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Characterization of gas-phase organics using proton transfer reaction time-of-flight mass spectrometry: fresh and aged residential wood combustion emissions

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Potential fragmentation

In most cases, possible reactions of species relevant to wood combustion to produce ions other than $[M+H]^+$ in the PTR-ToF mass spectrometer are negligible, however, these reactions can be more considerable in the following cases (the nominal $[M+H]^+$ *m/z* is given in parentheses and only literature results obtained under the most similar instrumental conditions are discussed):

- *Ethanol (m/z 47)*: Reactions to produce ions in addition to $[M+H]^+$ are highly dependent on instrumental conditions and do not necessarily correlate with *E*/*N*. At an *E*/*N* of 142 Td, Baasandorj et al. (2015) found that ~80 to >95% of the signal undergoes dehydration to form *m/z* 29, whereas Blake et al. (2006) observed only 7% of the signal at *m/z* 29 at a higher *E*/*N* of 165 Td and Buhr et al. (2002) observed ~25% of the signal at *m/z* 45 and ~4% at *m/z* 65 at a lower *E*/*N* of ~120 Td.

- *Isomers of acetic acid/glycolaldehyde (m/z 61)*: At an *E*/*N* of 142 Td, Baasandorj et al. (2015) found that ~30-50% of acetic acid and ~45-60% of glycolaldehyde undergoes dehydration to form $[C_2H_3O]^+$ at *m/z* 43. At an *E*/*N* of 132 Td, Haase et al. (2012) also found that ~30-50% of acetic acid fragments to *m/z* 43. Reported emission factors for $[C_2H_4O_2+H]^+$ are thus lower limits and could increase by up to ~30% (assuming $[C_2H_3O]^+$ originates entirely from $[C_2H_4O_2+H]^+$).

- *Isomers of (E)-pent-2-ene/(Z)-pent-2-ene/2-methylbut-1-ene/2-methylbut-2-ene/pent-1-ene/3-methylbut-1-ene (m/z 71)*: Gueneron et al. (2015) found 46-48% of the signal at *m/z* 43 and ~10% of the signal at *m/z* 41 at an *E/N* of 120 Td.

- *Butane-2,3-dione (m/z 87)*: At an *E*/*N* of ~120 Td, Buhr et al. (2002) found ~7% of the signal at *m/z* 59 and 5% at *m/z* 88, whereas Karl et al. (2007) found 34% of the signal at *m/z* 59 and the remainder at $[M+H]^+$ at an *E*/*N* of ~120 Td.

- *Ethyl benzene (m/z 107)*: At an *E*/*N* of 145 Td, Jobson et al. (2010) found that 40% of ethylbenzene fragments to interfere with the benzene signal at *m/z* 79. Using this fragmentation pattern in the current experiments, if the entire signal at $[C_8H_{10}+H]^+$ arises from ethylbenzene, this would result in a negligible overestimate of the benzene signal of less than 1% in experiments 1 and 3-5 and ~2% in experiment 2.

Table S1. Experimental parameters and primary emissions

exp.	wood burned per chamber volume (g m^{-3})	MCE	CO_2 (ppmv)	CO (ppmv)	CH_4 (ppmv)	NO_x (ppbv)	eBC (µg m^{-3})[b]	POA (µg m^{-3})[b]	OH exposure (molec cm^{-3} h)	OA (µg m^{-3})[b]
	primary								aged	
1	0.54231 (0.00007)	0.9763 (0.0002)	537.03 (0.07)	13.03 (0.02)	0.8353 (0.0004)	N/A	104.5 (0.5)	22.9 (0.3)	4.5×10^{7}	71
2	0.33360 (0.00007)	0.9776 (0.0003)	330.40 (0.07)	7.58 (0.02)	0.4754 (0.0004)	159 (2)	52 (1)	17.6 (0.4)	5.5×10^{7}	117
3	0.37052 (0.00006)	0.9767 (0.0002)	366.17 (0.06)	8.73 (0.02)	0.6384 (0.0004)	232 (2)	113.2 (0.7)	18.8 (0.3)	5.3×10^{7}	99
4	0.59413 (0.00009)	0.9737 (0.0002)	585.73 (0.09)	15.80 (0.02)	1.4352 (0.0005)	273 (2)	58.1 (0.3)	18.7 (0.3)	5.2×10^{7}	114
5	0.5726 (0.0007)	0.978 (0.002)	568.5 (0.7)	12.89 (0.02)	0.945 (0.001)	355 (2)	50.6 (0.4)	14.9 (0.2)	4.7×10^{7}	45

