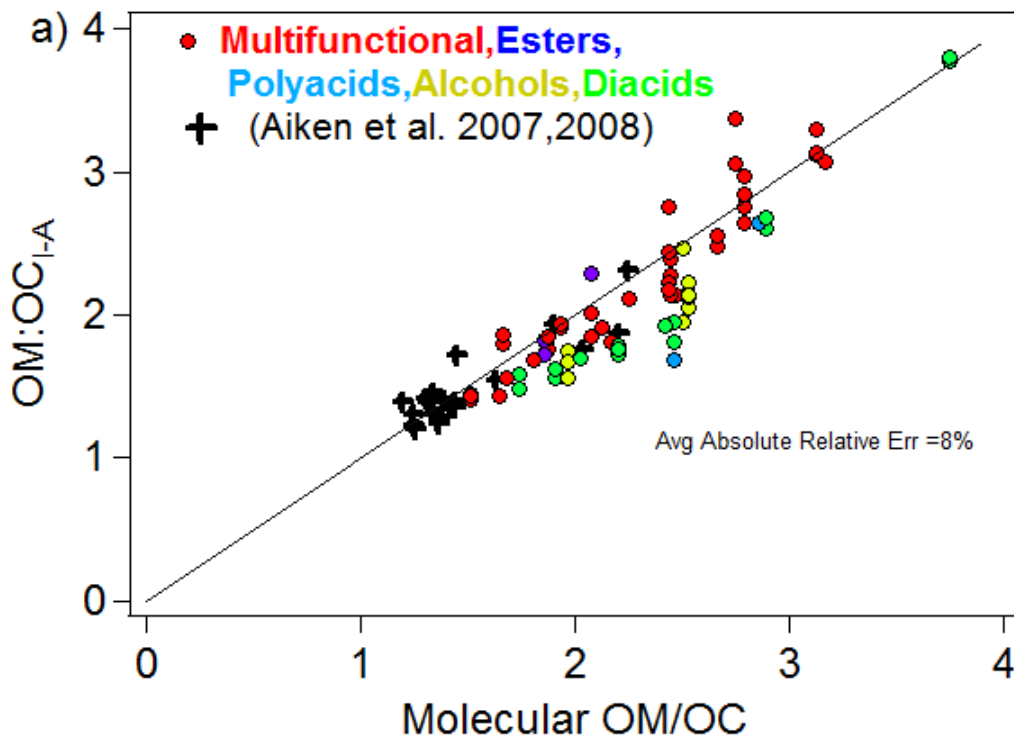


Figure S4: Scatter plots of key ions observed in the AMS spectra of laboratory standards. Panel a shows  $\text{OH}^+$  vs.  $\text{H}_2\text{O}^+$ , panel b shows  $\text{O}^+$  vs.  $\text{H}_2\text{O}^+$ , and panel c shows  $\text{O}^+$  vs.  $\text{CO}_2^+$ . The dashed line denotes the default relationships assumed when analyzing AMS data with the Aiken-Ambient method.

