S1 Combustion conditions and primary emission
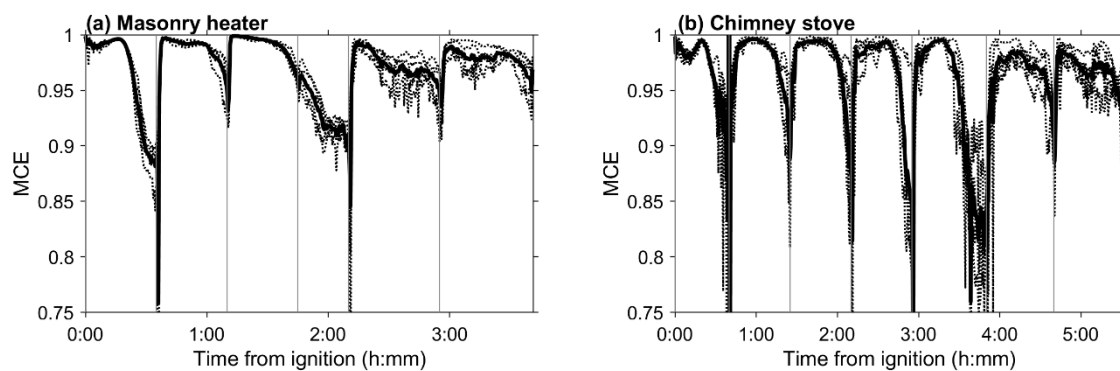

Figure S1: Modified combustion efficiency (MCE = ΔCO_2 / (ΔCO_2 + ΔCO)) during experiments. Averages over all experiments are shown with the solid lines and different experiments are shown with the dotted lines.

5 Table S1: Compounds measured by FTIR and their reaction rates with OH (k_{OH})utilized for OHR_{ext} calculations.

Compound	Formula	VOC group	CAS	Range	unit	k_{OH} [cm^3/molec. s]
Water vapor	H_2O		7732-18-5	30	vol-%	2.10E-16
Carbon dioxide	CO_2		124-38-9	25	vol-%	
Carbon monoxide	CO		630-08-0	5000	ppm	2.41E-13
				2	vol-%	
Nitrous oxide	N_2O		10024-97-2	200	ppm	2.01E-16
Nitrogen monoxide	NO		10102-43-9	1000	ppm	1.00E-11
Nitrogen dioxide	NO_2		19192-44-0	200	ppm	1.06E-11
Sulfur dioxide	SO_2		7446-09-5	1000	ppm	1.31E-12
Carbonyl sulfide	COS		483-58-1	100	ppm	1.96E-15
Ammonia	NH_3		7664-41-7	500	ppm	1.57E-13
Hydrogen chloride	HCl		7647-01-0	200	ppm	7.86E-13
Hydrogen cyanide	HCN		74-90-8	100	ppm	3.13E-14
Hydrogen fluoride	HF		7664-39-3	100	ppm	
Methane	CH_4		74-82-8	1000	ppm	6.28E-15
Ethane	C_2H_6	Alkane	74-84-0	200	ppm	2.41E-13
Propane	C_3H_8	Alkane	74-98-6	200	ppm	1.10E-12
Butane	C_4H_{10}	Alkane	106-97-8	200	ppm	2.33E-12
Pentane	C_5H_{12}	Alkane	109-66-0	200	ppm	3.80E-12
Hexane	C_6H_{14}	Alkane	110-54-3	200	ppm	5.20E-12
Heptane	C_7H_{16}	Alkane	142-82-5	200	ppm	6.76E-12
Octane	C_8H_{18}	Alkane	111-65-9	200	ppm	7.43E-12
Acetylene	C_2H_2	Unsaturated	75-86-2	200	ppm	7.75E-13
Ethylene	C_2H_4	Unsaturated	74-85-1	200	ppm	8.51E-12
Propene	C_3H_6	Unsaturated	115-07-1	200	ppm	3.01E-11
1,3-Butadiene	C_4H_6	Unsaturated	106-99-0	200	ppm	6.93E-11
Benzene	C_6H_6	Aromatic	71-43-2	200	ppm	1.28E-12
Toluene	C_7H_8	Aromatic	108-88-3	200	ppm	6.16E-12
m-Xylene	C_8H_{10}	Aromatic	108-38-3	200	ppm	2.45E-11
o-Xylene	C_8H_{10}	Aromatic	95-47-6	200	ppm	1.47E-11
p-Xylene	C_8H_{10}	Aromatic	106-42-3	200	ppm	1.52E-11
1,2,3-Trimethylbenzene	C_9H_{12}	Aromatic	526-73-8	200	ppm	2.89E-11
1,2,4-Trimethylbenzene	C_9H_{12}	Aromatic	95-63-6	200	ppm	3.16E-11
1,35-Trimethylbenzene	C_9H_{12}	Aromatic	106-42-3	200	ppm	5.98E-11
Formic acid	CH_2O	Oxygenated	64-18-6	200	ppm	4.50E-13
Acetic acid	$C_2H_4O_2$	Oxygenated	64-19-7	200	ppm	6.69E-13
Formaldehyde	CHOH	Oxygenated	50-0-0	200	ppm	9.38E-12
Acetaldehyde	C_2H_4O	Oxygenated	75-07-0	200	ppm	1.63E-11
Methanol	CH_4O	Oxygenated	67-56-1	500	ppm	9.28E-13
Ethanol	C_2H_6O	Oxygenated	64-17-5	500	ppm	3.24E-12
Propanol	C_3H_8O	Oxygenated	71-23-8	500	ppm	5.82E-12

Table S2: Average (± standard deviation) concentrations of gaseous emissions measured by FTIR for each experiment type (five experiments each).

Compound	unit	Masonry heater		Chimney stove		p* Masonry heater; S-5% v. S-22%	p* Chimney stove; B-17% v. S-22%	p** S-22%; Masonry heater v. Chimney stove
		S-5%	S-22%	B-17%	S-22%			
Water vapor	vol-%	4.5 ± 0.3	4.1 ± 0.4	7.2 ± 0.4	6.4 ± 0.5	<0.01	<0.01	<0.01
Carbon dioxide	vol-%	4.6 ± 0.1	3.26 ± 0.08	6.2 ± 0.3	5.9 ± 0.4	<0.01	0.05	<0.01
Carbon monoxide	ppm	638 ± 26.5	735 ± 129	1680 ± 249	1880 ± 144	0.24	0.28	<0.01
Nitrogen monoxide	ppm	25.6 ± 1.8	21.4 ± 2.6	45.9 ± 2.9	31.6 ± 3.7	<0.01	<0.01	<0.01
Nitrogen dioxide	ppm	1.0 ± 0.4	2.7 ± 1.1	1.3 ± 0.7	2.6 ± 1.6	0.03	0.08	0.98
Sulfur dioxide	ppm	0.5 ± 0.3	0.6 ± 0.3	1.4 ± 0.3	0.9 ± 0.1	0.05	<0.01	0.1
Ammonia	ppm	1 ± 0.3	1.9 ± 0.3	6.2 ± 0.4	5.1 ± 0.5	<0.01	0.01	<0.01
Methane	mgC m^{-3}	10.5 ± 0.5	23.6 ± 5.6	28.7 ± 4.5	46.3 ± 4.4	<0.01	0.01	<0.01
Non-methane VOCs	mgC m^{-3}	42.6 ± 9.2	101 ± 24.6	89.4 ± 11.3	148.3 ± 23	0.01	<0.01	0.05
Alkanes	mgC m^{-3}	9.7 ± 2.6	23.8 ± 6.6	16.9 ± 3	29.5 ± 6.7	<0.01	0.01	0.31
Unsaturated	mgC m^{-3}	12.8 ± 2.9	33.1 ± 8.1	30.8 ± 4.7	54.2 ± 5.4	<0.01	<0.01	<0.01
Aromatics	mgC m^{-3}	14.2 ± 3.5	20.7 ± 2.6	27.9 ± 4.9	36.0 ± 3.4	0.02	0.02	<0.01
Oxygenated	mgC m^{-3}	5.6 ± 1.2	21.9 ± 8.1	13.3 ± 0.6	27.1 ± 8.9	0.01	0.04	0.51