[a]Errors are given in parentheses and are ±1s calculated from the error propagation of the sample standard deviation of the measurements. Primary organic aerosol (POA) and organic aerosol (OA), are measured with an aerosol mass spectrometer, NMOGs are measured with a PTR-ToF-MS, equivalent black carbon (eBC) is measured with an Aethalometer and cavity ring-down spectroscopy is used to measure CO_2, CO and CH_4.
[b]Values are wall loss corrected and correspond to the given OH exposure.

Table S2. Applied reaction rates[a,b]

ion	species	reaction rate (10^{-9} cm^3 s^{-1})
$[CH_3OH+H]^+$	methanol	2.14
$[C_2H_3N+H]^+$	acetonitrile	3.82
$[C_2H_4O+H]^+$	acetaldehyde	3.02
$[C_3H_6+H]^+$	propene	1.62
$[CH_2O_2+H]^+$	formic acid	1.89
$[C_2H_6O+H]^+$	ethanol	2.04
$[C_4H_6+H]^+$	buta-1,3-diene	1.82
$[C_3H_4O+H]^+$	prop-2-enal	3.43
$[C_2H_2O_2+H]^+$	oxaldehyde	1.35
$[C_3H_6O+H]^+$	propan-2-one/propanal	3.10
$[C_2H_4O_2+H]^+$	acetic acid/glycolaldehyde	2.11
$[C_4H_4O+H]^+$	furan	1.69
$[C_5H_8+H]^+$	isoprene/cyclopentene	1.88
$[C_4H_6O+H]^+$	(*E*)-but-2-enal	3.45
	3-buten-2-one	
	2-methylprop-2-enal	
$[C_5H_{10}+H]^+$	(*E*)-/(*Z*)-pent-2-ene	1.91
	2-methylbut-1-ene	
	2-methylbut-2-ene	
	pent-1-ene	
	3-methylbut-1-ene	
$[C_4H_8O+H]^+$	butan-2-one	3.09
	butanal	
	2-methylpropanal	
$[C_3H_6O_2+H]^+$	methyl acetate	2.24
$[C_6H_6+H]^+$	benzene	1.93
$[C_6H_{12}+H]^+$	(*E*)-hex-2-ene	2.02
	2-methyl-pent-2-ene	
$[C_4H_6O_2+H]^+$	butane-2,3-dione	1.70
$[C_4H_8O_2+H]^+$	ethyl acetate	4.15
$[C_7H_8+H]^+$	toluene	2.08
$[C_6H_6O+H]^+$	phenol	2.13
$[C_5H_4O_2+H]^+$	2-furaldehyde	3.88
$[C_7H_{14}+H]^+$	methylcyclohexane/hept-1-ene	2.13
$[C_6H_{12}O+H]^+$	hexanal	3.15
$[C_8H_8+H]^+$	styrene	2.27
$[C_7H_6O+H]^+$	benzaldehyde	3.63
$[C_8H_{10}+H]^+$	*m*-/*o*-/*p*-xylene	2.26
	ethylbenzene	
$[C_7H_8O+H]^+$	*m*-/*o*-/*p*-cresol	2.27
$[C_7H_{14}O+H]^+$	heptanal	2.74
$[C_9H_8+H]^+$	1*H*-indene	2.42
$[C_8H_8O+H]^+$	1-phenylethanone	3.23
	3-/4-methylbenzaldehyde	
$[C_9H_{12}+H]^+$	*i*-propylbenzene	2.39
	n-propylbenzene	
	1,3,5-trimethylbenzene	
$[C_{10}H_8+H]^+$	naphthalene	2.45
$[C_{11}H_{10}+H]^+$	1-/2-methylnaphthalene	2.71
$[C_{12}H_8+H]^+$	acenaphthylene	2.86
$[C_{12}H_{10}+H]^+$	1,1'-biphenyl	2.81
	1,2-dihydroacenaphthylene	
$[C_{13}H_{10}+H]^+$	fluorene	2.88
$[C_{14}H_{10}+H]^+$	phenanthrene	2.97
	anthracene	
$[C_{16}H_{10}+H]^+$	fluoranthene	3.37
	pyrene	
	acephenanthrylene	

[a]Reaction rates taken from Cappellin et al. (2012) at a drift temperature of 90 °C and an *E*/*N* of 140 Td. A reaction rate of 2×10^{-9} cm^3 s^{-1} is applied to all other ions.