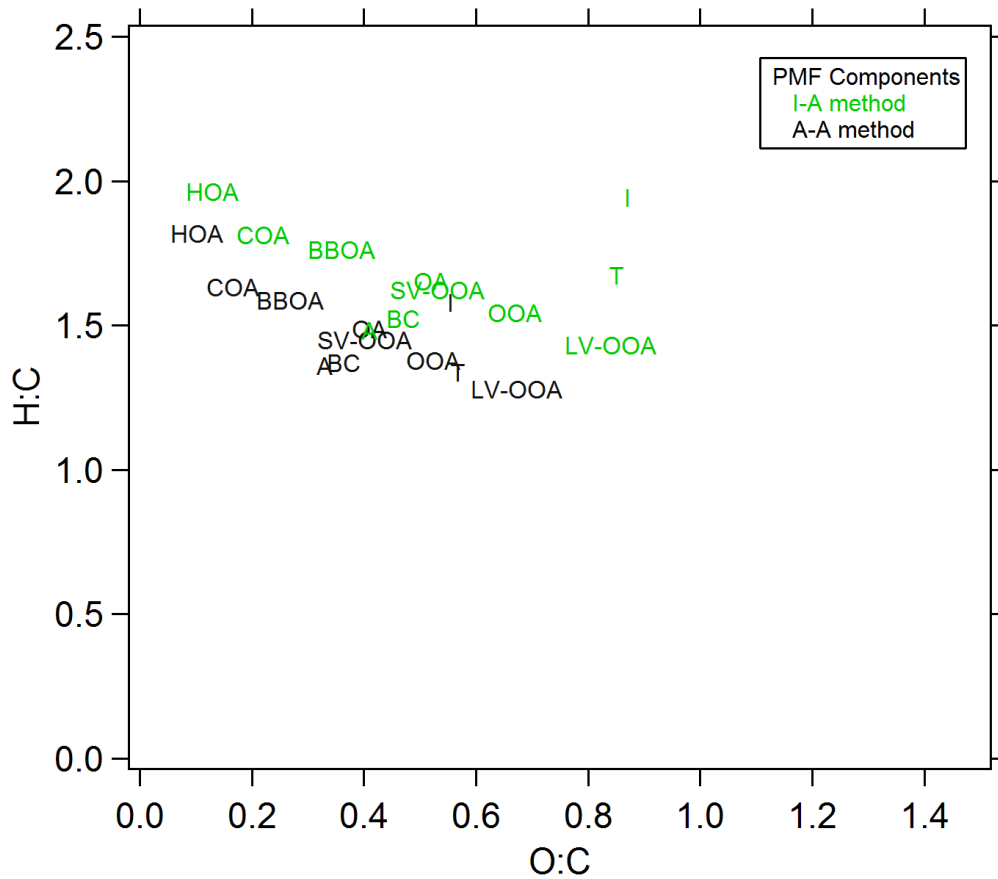


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2 Figure S5: Dependence of the fractional  $\text{CHO}^+$  ion intensity on functional composition of  
 3 OA standards. The standards are separated into two groups according to those that  
 4 contain at least one -OH or -OR functional group and those that don't contain any of  
 5 those functional groups.



- 1 Figure S6: a) Scatter plot between OM/OC values calculated with Improved-Ambient
- 2 method and the known OM/OC values for standard molecules. b) Scatter plot between
- 3 OM/OC values and O:C values calculated with the Improved-Ambient method for
- 4 standard molecules. The black line shows a linear fit through the data.



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3 Figure S7: a) Van Krevelen plots of ambient PMF components (HOA, COA, BBOA, SV-  
 4 OOA, LV-OOA, OOA) and chamber SOA ( $\alpha$ -pinene(A), toluene (T) and isoprene (I))  
 5 obtained when the O:C and H:C values are calculated with the A-A and I-A methods.