* paired Student's t-test; comparison of averages of the consecutive experiments; ** unpaired Student's t-test of independent experiments
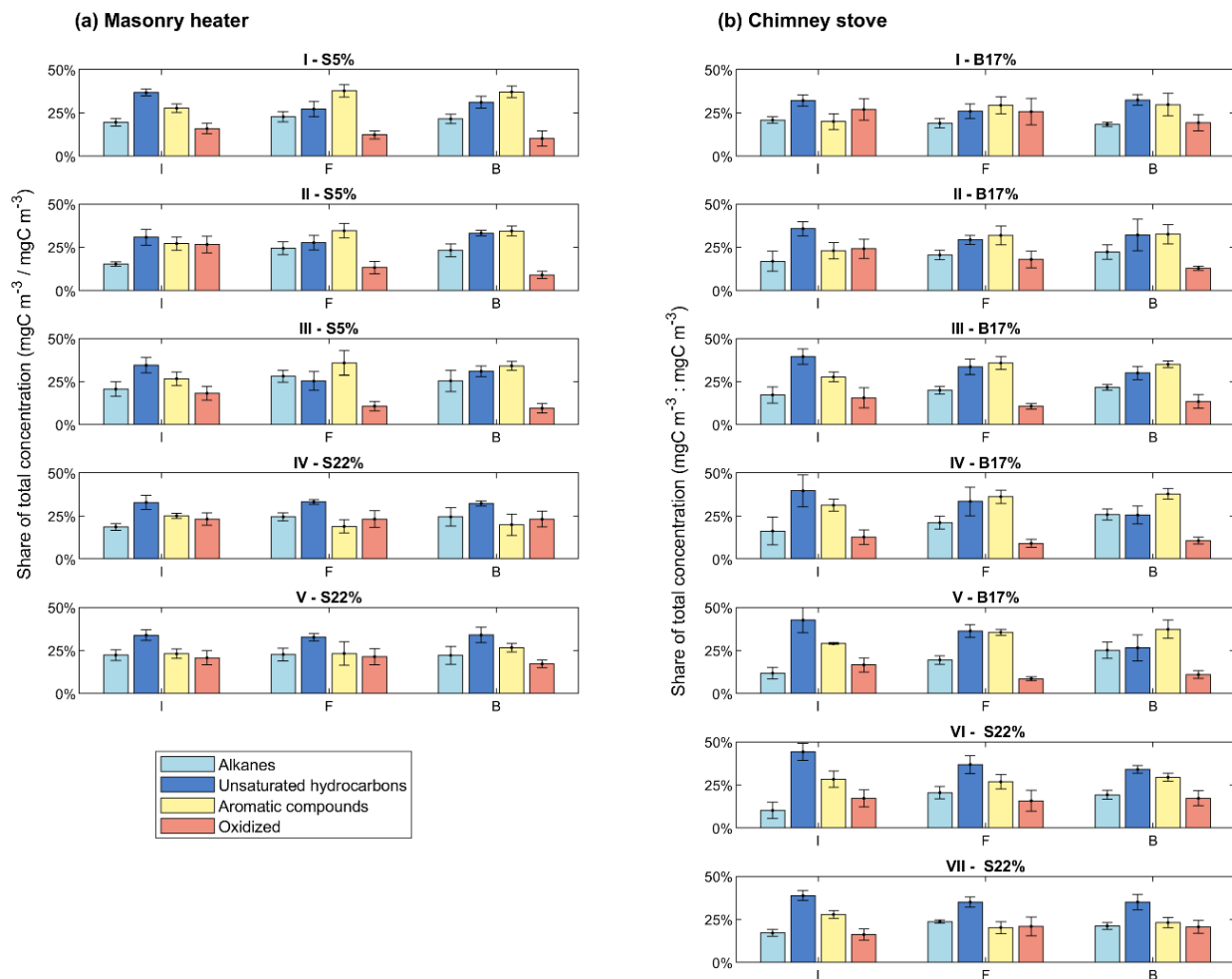
(a) Masonry heater
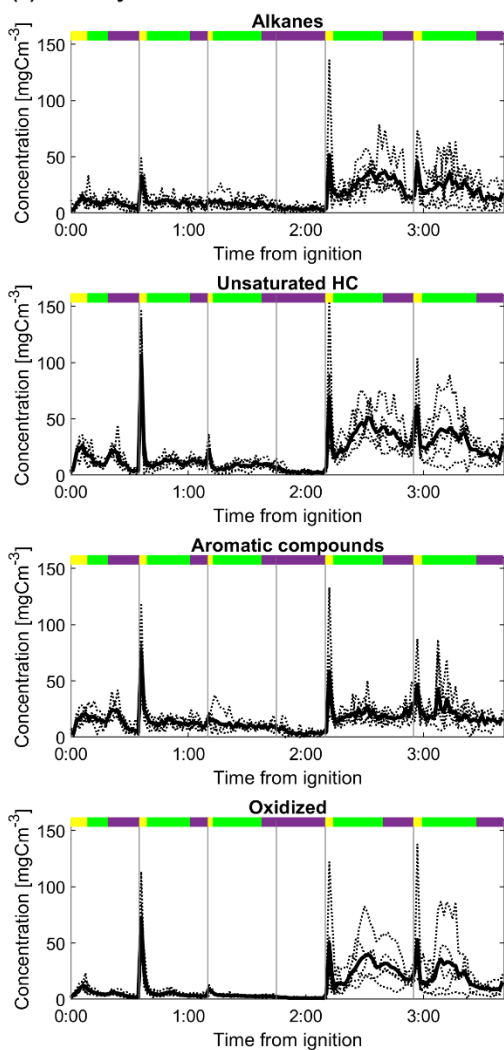
(b) Chimney stove
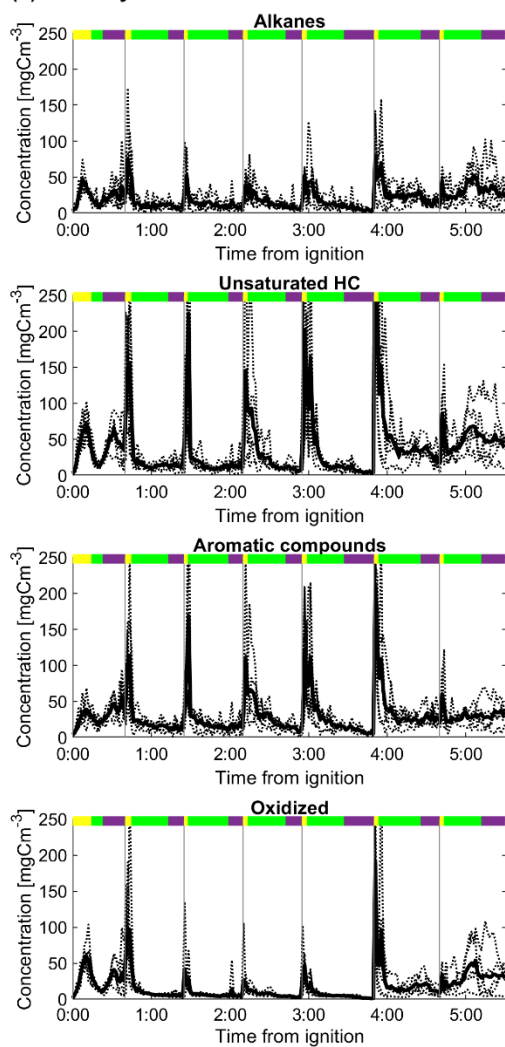
10

Figure S2: Timeseries of VOC emissions from (a) masonry heater and (b) chimney stove for the four VOC groups. Average lengths of the combustion phases are marked on the top panel with the yellow (ignition), green (flaming), and purple (burnout).

(a) Masonry heater

(b) Chimney stove

15   **Figure S3: Average shares of the VOC groups at different combustion phases (I = ignition, F = flaming, B = burnout) for each batch (I-V for masonry heater, I-VII for chimney stove) as a relation to the total VOC concentration (mgC m⁻³ / mgC m⁻³) measured by FTIR from the fresh exhaust. Error bars denote standard deviations.**