[b]In the cases of isomers, an average of the available rate constants is applied.

Table S3. Aged gas-phase emissions (mg kg^{-1}) measured using PTR-ToF-MS at an OH exposure of (4.5-5.5)×10^7 molec cm^{-3} h[a]

species	monoisotopic *m/z*	structural assignment[b]	functional group	experiment 1	2	3	4	5	average[c]
NMOG				2600	9100	7100	2600	1600	5000±3000
acid				990	4600	3500	960	570	2000±2000
O-containing				680	2600	1900	620	390	1000±1000
carbonyl				210	730	490	170	130	300±300
oxygenated aromatic				34	56	55	35	26	40±10
alcohol				140	590	350	100	65	200±200
furan				24	72	51	20	14	40±20
O- and N-containing				120	87	110	120	100	100±20
C_xH_y				59	69	83	61	42	60±10
aromatic hydrocarbon				180	81	270	380	140	200±100
N-containing				31	46	42	28	24	34±9
other				180	260	280	180	140	210±60
$[CH_3OH+H]^+$	33.034	methanol	alcohol	110	590	340	92	56	200±200
$[C_2H_3N+H]^+$	42.034	acetonitrile	N-containing	3.6	3.6	4.4	3.3	2.7	3.5±0.6
$[C_3H_6+H]^+$	43.055	propene	C_xH_y	7.7	17	13	6.4	5.2	10±5
$[C_2H_4O+H]^+$	45.034	acetaldehyde	carbonyl	50	130	94	42	30	70±40
$[CH_2O_2+H]+$	47.013	formic acid	acid	280	580	570	310	190	400±200
$[C_2H_6O+H]^+$	47.050	ethanol	alcohol	33	1.1	6.8	8.9	9.1	10±10
$[C_4H_6+H]^+$	55.055	buta-1,3-diene	C_xH_y	6.4	8.0	9.7	5.9	7.5	7±2
$[C_3H_4O+H]^+$	57.034	prop-2-enal	carbonyl	6.9	39	27	7.4	1.8	20±20
$[C_2H_2O_2+H]^+$	59.013	oxaldehyde	carbonyl	BDL	BDL	BDL	BDL	BDL	BDL
$[C_3H_6O+H]^+$	59.050	propan-2-one propanal	carbonyl	64	170	120	46	48	90±50
$[C_2H_4O_2+H]^+$	61.029	acetic acid glycolaldehyde	acid	710	4000	2900	650	370	2000±2000
$[C_4H_4O+H]^+$	69.034	furan	furan	4.1	16	11	3.9	2.3	8±6
$[C_5H_8+H]^+$	69.070	isoprene cyclopentene	C_xH_y	3.3	4.4	4.5	1.6	1.6	3±1
$[C_4H_6O+H]^+$	71.050	(*E*)-but-2-enal 3-buten-2-one 2-methylprop-2-enal	carbonyl	4.3	16	11	3.7	2.1	7±6