1 Table S1. Improved-Ambient (I-A) results for previously reported ambient OA components.

Ambient PMF components	Reference	O:C <sub>I-A</sub>	Change (%)	H:C <sub>I-A</sub>	Change (%)	OM:OC <sub>I-A</sub>	Change (%)	$\overline{OS}_c(I-A)$	Absolute Change
HOA	(Aiken et al., 2009)	0.21	31	2.03	10	1.45	6	-1.61	-0.09
	(DeCarlo et al., 2010)	0.07	24	1.92	7	1.26	2	-1.77	-0.10
	(Docherty et al., 2011)	0.02	26	2.10	7	1.21	2	-2.05	-0.13
	(Ge et al., 2012)	0.11	27	1.95	8	1.33	3	-1.72	-0.10
	(Gong et al., 2012)	0.11	26	1.94	8	1.34	3	-1.71	-0.09
	(He et al., 2011)	0.14	25	1.83	8	1.35	4	-1.55	-0.07
	(Wang et al., 2010)	0.14	28	1.88	9	1.34	4	-1.60	-0.09
	(Huang et al., 2012)	0.20	28	1.93	9	1.43	5	-1.52	-0.07
	(Huang et al., 2013) (Winter)	0.14	29	2.02	9	1.38	4	-1.74	-0.11
	(Huang et al., 2013) (Summer)	0.17	29	1.93	9	1.40	5	-1.60	-0.09
	(Mohr et al., 2012)	0.04	26	2.10	7	1.23	2	-2.03	-0.13
	(Saarikoski et al., 2012)	0.26	24	1.84	8	1.50	5	-1.32	-0.03
	(Setyan et al., 2012)	0.11	34	1.99	11	1.31	4	-1.77	-0.14
(Sun et al., 2011)	0.08	26	1.96	7	1.26	3	-1.81	-0.10	
BBOA	(Aiken et al., 2009)	0.40	34	1.88	11	1.69	10	-1.08	0.01
	(DeCarlo et al., 2010)	0.55	31	1.60	12	1.88	11	-0.49	0.09
	(Ge et al., 2012)	0.46	40	1.78	14	1.79	12	-0.86	0.04
	(Gong et al., 2012)	0.25	31	1.55	10	1.50	6	-1.05	-0.02
	(He et al., 2011)	0.45	42	1.69	15	1.81	12	-0.79	0.04
	(Huang et al., 2011)	0.34	32	1.79	10	1.60	8	-1.10	0.00
	(Huang et al., 2013) (Winter)	0.36	35	1.70	12	1.66	9	-0.97	0.00
	(Mohr et al., 2012)	0.31	30	1.94	10	1.58	8	-1.31	-0.02
	(Saarikoski et al., 2012)	0.33	44	1.77	16	1.59	11	-1.11	-0.04
COA	(Ge et al., 2012)	0.14	27	1.85	8	1.35	4	-1.57	-0.07
	(Wang et al., 2010)	0.14	28	1.88	9	1.34	4	-1.60	-0.09
	(Mohr et al., 2012)	0.27	31	1.73	10	1.51	7	-1.18	-0.03
	(Sun et al., 2011)	0.23	26	1.71	8	1.44	5	-1.26	-0.04
OOA	(Aiken et al., 2009) (OOA)	0.80	33	1.67	14	2.20	15	-0.07	0.20
	(DeCarlo et al., 2010) (SVOOA)	0.83	29	1.46	13	2.23	13	0.20	0.21
	(DeCarlo et al., 2010) (LVOOA)	1.32	29	1.29	16	2.87	17	1.34	0.42
	(Docherty et al., 2011) (SV-OOA)	0.32	40	1.88	14	1.59	10	-1.23	-0.04
	(Docherty et al., 2011) (LVOOA)	0.86	20	1.40	10	2.26	10	0.32	0.16
	(Ge et al., 2012) (OOA)	0.55	31	1.60	12	1.88	11	-0.50	0.09
	(Gong et al., 2012) (SV-OOA)	0.46	25	1.46	10	1.75	8	-0.53	0.06
	(Gong et al., 2012) (LV-OOA)	0.68	24	1.43	10	2.06	10	-0.06	0.14
	(He et al., 2011) (SV-OOA)	0.60	32	1.64	13	1.95	12	-0.45	0.10
	(He et al., 2011) (LV-OOA)	0.76	29	1.43	14	2.17	12	0.09	0.17
	(Wang et al., 2010) (OOA1)	0.56	17	1.48	7	1.87	7	-0.35	0.07
	(Wang et al., 2010) (OOA2)	0.65	38	1.54	16	1.99	15	-0.24	0.15
	(Huang et al., 2011) (SV-OOA)	0.49	27	1.63	10	1.79	9	-0.64	0.06
	(Huang et al., 2011) (LV-OOA)	0.80	26	1.45	11	2.19	12	0.16	0.18
	(Huang et al., 2012) (SV-OOA)	0.45	30	1.65	12	1.74	10	-0.74	0.04
	(Huang et al., 2012) (LV-OOA)	0.81	25	1.66	11	2.22	11	-0.04	0.15
	(Huang et al., 2013) (Winter; OOA)	0.75	28	1.45	12	2.16	12	0.06	0.17
	(Huang et al., 2013) (Summer; OOA)	0.53	28	1.63	11	1.86	10	-0.58	0.07
	(Mohr et al., 2012) (SV-OOA)	0.41	29	1.75	11	1.70	9	-0.92	0.02
	(Mohr et al., 2012) (LV-OOA)	0.98	31	1.35	14	2.42	16	0.62	0.30
	(Saarikoski et al., 2012) (OOAa)	0.95	20	1.46	10	2.39	10	0.44	0.19
	(Saarikoski et al., 2012) (OOAb)	0.80	28	1.58	12	2.20	13	0.03	0.18
	(Saarikoski et al., 2012) (OOAc)	0.69	21	1.36	9	2.03	9	0.01	0.12
	(Setyan et al., 2012) (LO-OOA)	0.51	21	1.46	8	1.80	7	-0.45	0.06
	(Setyan et al., 2012) (MO-OOA)	0.74	37	1.52	15	2.11	15	-0.04	0.20
	(Sun et al., 2011) (SVOOA)	0.51	33	1.58	13	1.81	11	-0.56	0.08
(Sun et al., 2011) (LVOOA)	0.78	24	1.43	11	2.16	11	0.14	0.17	

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2 Table S2. Improved-Ambient (I-A) results for total OA from previously reported ambient  
 3 field campaigns.