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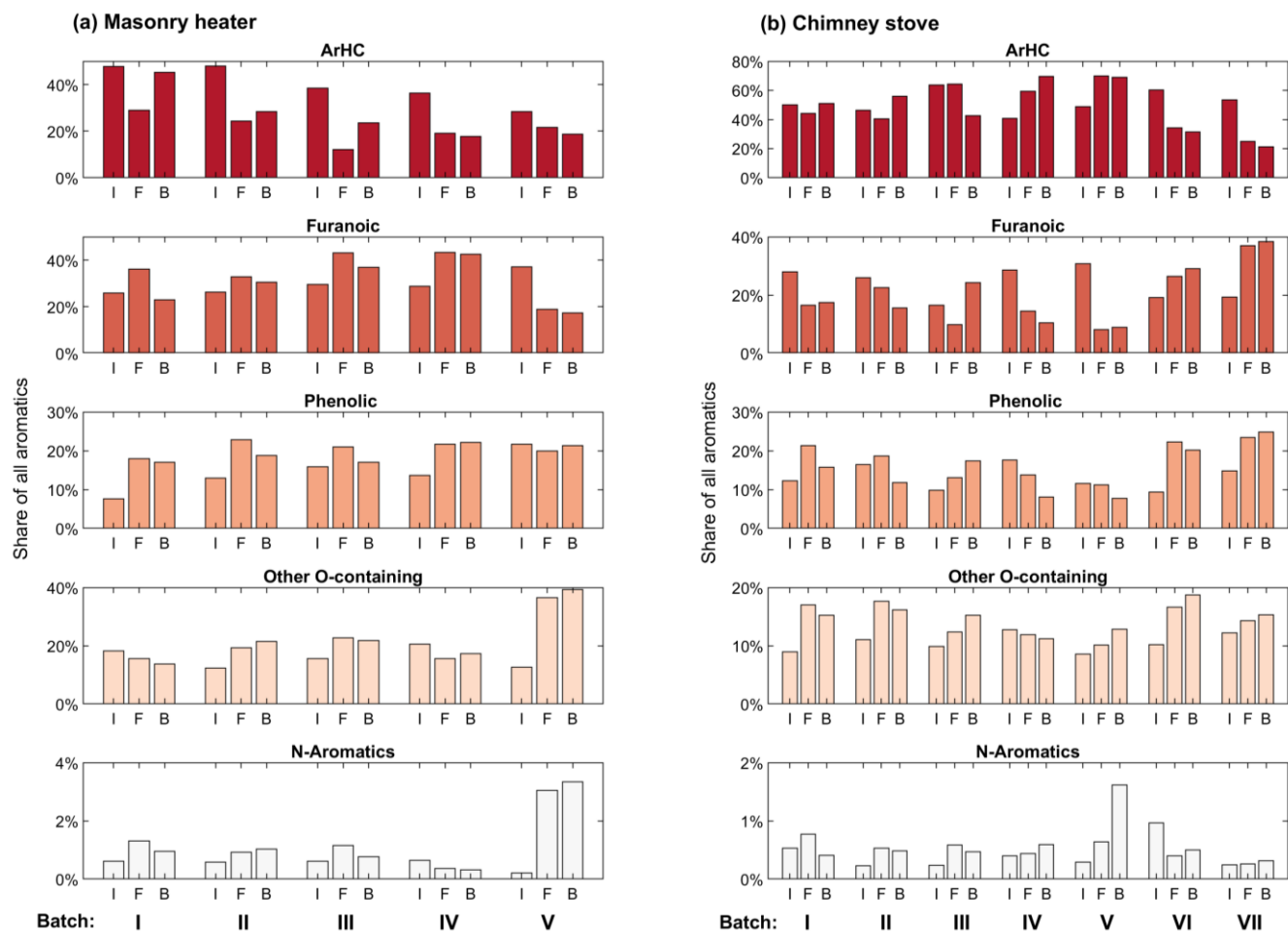
Figure S4: Shares of aromatic compound groups measured by the PTR-ToF-MS from unaged aerosol, in relation to the total aromatic concentration (ppm/ppm) during different combustion phases (I = ignition, F = flaming, B = burnout) for each batch.
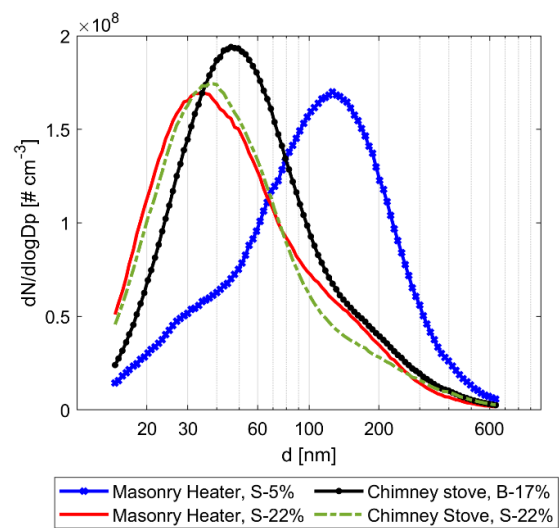
Figure S5: Average size distributions in the primary exhaust (dry, 13 % O₂) measured by SMPS.

S2 Conditions in PEAR

S2.1 External OH reactivity

External OH reactivity (OHR$_{ext}$ = Σ k$_i$*c$_i$) was calculated from the compounds measured by FTIR from the primary exhaust, using their reaction rate constants with OH (k$_i$; Table S1) and dilution corrected concentrations (c$_i$) in the PEAR.
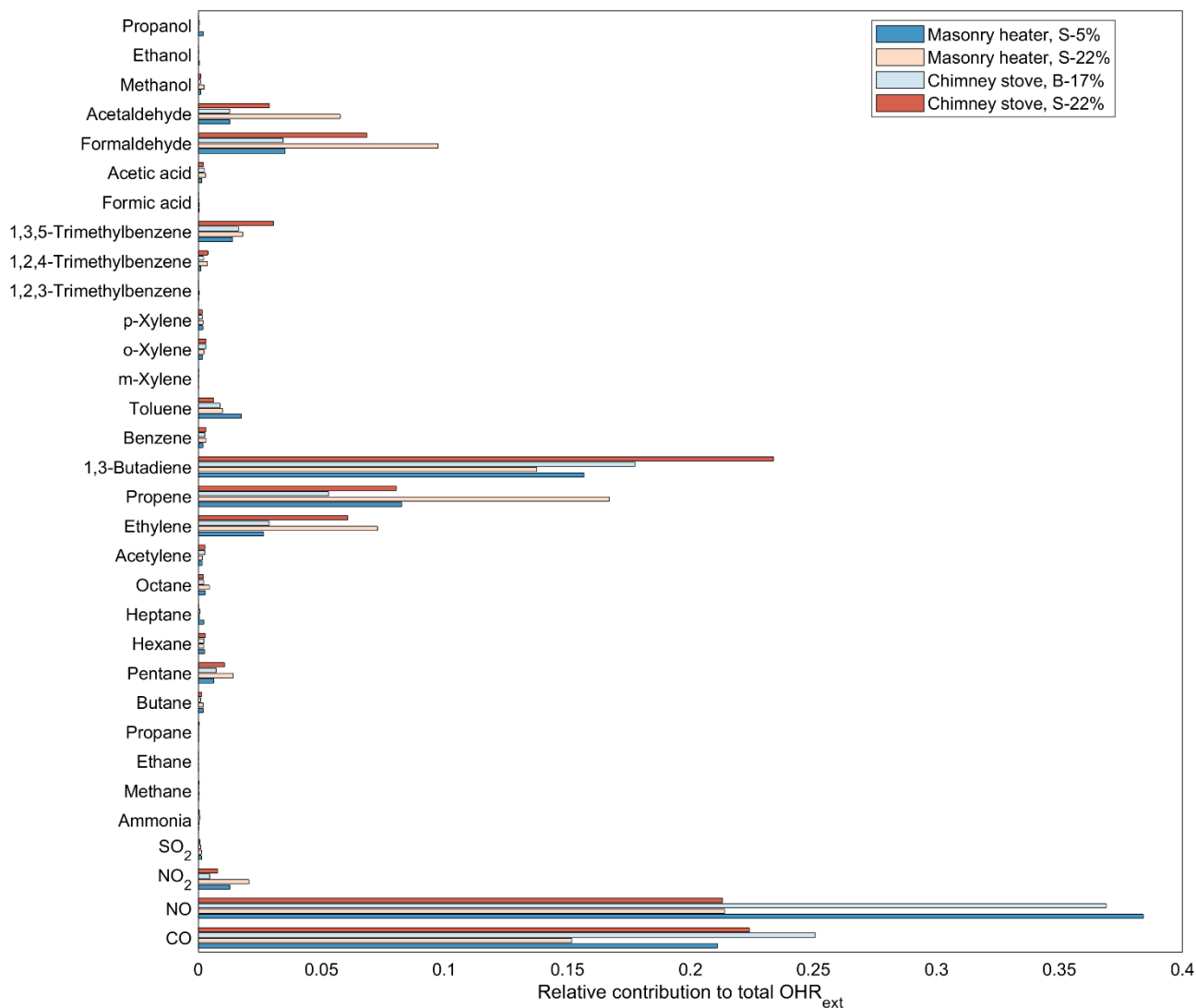

Figure S6: Average relative contributions of the measured gaseous compounds to the total OHR$_{ext}$.
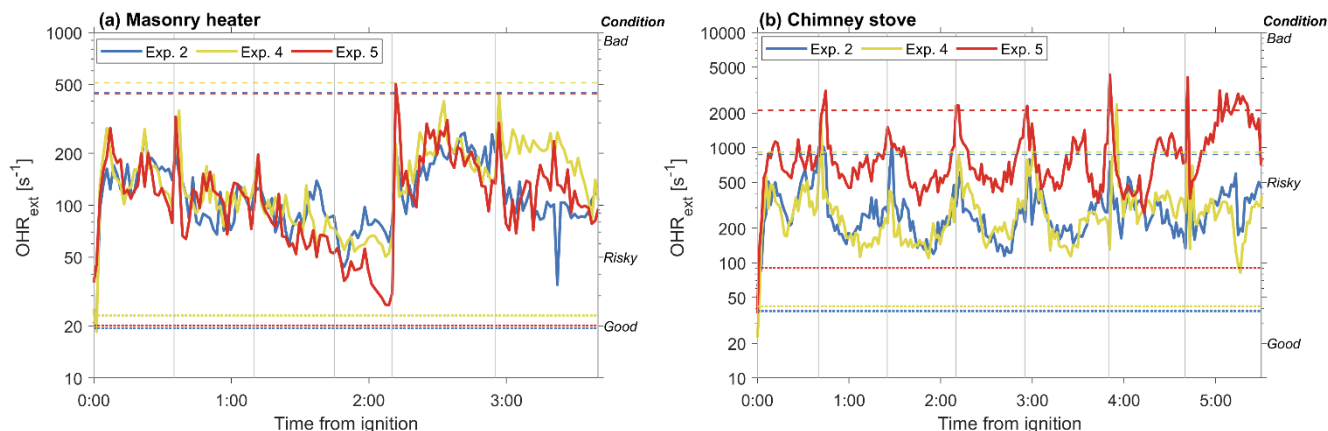
Figure S7: External OH reactivity in the PEAR for the compounds measured by FTIR during photochemical aging experiments. Risk levels for OHR$_{ext}$, determined by Peng and Jimenez (2017), are marked with dotted (good/risky: $\frac{Flux_{254nm}\ exposure}{OH\ exposure} > 4E5\ cm\ s^{-1}$) and dashed (risky/bad: $\frac{Flux_{254nm}\ exposure}{OH\ exposure} > 1E7\ cm\ s^{-1}$) lines for each experiment.