$[C_5H_{10}+H]^+$	71.086	(*E*)-/(*Z*)-pent-2-ene 2-methylbut-1-ene 2-methylbut-2-ene pent-1-ene 3-methylbut-1-ene	C_xH_y	BDL	BDL	1.1	BDL	BDL	0.2±0.5
$[C_3H_4O_2+H]^+$	73.029	2-oxopropanal	carbonyl	18	60	48	17	11	30±20
$[C_4H_8O+H]^+$	73.065	butan-2-one butanal 2-methylpropanal	carbonyl	9.7	39	24	6.7	7.0	20±10
$[C_3H_6O_2+H]^+$	75.045	methyl acetate	O-containing	59	350	220	46	30	100±100
$[C_6H_6+H]^+$	79.055	benzene	aromatic hydrocarbon	170	65	240	350	130	200±100
$[C_5H_6O+H]^+$	83.050	2-methylfuran	furan	2.1	9.0	5.9	2.1	1.4	4±3
$[C_5H_8O+H]^+$	85.065	3-methyl-3-buten-2-one	carbonyl	3.0	14	8.9	2.7	2.1	6±5
$[C_6H_{12}+H]^+$	85.102	(*E*)-hex-2-ene 2-methyl-pent-2-ene	C_xH_y	BDL	BDL	BDL	BDL	BDL	BDL
$[C_4H_6O_2+H]^+$	87.045	butane-2,3-dione	carbonyl	48	250	150	39	24	100±100
$[C_7H_8+H]^+$	93.070	toluene	aromatic hydrocarbon	11	7.4	15	17	6.6	11±5
$[C_6H_6O+H]^+$	95.050	phenol	oxygenated aromatic	2.3	3.5	6.4	6.1	4.0	4±2
$[C_5H_4O_2+H]^+$	97.029	furan-2-carbaldehyde	furan	11	23	19	8.4	5.8	13±7
$[C_6H_8O+H]^+$	97.065	2,4-/2,5-dimethylfuran	furan	0.99	3.5	2.6	1.2	0.82	2±1
$[C_4H_2O_3+H]^+$	99.008	maleic anhydride[d]	O-containing	110	260	200	92	74	140±80
$[C_8H_8+H]^+$	105.070	styrene	aromatic hydrocarbon	0.88	1.5	1.5	1.3	0.61	1.2±0.4
$[C_7H_6O+H]^+$	107.050	benzaldehyde	oxygenated aromatic	5.8	4.0	6.6	7.4	4.0	6±2
$[C_8H_{10}+H]^+$	107.086	*m*-/*o*-/*p*-xylene ethylbenzene	aromatic hydrocarbon	0.95	BDL	1.5	1.5	BDL	0.8±0.8
$[C_7H_8O+H]^+$	109.065	*m*-/*o*-/*p*-cresol	oxygenated aromatic	BDL	0.96	1.1	0.94	0.48	0.7±0.4
$[C_6H_6O_2+H]^+$	111.045	*m*-/*o*-/*p*-benzenediol 2-methylfuraldehyde	oxygenated aromatic	6.8	18	13	5.0	3.8	9±6
$[C_9H_8+H]^+$	117.070	1*H*-indene	aromatic hydrocarbon	BDL	BDL	0.80	0.79	BDL	0.3±0.4
$[C_9H_{10}+H]^+$	119.086	2,3-dihydro-1*H*-indene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	BDL	BDL