Reference	O:C <sub>I-A</sub>	Change (%)	H:C <sub>I-A</sub>	Change (%)	OM:OC <sub>I-A</sub>	Change (%)	$\overline{OS}_c$ (I-A)	Absolute Change
(Aiken et al., 2009)	0.53	32	1.82	12	1.86	11	-0.77	0.06
(Docherty et al., 2011)	0.44	27	1.71	10	1.73	9	-0.82	0.03
(Chen et al., 2009 and 2014)	0.60	34	1.65	14	1.94	13	-0.44	0.11
(Ge et al., 2012)	0.35	30	1.75	10	1.63	8	-1.05	0.00
(Gong et al., 2012)	0.50	26	1.63	9	1.87	9	-0.62	0.07
(He et al., 2011)	0.39	31	1.83	12	1.71	9	-1.04	-0.01
(Wang et al., 2010)	0.41	26	1.63	9	1.69	8	-0.80	0.03
(Huang et al., 2011)	0.60	27	1.64	11	1.94	11	-0.44	0.10
(Huang et al., 2012)	0.40	28	1.92	11	1.69	8	-1.13	-0.02
(Huang et al., 2013) (Winter)	0.43	30	1.73	11	1.75	9	-0.87	0.03
(Huang et al., 2013) (Summer)	0.36	28	1.94	10	1.67	8	-1.22	-0.02
(Martin et al., 2008)	0.69	26	1.40	11	2.04	11	-0.01	0.15
(Mohr et al., 2012)	0.41	30	1.77	11	1.70	9	-0.94	0.01
(Ovadnevaite et al., 2011)	0.70	17	1.34	8	2.05	8	0.06	0.11
(Poulain et al., 2011) (Summer)	0.52	17	1.51	7	1.83	6	-0.47	0.05
(Poulain et al., 2011) (Autumn)	0.54	14	1.48	7	1.84	6	-0.40	0.04
(Poulain et al., 2011) (Winter)	0.53	16	1.48	7	1.83	6	-0.41	0.05
(Robinson et al., 2011)	0.71	45	1.62	20	2.08	18	-0.20	0.17
(Saarikoski et al., 2012)	0.59	26	1.64	10	1.92	10	-0.46	0.09
(Setyan et al., 2012)	0.56	28	1.53	11	1.88	10	-0.40	0.10
(Sun et al., 2011)	0.46	28	1.65	11	1.75	9	-0.73	0.04

4

1 Table S3. Comparison of different versions of the organic fragmentation waves that can  
2 be used for AMS analysis

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4

Frag_organic		
m/z	Default AMS Frag. Table Allan et al., (2004)	Hildebrandt Ruiz et al., (2014) Correction
1		<b>Hwave</b> *frag_organic [18]
16	0.04*frag_organic [18]	0.04*frag_organic [18]
17	0.25*frag_organic [18]	0.25*frag_organic [18]
18	1*frag_organic [44]	1*frag_organic [44]

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1 **Calculation of H<sup>+</sup>/H<sub>2</sub>O<sup>+</sup> for Organic Frag Wave in AMS fragmentation table.**

2 The current treatment of water fragmentation does not account for the H-atoms which  
3 were bound to HO<sup>+</sup> and O<sup>+</sup> before fragmentation. The neglected mass from H-atoms is  
4 negligible when calculating organic aerosol mass concentrations. However, the neglected  
5 H<sup>+</sup> signal does affect oxidation state ( $\overline{OS}_c \sim 2 \times O:C - H:C$ ) calculations from current H:C  
6 and O:C AMS values. In particular, the  $\overline{OS}_c$  values are not invariant with respect to  
7 hydration/dehydration processes as they should be; the oxidation state decreases with  
8 dehydration because the prescribed H/O ratio of water in the AMS analysis is less than 2.  
9 Here we calculate H<sup>+</sup>/H<sub>2</sub>O<sup>+</sup> that needs to be added to the standard organic fragmentation  
10 wave to obtain an H/O ratio of 2 in the total signal of H<sub>2</sub>O determined by AMS data  
11 analysis. The addition of H<sup>+</sup> changes the organic H:C ratio calculated in elemental  
12 analysis of the organic aerosol (OA) and therefore the oxidation state ( $\overline{OS}_c$ ) estimated  
13 from O:C and H:C ratios.