S2.2 Fate of low-volatility organic compounds in the PEAR

Estimations of the LVOC fates in the PEAR were done based on Palm et al. (2016). Three possible pathways for LVOC depletion in the PEAR were considered: condensation onto particles, reactions with OH, and condensation onto walls of the PEAR. The rest exit the PEAR in the gas-phase after which they are expected to condense on the walls of sampling lines.

The LVOC lifetime due to aerosol losses (τ_{aer}) was calculated using Eq. (S1):

$$\tau_{aer} = \frac{1}{CS} \tag{S1}$$

where condensation sink (CS, s^{-1}; Fig 4) was calculated according to Lehtinen et al. (2003), using an average of the particle size distributions before and after the PEAR. The diffusion coefficient in use (D; 7x10^{-6} m^2 s^{-1}) was chosen as representative for oxidised, ~200 g mol^{-1} molecules (Tang et al., 2015).

LVOC lifetime due to wall losses (τ_{wall}) was calculated with Eq. (S2):

$$\tau_{wall} = \left(\frac{A}{V} * \frac{2}{\pi} \sqrt{k_e D} \right)^{-1} \tag{S2}$$

τ_{wall} depends on the area-to-wall ratio ($\frac{A}{V}$), which in the PEAR is 2.28 m^2:0.139 m^3 = 16.4 m^{-1}. The applied eddy diffusion coefficient, k$_e$, was 0.0036 s^{-1}, similarly to Palm et al. (2016). τ_{wall} was constant (603 s) for all of the PEAR experiments.

Lifetime for LVOCs due to reactions with OH radicals was calculated with Eq. (S3):

$$\tau_{OH} = \frac{5}{k_{OH} * [OH]} \tag{S3}$$

based on the estimations that LVOCs as a total have an OH reaction rate (k$_{OH}$) of 1×10^{-11} cm^3 molec.$^{-1}$ s^{-1} and that it takes five such reactions for LVOC fragmentation into higher-volatility compounds.

The resulting total lifetime was calculated with Eq. (S4):

$$\tau_{total} = \left(\frac{1}{\tau_{aer}} + \frac{1}{\tau_{wall}} + \frac{1}{\tau_{OH}}\right)^{-1} \tag{S4}$$

from which the fraction of each pathway (F_x, where footnote denotes the degradation pathway: aerosol, wall, or OH) to the total loss was derived with eq. (S5).

$$F_x = (1 - F_{exit}) * \frac{\tau_x^{-1}}{\tau_{total}^{-1}} \tag{S5}$$

The fraction of LVOCs exiting the PEAR in gas-phase (F_{exit}) was by eq. (S6):

$$F_{exit} = e^{-\frac{RT}{\tau_{total}^{-1}}} \tag{S6}$$

with the residence time (RT) being 139s for the 60lpm flow rate.

S2.3 Photochemical exposure in PEAR

OH exposure during photochemical aging was measured directly from the decay of added butanol-*d9*, similarly to Barmet et al. (2012). Timeseries for the OH exposures during each experiment are shown in Figure S8.
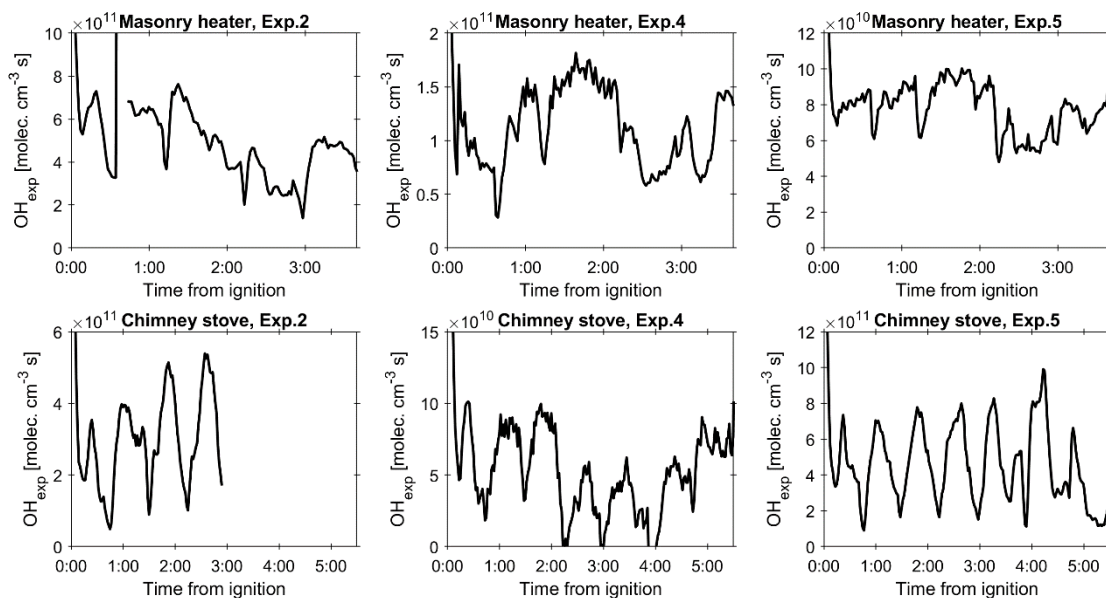
Figure 8: OH exposure during photochemical aging experiments.

The relevance of other reaction pathways in the PEAR, including photolysis as well as exposure to ozone, excited singlet oxygen (O(^{1}D)), and triplet atomic oxygen (O(^{3}P)), was estimated based on the estimation equations by Peng et al. (2016) (Table S3). The conditions in the PEAR were assessed utilising the directly measured OH exposure (OH$_{exp}$), initial O$_3$ input concentration (O$_{3, init}$), residence time in PEAR (RT; 139s), and water mixing ratio (H$_2$O; 0.015 for 45 % relative humidity). Average exposures during each experiment are shown in Table S4, and their relevance towards ambient atmospheric conditions are displayed in Figure S11.

Table S3: Estimation equations used for determination of photon flux (F_{254nm}), $O(^3P)$, $O(^1D)$, and O_3 exposures, based on Peng et al. (2016).

Reactant	Exposure estimation equation	eq.no
Photon flux	$$flux_{254nm,exp} = 10^{\left(1.0238 + \sqrt{(-1.0238)^2 - 4*0.060786*c}\right)/2*0.060786}$$ $$\text{where } c = -\left[log_{10}\left(OH_{exp} * \frac{180}{RT}\right) - \left\{15.514 + 0.79292 * log_{10}(H_2O) + 0.023076 * log_{10}(H_2O)^2 - \right.\right.$$ $$\left.\left. log_{10}\left(1 + e^{\frac{-0.42602 - log_{10}\left(\frac{O_{3,init}}{OHR_{ext}}\right)}{0.39479}}\right)\right\}\right]$$	(S7)
$O(^1D)$	$$log_{10}\left(O(^1D)_{exp}\right) = 3.7371 + 0.1608 * log_{10}\left(-log_{10} d^{\frac{180}{RT}}\right)$$ $$- 1.1344 * log_{10} H_2O + 0.59179 * log_{10} O_3, init$$ $$- 0.17019 * log_{10}\left(-log_{10} d^{\frac{180}{RT}}\right) * log_{10} H_2O$$ $$- 0.37983 * log_{10}\left(-log_{10} d^{\frac{180}{RT}}\right)^2 + 0.099941 * log_{10} OHR_{ext}$$	(S8)
$O(^3P)$	$$log_{10}\left(O(^3P)_{exp}\right) = 7.6621 + 0.16135 * log_{10}\left(-log_{10} d^{\frac{180}{RT}}\right)$$ $$- 1.1342 * log_{10} H_2O + 0.59182 * log_{10} O_{3,init}$$ $$- 0.17007 * log_{10}(-log_{10} d^{\frac{180}{RT}}) * log_{1o} H_2O$$ $$- 0.3797 * (log_{10}(-log_{10} d^{\frac{180}{RT}}))^2 + 0.099902 * log_{10}(OHR_{ext})$$	(S9)
O_3	$$log_{10}(O_{3,exp}) = 15.559 + log_{10} O_{3,init} + 0.42073 * log_e(d^{180/RT})$$	(S10)
for eq. S8-S10:	$$d = \left(10^{-10^{log_{10}(OH_{exp}/RT*180) - 13.322 + 0.22101*\left(\frac{OHR_{ext}}{O_{3,init}}\right)^{0.43529}}}\right)^{RT/180}$$	(S11)

75 **Table S4: Average exposures to radicals in the PEAR during photochemical aging experiments.**

	Exp.	O_3 input [ppm]	photon flux [photons cm^{-2}]	Fuel	OH_{exp} [# s cm^{-3}]	$O(^1D)_{exp}$ [# s cm^{-3}]	$O(^3P)_{exp}$ [# s cm^{-3}]	$O_{3\,exp}$ [# s cm^{-3}]
Masonry heater	2	2.3	8.3E+15	S-5%	5.8E+11	1.28E+6	1.07E+10	2.42E+15
				S-22%	3.5E+11	1.16E+6	9.75E+9	2.97E+15
	4	2.2	1.9E+15	S-5%	8.1E+10	3.74E+5	3.14E+9	5.85E+15
				S-22%	6.3E+10	6.25E+5	5.25E+9	4.87E+15
	5	1.8	1.36E+15	S-5%	8.1E+10	2.88E+5	2.42E+9	4.99E+15
				S-22%	6.3E+10	3.83E+5	3.22E+9	4.65E+15
Chimney stove	2	4.3	5.42E+15	B-17%	3.1E+11	1.51E+6	1.27E+10	6.86E+15
				S-22%*	not meas.	not meas.	not meas.	not meas.
	4	3.6	1.09E+15	B-17%	5.4E+10	3.08E+5	2.59E+09	1.08E+16
				S-22%	6.1E+10	4.10E+5	3.45E+09	1.03E+16
	5	11	1.06E+16	B-17%	5.0E+11	4.22E+6	3.55E+10	9.75E+15
				S-22%	5.0E+11	5.04E+6	4.24E+10	7.47E+15