$[C_8H_8O+H]^+$	121.065	1-phenylethanone 3-/4-methylbenzaldehyde	oxygenated aromatic	3.0	2.8	3.3	2.7	1.8	2.7±0.6
$[C_9H_{12}+H]^+$	121.102	*i*-propylbenzene *n*-propylbenzene 1,3,5-trimethylbenzene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	BDL	BDL
$[C_8H_{10}O+H]^+$	123.081	2,4-/2,6-/3,5-dimethylphenol	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_7H_8O_2+H]^+$	125.060	2-methoxyphenol methylbenzenediols	oxygenated aromatic	BDL	5.5	3.7	0.94	BDL	2±2
$[C_6H_6O_3+H]^+$	127.040	5-(hydroxymethyl)furan-2-carbaldehyde	furan	6.1	21	13	4.4	3.4	9±7
$[C_{10}H_8+H]^+$	129.070	naphthalene	aromatic hydrocarbon	BDL	BDL	4.6	4.6	0.84	2±2
$[C_8H_{10}O_2+H]^+$	139.076	2-methoxy-4-methylphenol 4-(2-hydroxyethyl)phenol	oxygenated aromatic	BDL	2.9	2.1	0.71	BDL	1±1
$[C_{11}H_{10}+H]^+$	143.086	1-/2-methylnaphthalene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	0.4	0.07±0.2
$[C_9H_6O_2+H]^+$	147.045	2,3-dihydroinden-1-one	oxygenated aromatic	3.8	3.6	4.8	3.8	2.5	3.7±0.8
$[C_8H_4O_3+H]^+$	149.024	phthalic anhydride[d]	O-containing	55	71	74	59	55	63±9
$[C_8H_8O_3+H]^+$	153.055	4-hydroxy-3-methoxybenzaldehyde	oxygenated aromatic	1.2	3.3	2.6	1.4	BDL	2±1
$[C_{12}H_8+H]^+$	153.070	acenaphthylene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	BDL	BDL
$[C_9H_{12}O_2+H]^+$	153.092	4-ethyl-2-methoxyphenol 1,2-dimethoxy-4-methylbenzene	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_8H_{10}O_3+H]^+$	155.071	2,6-dimethoxyphenol	oxygenated aromatic	BDL	BDL	BDL	BDL	0.5	0.1±0.2
$[C_{12}H_{10}+H]^+$	155.086	1,1'-biphenyl 1,2-dihydroacenaphthylene	aromatic hydrocarbon	BDL	BDL	BDL	1.2	BDL	0.2±0.5
$[C_{12}H_{12}+H]^+$	157.102	dimethylnaphthalene	aromatic hydrocarbon	BDL	1.7	1.4	BDL	BDL	0.6±0.9
$[C_{10}H_{12}O_2+H]^+$	165.092	2-methoxy-4-[(*E*)-prop-1-enyl]phenol 2-methoxy-4-prop-2-enylphenol 2-methoxy-4-[(*Z*)-prop-1-enyl]phenol	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_9H_{10}O_3+H]^+$	167.071	1-(4-hydroxy-3-methoxyphenyl)ethanone 2,5-dimethylbenzaldehyde 3,4-dimethoxybenzaldehyde	oxygenated aromatic	1.4	1.9	1.9	0.96	BDL	1.2±0.8
$[C_{13}H_{10}+H]^+$	167.086	fluorene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	BDL	BDL
$[C_{10}H_{14}O_2+H]^+$)	167.107	2-methoxy-4-propylphenol	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_9H_{12}O_3+H]^+$	169.086	2,6-dimethoxy-4-methylphenol	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_{14}H_{10}+H]^+$	179.086	phenanthrene anthracene	aromatic hydrocarbon	2.8	2.1	1.9	1.6	3.9	2.5±0.9
$[C_{13}H_8O+H]^+$	181.065	fluoren-9-one phenalen-1-one	oxygenated aromatic	4.2	4.1	3.7	2.5	4.2	3.7±0.7
$[C_{10}H_{12}O_3+H]^+$	181.086	1-(4-hydroxy-3-methoxyphenyl)propan-2-one	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_9H_{10}O_4+H]^+$	183.066	3,4-dimethoxybenzoic acid 4-hydroxy-3,5-dimethoxybenzaldehyde	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_{10}H_{14}O_3+H]^+$	183.102	4-ethyl-2,6-dimethoxyphenol	oxygenated aromatic	BDL	BDL	BDL	BDL	BDL	BDL
$[C_{15}H_{12}+H]^+$	193.102	1-/2-/3-/9-methylphenanthrene 2-methylanthracene	aromatic hydrocarbon	0.71	BDL	0.60	BDL	0.77	0.4±0.4
$[C_{11}H_{14}O_3+H]^+$	195.102	1,3-dimethoxy-2-prop-2-enoxybenzene 2,6-dimethoxy-4-[(*Z*)-prop-1-enyl]phenol	oxygenated aromatic	BDL	BDL	BDL	BDL	0.43	0.09±0.2
$[C_{16}H_{10}+H]^+$	203.086	fluoranthene pyrene acephenanthrylene	aromatic hydrocarbon	BDL	BDL	BDL	BDL	0.45	0.09±0.2