14 The ratio of H<sup>+</sup>/H<sub>2</sub>O<sup>+</sup> needed to add to the updated fragmentation table to keep  $\overline{OS}_c$   
15 constant upon (de-) hydration can be calculated as follows:

16

17 For any given time point, let:

18  $x = H^+/H_2O^+$

19  $z =$  initial mass of H<sub>2</sub>O<sup>+</sup>,  $y =$  factor by which H<sub>2</sub>O changes,

20  $f_{OH} = OH^+/H_2O^+ = 0.25$  and  $f_O = O^+/H_2O^+ = 0.04$

21 Accounting for the contribution of <sup>18</sup>O:

22  $f'_{OH} = 1.00205499 * f_{OH}$  and  $f'_O = 1.00205499 * f_O$

23  $mw_i =$  molecular weights

24  $f_{cal}^{OC}, f_{cal}^{HC}$  = calibration factors for O:C and H:C

25 The Aiken-Ambient and Aiken-Explicit values are 0.75 and 0.91, respectively

26 The Improved-Ambient values are (See Equations 8 and 9) :

27  $0.75 * (1.26 - 0.623 * f_{CO_2} + 2.28 * f_{CHO})$  for O/C

1 0.91\* ( 1.07+1.07\*f<sub>CHO</sub>) for H:C

2 NOTE: f<sub>CO2</sub> and f<sub>CHO</sub> are calculated using the default organic fragmentation wave  
3 that does not include the H<sup>+</sup> fragment since inclusion of the H<sup>+</sup> fragment does not  
4 significantly affect the calculated organic mass.

5 Then:

6 
$$\text{moles of O} = z \times \left( \frac{f'_O}{mw_O} + \frac{f'_{OH}}{mw_{OH}} + \frac{1}{mw_{H_2O}} \right)$$

7 
$$\text{moles of H} = z \times \left( \frac{f'_{OH}}{mw_{OH}} + \frac{2}{mw_{H_2O}} + \frac{x}{mw_H} \right)$$

8 To simplify the equations, let  $\frac{f'_O}{mw_O} + \frac{f'_{OH}}{mw_{OH}} + \frac{1}{mw_{H_2O}} = a$  and  $\frac{f'_{OH}}{mw_{OH}} + \frac{2}{mw_{H_2O}} = b$

9 Then:

10 
$$\text{moles of O} = z \times a \quad \text{and} \quad \text{moles of H} = z \times \left( b + \frac{x}{mw_H} \right)$$

11 Keeping oxidation state constant when H<sub>2</sub>O is changed by y:

12 
$$\frac{2}{f_{cal}^{OC}} \times z a y - \frac{1}{f_{cal}^{HC}} z y \times \left( b + \frac{x}{mw_H} \right) = \frac{2}{f_{cal}^{OC}} \times z a - \frac{1}{f_{cal}^{HC}} z \times \left( b + \frac{x}{mw_H} \right)$$

13 Dividing by z and rearranging:

14 
$$\frac{2}{f_{cal}^{OC}} \times a (y - 1) = \frac{1}{f_{cal}^{HC}} \times \left( b + \frac{x}{mw_H} \right) (y - 1)$$

15 Dividing by (y-1) and rearranging:

16 
$$x = \left( \frac{2f_{cal}^{HC}}{f_{cal}^{OC}} \times a - b \right) \times mw_H$$

17

18 The calculated H<sup>+</sup>/H<sub>2</sub>O<sup>+</sup> ratio from the above equations can be directly

19 incorporated into the AMS analysis frag\_organic wave as shown in Table S3. The  
20 modified Frag\_organic wave refers to HWave, which is a new wave that contains time  
21 varying values of the H<sup>+</sup>/H<sub>2</sub>O<sup>+</sup> ratios over the time period being analyzed. For the  
22 Aiken-Ambient and Aiken Explicit methods, where the constant Aiken et al. (2008)

1 values are used for  $f_{cal}^{OC}$  and  $f_{cal}^{HC}$ , the HWave is constant over time at a value of 0.05.

2 For the Improved-Ambient method, the variations in  $f_{cal}^{OC}$  and  $f_{cal}^{HC}$  introduce a time  
3 variation in HWave.

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