* Direct OH exposure measurements not available for Chimney stove Exp. 2 S-22%; approximately similar as in Exp.2 B-17%.
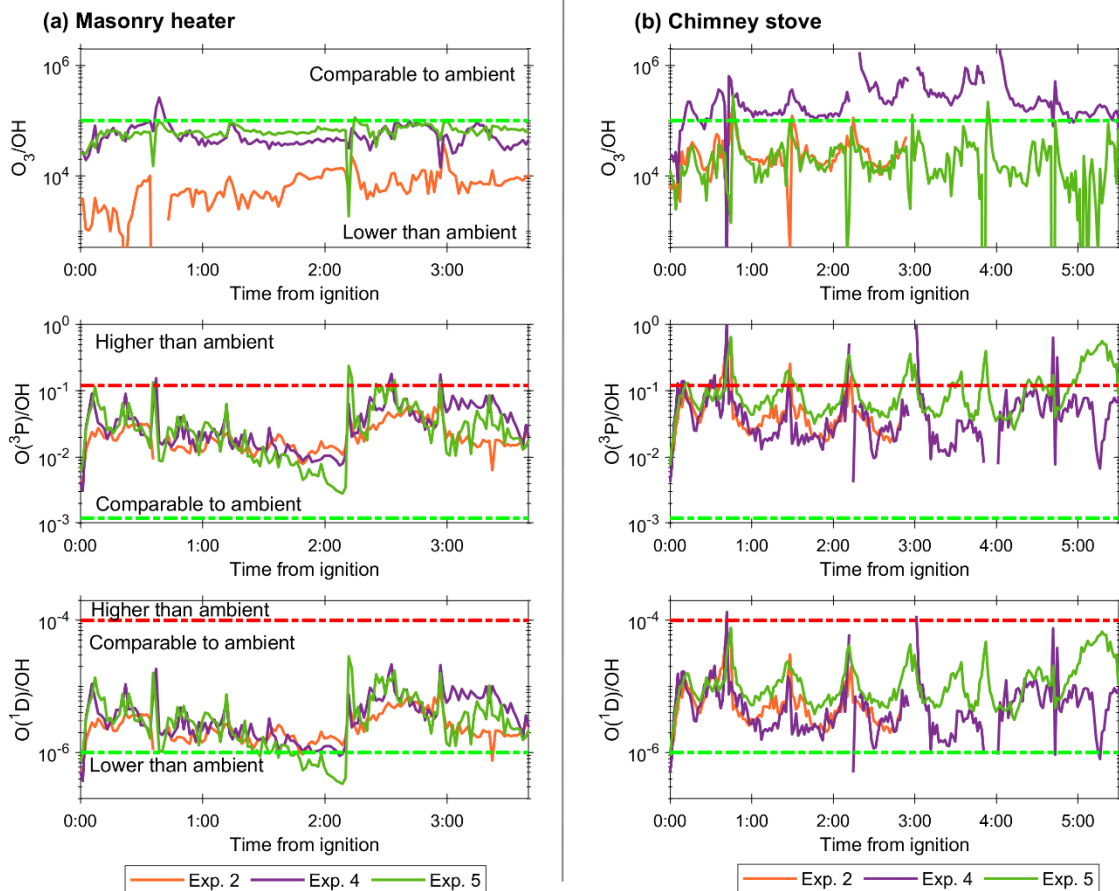
Figure S9: Ratios of alternative reaction pathways to the OH exposure. Ambient ratios as in Peng et al. (2016)

80   As an example of the relevance of the different reaction pathways, their importance towards the VOC fates was assessed for two important SOA precursors common for RWC: benzene and toluene (Fig. S10). The ratio of degradation by photolysis (D_{flux}) and reactions with alternative oxidants (D_X) to the degradation by OH reactions were calculated with Eq. (S12-S13), which show the inverse relationship between the importance of the pathway and the ratio of the photoabsorption cross section (σ_{abs}) or reaction rate of the compound with the considered radical ($k[X]_{VOC}$) to its reaction rate with OH ($k[OH]_{VOC}$).

85

$$D_{flux} = \frac{flux_{exp}}{[OH]_{exp}} * \frac{\sigma_{abs,VOC}}{k[OH]_{VOC}} \tag{S12}$$

$$D_x = \frac{[X]_{exp}}{[OH]_{exp}} * \frac{k[X]_{VOC}}{k[OH]_{VOC}} \tag{S13}$$

13

The absorption cross sections and reactivities used for the exemplary pathway estimations for toluene and benzene are listed

90 in Table S5. The total degradation in relation to degradation by OH was calculated with eq. (S14) by considering reactions with OH, O_3, and $O(^3P)$ and photolysis.

$$D_{total} = 1 + D_{flux} + D_{O3} + D_{O(3P)} \tag{S14}$$

Table S5: Absorption cross sections ($\sigma_{abs,}$ cm^2 molecule^{-1}) and reaction rates [cm^3 molec.$^{-1}$ s] of major RWC VOCs towards oxidants in PEAR

|	Benzene	Toluene	Phenol	Furan	Acetic acid	Acetaldehyde	Isoprene	References
$\sigma_{abs, 254nm}$	3.0E-19	2.3E-19	1.6E-18	7.1E-19*	4.8E-21	1.5e-20	5.3E-20	Keller-Rudek et al. (2015); * at 225nm
k[OH]	1.8E-12 [A]	6.16E-12 [A]	1.00E-11 [B]	4.04E-11 [A]	8.00E-13 [C]	1.62E-11 [A]	1.00E-10 [A]	A: (Atkinson, 1986), B: (Baulch et al., 1992); C: (Atkinson. et al., 2001)
k[O$_3$]	1.72E-22 [A]	3.90E-22 [A]		2.42E-18 [B]		3.40E-20 [c]	1.29E-17 [D]	A: (Toby et al., 1985), B: (Atkinson et al., 1983) C: (Stedman and Niki, 1973), D: (Grosjean and Grosjean, 1996)
k[O(^3P)]	1.99E-14 [A]	7.63E-14 [A]	1.56E-13 [B]		1.73E-15 [C]	4.58E-13 [B]	3.50E-11	A: (Cvetanović, 1987), B: (Baulch et al., 1994), C: (Herron, 1988), (Paulson et al., 1995)

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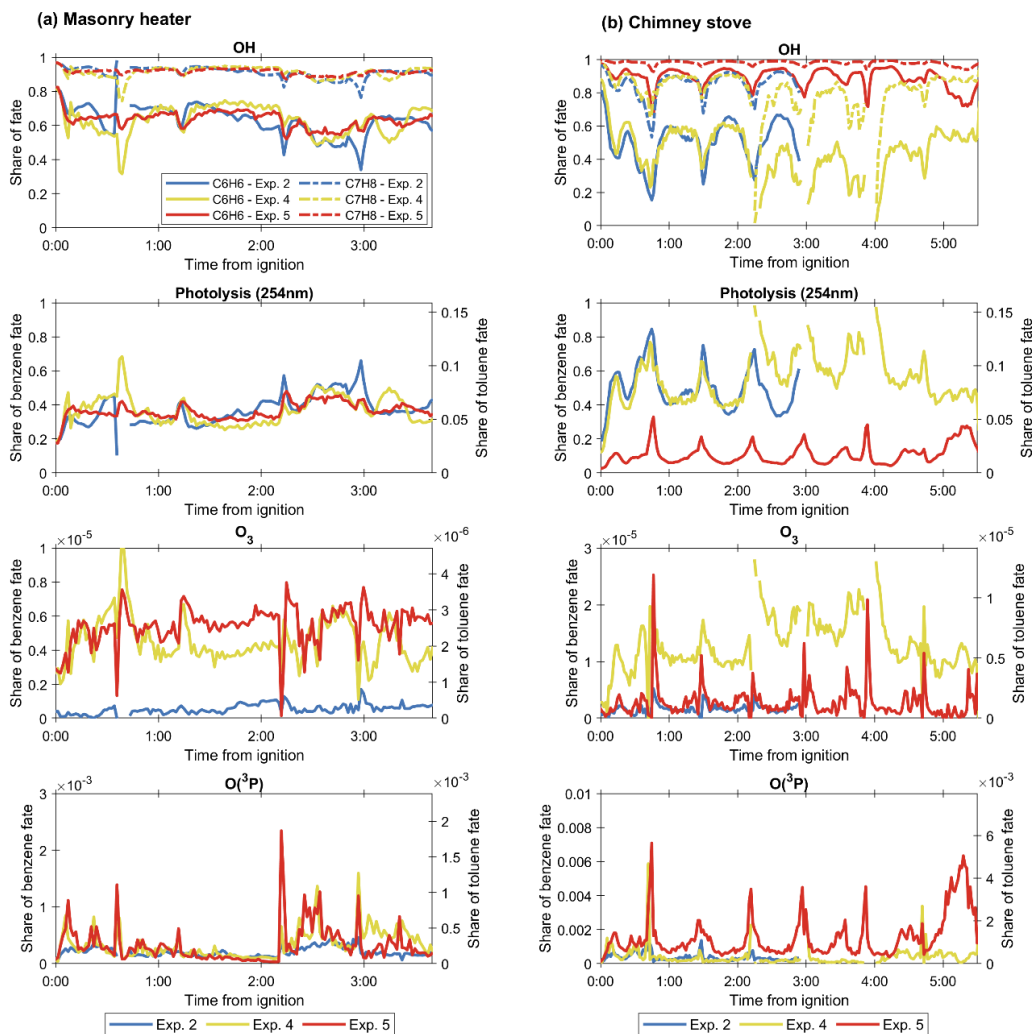
Figure S10: Share of the considered pathways in the total degradation of benzene and toluene in the PEAR during the experiments.