[a]BDL indicates value is below the detection limit, the determination of which is described in the text.

[b]Multiple structural assignments for a given ion correspond to possible isomers.

[c]Uncertainties correspond to one sample standard deviation of the replicates.

[d]Structural assignment based on known products produced during oxidation of aromatics (Bandow et al., 1985; Chan et al., 2009; Praplan et al., 2014).

Figure S1. Photograph of residential wood burner used for all burning. The stove is not fitted with a catalyst or other emission control device.

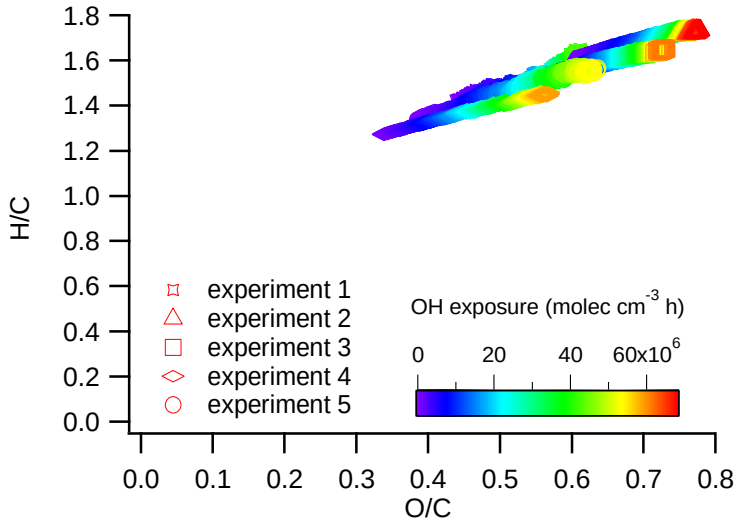

Figure S2. Elemental composition of the speciated NMOG bulk as a function of OH exposure for all experiments.

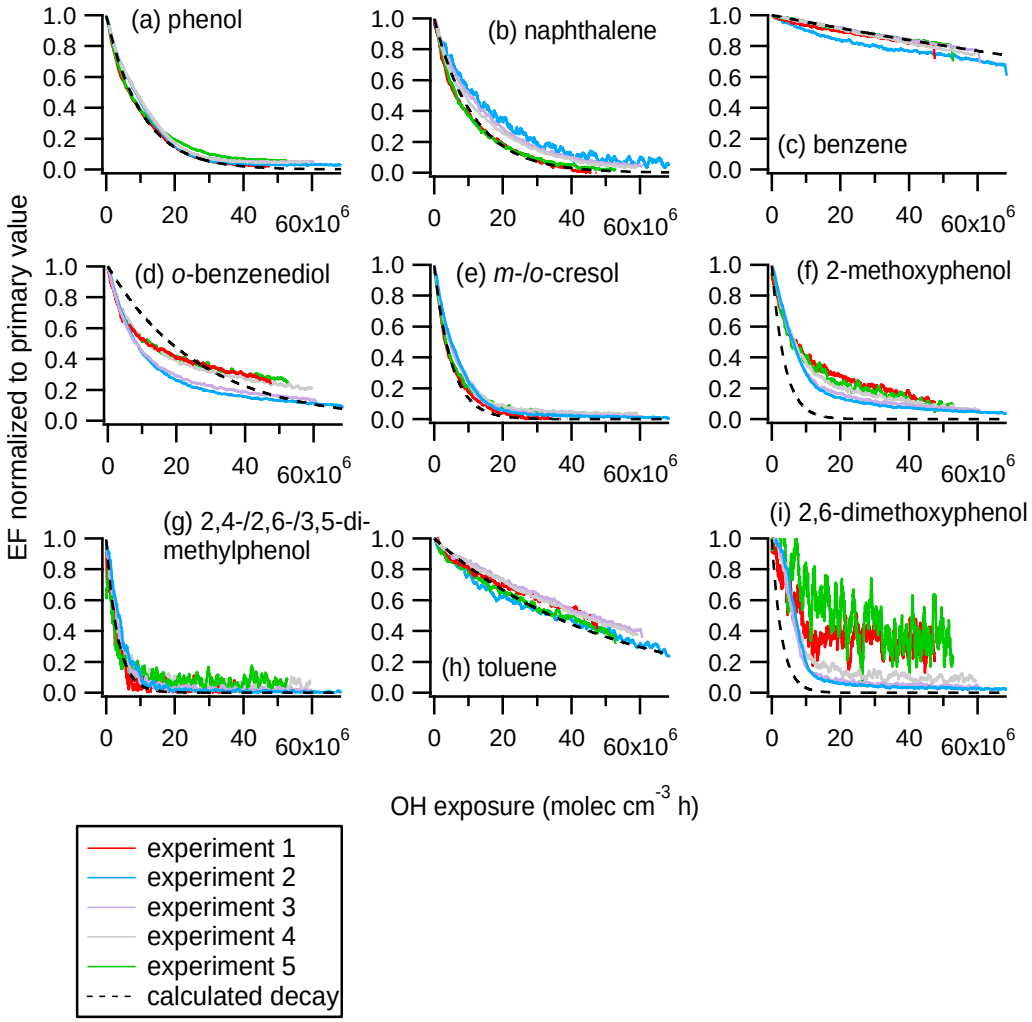

Figure S3. (a-i) Behavior with aging of individual compounds contributing the most to SOA formation for experiments 1-5 (5 min moving average). The black, dashed line in each panel is the expected decay rate of each species based on the OH concentration and reaction rate with OH.

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