S3 Secondary gaseous phase

Table S6: Compounds identified from the RWC exhaust by PTR-ToF-MS and the applied reaction rates with H₃O⁺ (Cappellin et al., 2012).

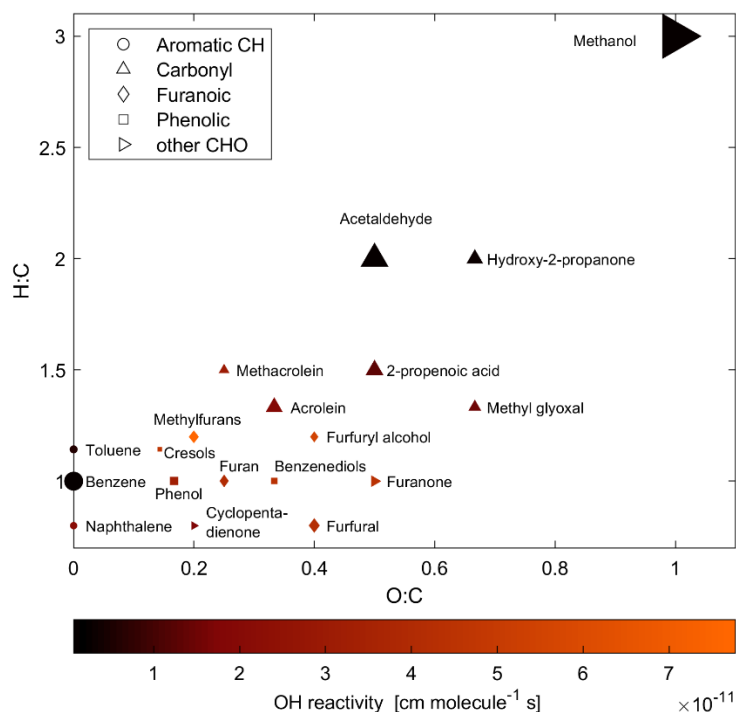| Group	m/z	Ion	Suggested compound	k-rate
Aliphatic hydrocarbons				
	53.04	C4H4-H+	Butenyne	2.00
	55.06	C4H6-H+	1,3-Butadiene	1.80
	67.06	C5H6-H+	1,3-cyclopentadiene	1.83
	69.07	C5H8-H+	Isoprene	1.95
	97.11	C7H12-H+	Cycloheptene	2.00
	109.10	C8H12-H+	e.g. Cyclohexene, 4-ethenyl-	2.00
	137.13	C10H16-H+	Monoterpenes	2.42
Aromatic hydrocarbons				
	79.05	C6H6-H+	Benzene	1.93
	93.07	C7H8-H+	Toluene	2.08
	105.07	C8H8-H+	Styrene	2.27
	107.09	C8H10-H+	Xylene, ethylbenzenes	2.26
	119.08	C9H10-H+	e.g. Indane, methylstyrene	2.00
	121.10	C9H12-H+	C3 benzenes; e.g. Cumene	2.40
	129.07	C10H8-H+	Naphthalene	2.45
	143.09	C11H10-H+	Methylnaphthalene	2.71
	155.08	C12H10-H+	e.g. Acenaphthene	2.82
	167.08	C13H10-H+	Fluorene	2.88
Phenolic compounds				
	95.05	C6H6O2-H+	Phenol	2.20
	109.07	C7H8O-H+	Cresols	3.20
	111.04	C6H6O2-H+	Benzenediols	2.00
	123.08	C8H10O-H+	Dimethylphenol	2.00
	125.06	C7H8O2-H+	Guaiacol	2.00
	139.04	C7H6O3-H+	e.g. Salicylic acid	2.00
	139.08	C8H10O2-H+	Creosol, Trosol	2.00
	153.07	C8H8O3-H+	e.g. Methoxy-benzoic acid	2.00
Furanoic compounds				
	69.03	C4H4O-H+	Furan	1.70
	83.05	C5H6O-H+	Methylfurans	2.00
	85.03	C4H4O2-H+	Furanone	2.00
	97.03	C5H4O2-H+	Furfural	3.95
	97.07	C6H8O-H+	Dimethyl- & ethyl furan	2.00
	99.04	C5H6O2-H+	Furfuryl alcohol	2.00
	119.05	C8H6O-H+	Benzofuran	2.00
	125.02	C6H4O3-H+	2,5-Furandicarboxaldehyde	2.00
	127.04	C6H6O3-H+	5-Hydroxymethylfurfural	2.00
Nitroaromatics				
	140.03	C6H5NO3-H+	Nitrophenols	2.00
	154.06	C7H7NO3-H+	e.g. Nitrocresol	2.00
Other oxygenated aromatics				
	107.05	C7H6O-H+	Benzaldehyde	3.73
	109.03	C6H4O2-H+	Benzoquinones	1.99
	121.07	C8H8O-H+	E.g. Acetophenone	3.36
	123.04	C7H6O2-H+	Benzoic acid	2.68
	133.06	C9H8O-H+	e.g. Cinnamic acid	2.00
	135.04	C8H6O2-H+	e.g. Isophthalaldehyde, phenylglyoxal	2.00
	135.08	C9H10O-H+	e.g. 4-Ethylbenzaldehyde	2.00
	137.06	C8H8O2-H+	e.g. Benzeneacetic acid	2.00
	141.05	C7H8O3-H+		2.00
	147.06	C9H6O2-H+	e.g. Coumarin	2.00
	149.02	C8H4O3-H+	Phthalic anhydride	2.00
	151.04	C8H6O3-H+	e.g. Benzoylformic acid	2.00
	151.08	C9H10O2-H+	e.g. Hydrocinnamic acid	2.00
	151.11	C10H14O-H+	e.g. Verbenone	2.00
	159.04	C10H6O2-H+	Naphthalenedione	2.00
	163.04	C9H6O3-H+		2.00
	169.07	C12H8O-H+	e.g. dibenzofuran	2.00
CHN				
	42.04	C2H3N-H+	Acetonitrile	3.90
	54.03	C3H3N-H+	Propenenitrile	2.00
	56.05	C3H5N-H+	e.g. Propanenitrile	2.00
	68.05	C4H5N-H+	e.g. Pyrrole	2.38
	70.07	C4H7N-H+	e.g. Butanenitrile	2.00
	84.09	C5H9N-H+	e.g. Pentanenitrile	2.00
	100.08	C5H9N3-H+		2.00
	104.05	C7H5N-H+	Benzonitrile	2.00
	118.06	C8H7N-H+	Indole	2.00
Carbonyls - primary				
	45.03	C2H4O-H+	Acetaldehyde	3.07
	55.02	C3H2O-H+	2-propynal	2.00
	57.03	C3H4O-H+	Acrolein	3.49
	71.02	C3H2O2-H+	Propiolic acid	2.00
	71.05	C4H6O-H+	Methyl vinyl ketone, methacrolein	3.20
	73.03	C3H4O2-H+	2-Propenoic acid	2.60
	75.04	C3H6O2-H+	Hydroxy-2-propanone	2.33
	83.01	C4H2O2-H+	3-Cyclobutene-1,2-dione	2.00
	85.07	C5H8O-H+	e.g. Pentenal	2.00
	87.05	C4H6O2-H+	e.g. 2,3- butadione	1.70
	99.08	C6H10O-H+	e.g. Hexenal	3.74
	101.06	C5H8O2-H+	e.g. Acetylacetone	2.93
	103.04	C4H6O3-H+	Acetic anhydride	2.00
	103.08	C5H10O2-H+	e.g. Methyl-butanoic acid	2.00
	117.02	C4H4O4-H+	Fumaric acid	2.00
	117.06	C5H8O3-H+	e.g. Acetyloxypropanone	2.00
Carbonyls - secondary				
	59.05	C3H6O-H+	Acetone, propanal	3.20
	61.03	C2H4O2-H+	Acetic acid, glycolaldehyde	2.30
	73.07	C4H8O-H+	2-butanone and 2-methylpropanal	3.12
	77.02	C2H4O3-H+	Acetic acid, hydroxy-	2.00
	89.03	C3H4O3-H+	E.g. Acetic acid anhydride with formic acid	2.00
	89.06	C4H8O2-H+	Acetoin	4.23
	99.01	C4H2O3-H+	Maleic anhydride	2.00
	101.02	C4H4O3-H+	Succinic anhydride	2.00
	105.04	C4H8O3-H+	e.g. Acetic acid, methoxy-	2.00
Other CHO				
	47.05	C2H6O-H+	Ethanol	2.11
	49.03	CH4O2-H+	Methyl peroxide or methanediol	2.00
	63.04	C2H6O2-H+	ethanediol	1.47
	77.05	C3H8O2-H+	e.g. Ethanol 2-methoxy- ; 1,3-Propanediol	3.50
	81.04	C5H4O-H+	2,4-Cyclopentadiene-1-one	2.00
	91.07	C4H10O2-H+	Butanediols	2.00
	111.08	C7H10O-H+	e.g. Heptenedial or trimethylfuran	2.00
	113.02	C5H4O3-H+	e.g. 2-furoic acid	2.00
	113.06	C6H8O2-H+	eg. Methylcyclohexanone	2.00
	115.04	C5H6O3-H+	e.g. Hydroxymethylfuranone	2.00
	115.08	C7H10O2-H+		2.00
	115.11	C7H14O-H+	e.g. Heptanal	2.83
	117.09	C6H12O2-H+		2.00
	127.08	C7H10O2-H+		2.00
	139.11	C9H14O-H+		2.00
	145.05	C6H8O4-H+	e.g. Dimethyl fumarate	2.00
	157.08	C8H12O3-H+		2.00
CHNO				
	44.01	CHNO-H+	Isocyanic acid	2.00
	58.03	C2H3NO-H+	Methyl isocyanate	2.00
	60.05	C2H5NO-H+	e.g. Acetamide	2.00
	62.03	CH3NO2-H+	e.g. Nitromethane, methyl nitrite	2.00
	70.03	C3H3NO-H+	e.g. Vinyl isocyanate	2.00
	74.03	C2H3NO2-H+	e.g. Nitroethylene	2.00
	74.06	C3H7NO-H+	Propanamide	2.00
	84.05	C4H5NO-H+		2.00
	86.03	C3H3NO2-H+	Acetic acid, cyano-	2.00
	86.07	C4H7NO-H+	e.g. Methacrylamide	2.00
	88.05	C3H5NO2-H+	Nitropropene	2.00
	88.08	C4H9NO-H+		2.00
	96.06	C5H5NO-H+	Pyridine-N-oxide	2.00
	100.04	C4H5NO2-H+		2.00
	112.05	C5H5NO2-H+		2.00
	114.02	C4H3NO3-H+	Nitrofuran	2.00
	114.06	C5H7NO2-H+	e.g. Ethyl cyanoacetate	2.00
	120.05	C7H5NO-H+	e.g. Benzisoxazole	2.00
	122.02	C2H3NO5 -H+	peroxyacetyl nitrate	2.00
	122.07	C7H7NO-H+	e.g. Benzamide	2.00
	124.05	C6H5NO2-H+	Nitrobenzene	2.00
	136.04	C7H5NO2-H+	e.g. 2-Furoylacetonitrile	2.00

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Figure S11: Van Krevelen diagram of selected VOCs commonly emitted from RWC. The marker size is scaled based on concentration in the fresh exhaust from spruce combustion in a modern masonry heater (Hartikainen et al., 2018), whereas the marker color indicates the compounds reactivity with OH.
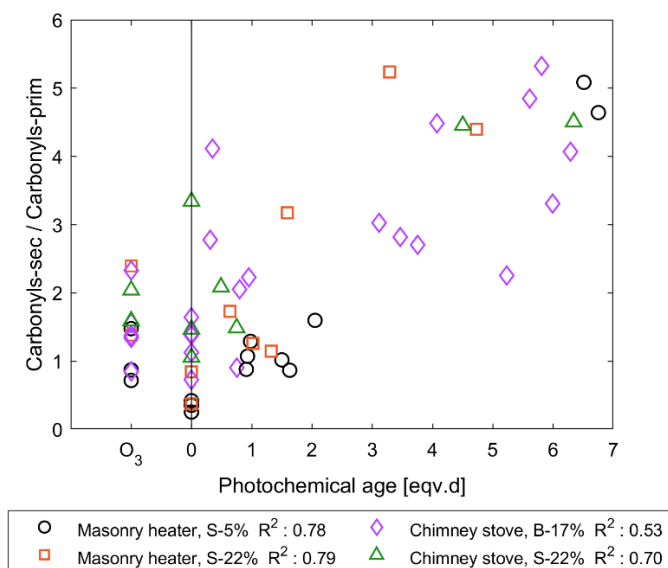

Figure S12: The ratios between the concentrations of the two carbonyl groups (secondary:primary) as the function of photochemical age.

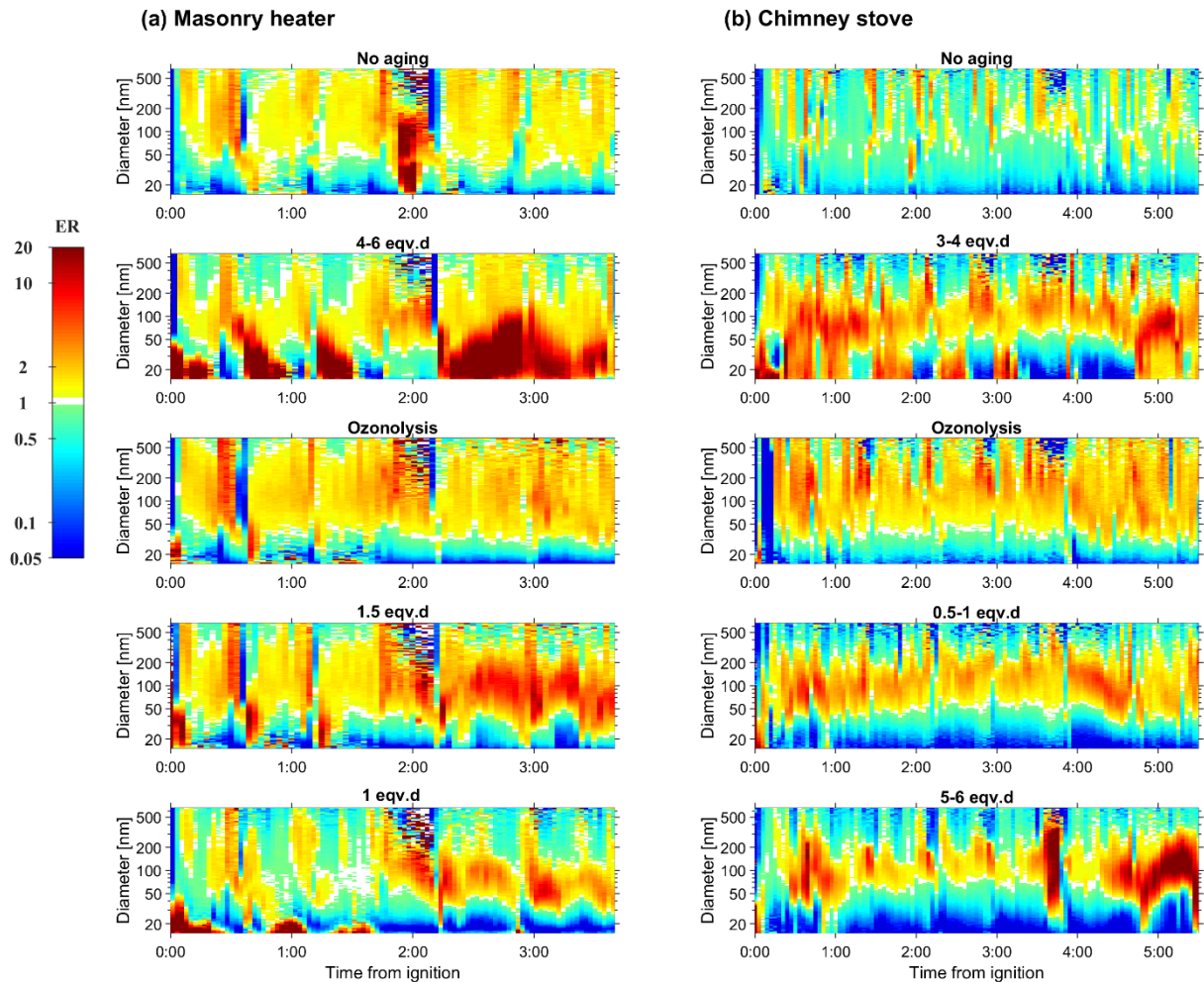110 **S4 Physical transformation of the particulate phase**

Figure S13: Particle number enhancement ratios (ER; secondary/primary) based on the SMPS measurements before and after PEAR.

S5 Aerosol mass spectrometry analyses

115   The measurements of the non-refractory chemical species (Org, NO$_3$, SO$_4$, NH$_4$ and Chl) were conducted with the standard tungsten vaporizer, while the SP-AMS dual vaporizer mode was used to characterise the refractory black carbon (rBC) (Onasch et al., 2012). Standard mass-based calibrations were performed for the ionisation efficiency in the AMS using ammonium nitrate and Regal Black (Regal 400R Pigment Black, Cabot Corp.) particles (Jayne et al., 2000; Onasch et al., 2012). SP-AMS

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was operated in V-mode from 12 to 555 m/z and the two vaporizer configurations were alternated every 120 s, including the
120 particle time-of-flight (PTOF) mode (duration 20 s). AMS data analysis was performed using the standard analysis tools
SQUIRREL v1.62A and PIKA v1.22A adapted in Igor Pro 8 (Wavemetrics). The data was corrected for time-dependent
background gas-phase CO_2 and for the interactions of inorganic salts with pre-deposited carbon on the tungsten vaporizer
(Pieber et al., 2016) using a relative ionisation efficiency (RIE) of 1.4.

125 **Figure S14: Correlations between variables measured by AMS after the PEAR and by thermal-optical analyses, respectively: (a) refractory black carbon (rBC) and elemental carbon (EC) before the PEAR; (b) rBC and EC after the PEAR; (c) organic aerosol (OA) and organic carbon (OC) after the PEAR.**

Figure S15: Concentrations and average carbon oxidation states (OS_C) of particulate OA from (a) masonry heater and (b) chimney
130 **stove after PEAR in exhaust normalised to dry, 13 % O_2 flue gas. Black dotted lines indicate ozonolysis experiments.**
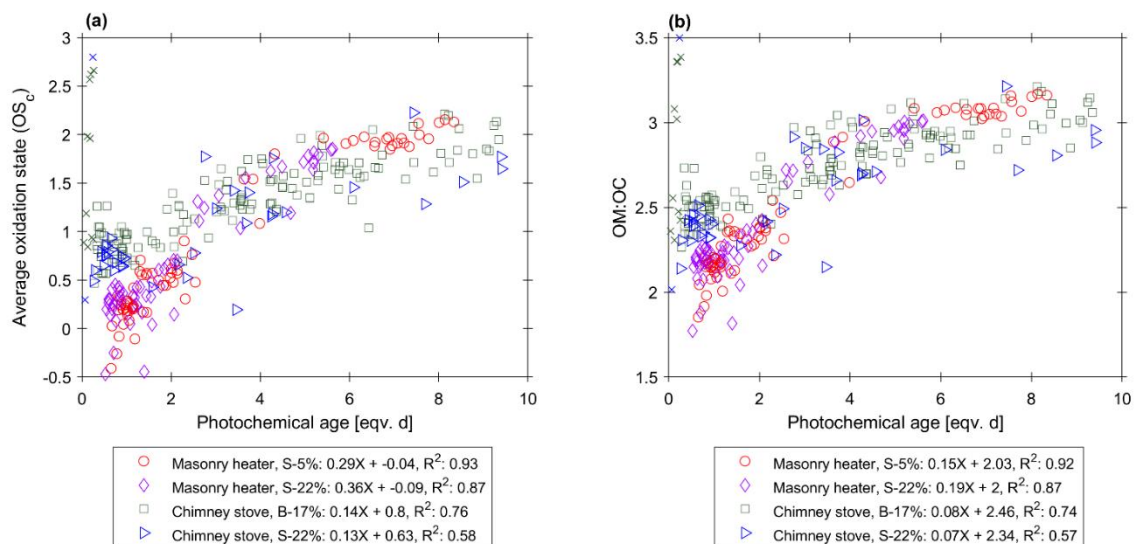
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Figure 16: Relationship between photochemical age and (a) oxidation state and (b) OM:OC ratio during the photochemical aging experiments. Coefficients of determination (R^2) were calculated for the photochemical exposure range of 0.25-10 eqv.d. Values outside this age range (crossed over) were not considered.

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Table S7: Average chemical composition of the OA (dilution corrected, dry, 13 % O_2) measured by AMS.

	Exp.	Fuel	Exposure [eqv.d]	O:C	H:C	OS_C	OM:OC	rBC mg m^{-3}	OA mg m^{-3}	PAH µg m^{-3}	NO$_3$ mg m^{-3}	SO$_4$ mg m^{-3}	NH$_4$ mg m^{-3}	Chl mg m^{-3}
Masonry heater	1	S-5%		0.55	1.51	-0.18	1.86	28.89	3.10	98	0.16	0.17	0.00	0.17
		S-22%		0.82	1.33	0.15	2.20	21.77	1.57	27	0.11	0.13	0.00	0.12
	2	S-5%	6.72	1.45	1.06	0.92	3.02	42.83	7.63	16	1.92	0.80	0.79	0.02
		S-22%	4.02	1.30	1.12	0.64	2.83	48.03	24.98	80	1.50	1.08	0.96	0.03
	3	S-5%	O$_3$	0.69	1.38	0.00	2.03	68.54	9.64	246	1.42	0.21	0.03	0.13
		S-22%	O$_3$	0.81	1.28	0.15	2.19	12.15	6.61	180	0.84	0.25	0.03	0.06
	4	S-5%	0.94	0.90	1.31	0.22	2.31	59.40	9.58	105	1.27	0.44	0.16	0.04
		S-22%	0.73	0.84	1.33	0.17	2.23	17.70	44.11	485	1.00	0.38	0.49	0.04
	5	S-5%	1.71	0.76	1.36	0.07	2.13	47.08	13.69	241	1.36	0.27	0.16	0.05
		S-22%	1.46	0.78	1.34	0.11	2.16	24.47	24.54	455	1.20	0.37	0.24	0.03
Chimney stove	1	B-17%		0.86	1.25	0.22	2.26	23.03	3.86	65	0.34	1.08	0.00	0.28
		S-22%		1.47	0.91	0.41	2.32	27.97	4.02	80	0.21	1.13	0.00	0.31
	2	B-17%	3.61	1.25	1.14	0.63	2.76	21.60	8.78	23	5.07	1.66	1.38	0.02
		S-22%	~4	1.47	1.19	0.90	2.80	19.70	10.39	26	4.05	1.33	0.96	0.02
	3	B-17%	O$_3$	1.01	1.15	0.41	2.44	26.24	8.33	114	3.32	1.82	0.22	0.12
		S-22%	O$_3$	0.82	1.30	0.16	2.20	24.89	10.34	271	2.41	1.12	0.16	0.13
	4	B-17%	0.62	1.09	1.17	0.47	2.55	20.93	9.95	52	10.90	2.93	1.88	0.08
		S-22%	0.70	1.04	1.23	0.05	2.37	27.91	11.46	215	6.40	1.89	1.02	0.08
	5	B-17%	5.78	1.36	1.11	0.80	2.91	21.81	9.45	22	7.34	2.68	1.66	0.02
		S-22%	5.83	0.87	1.32	0.10	2.74	19.63	32.71	158	4.63	1.26	1.31	0.03

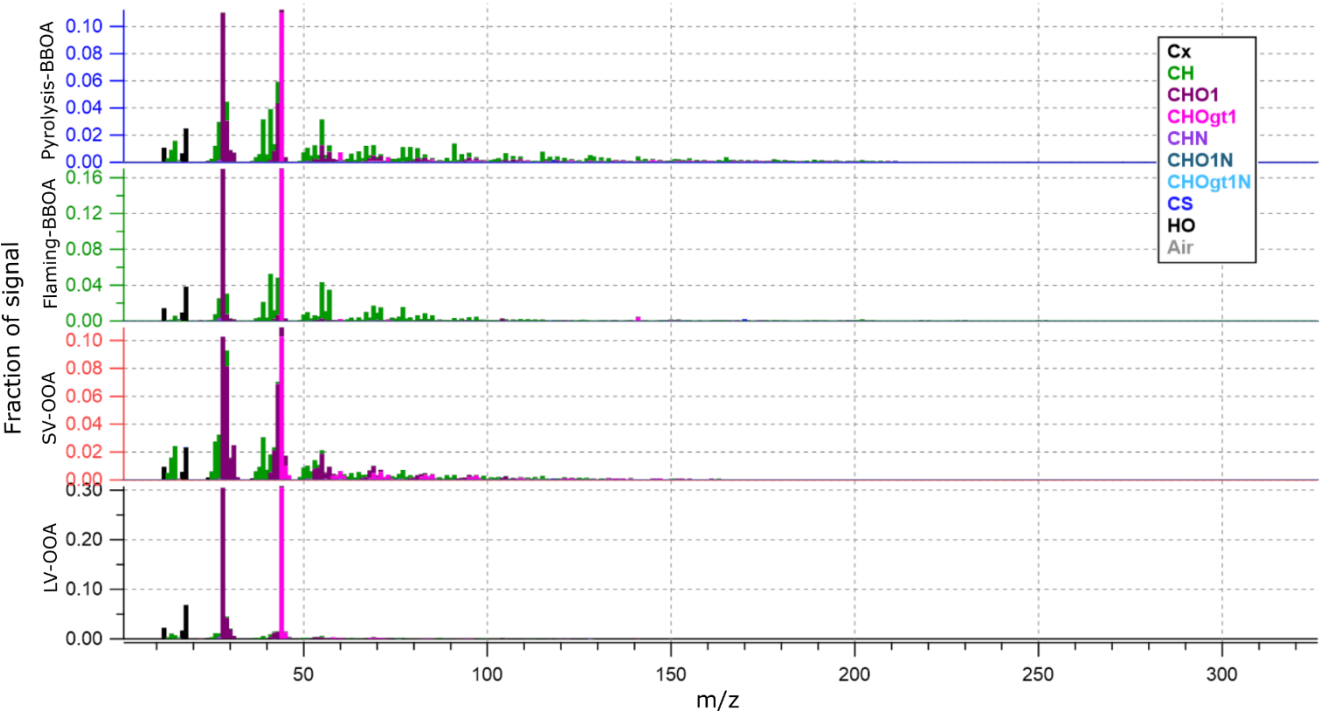**S5.1 Positive matrix factorisation analysis**

Figure S17: Spectra of the four factors identified from the AMS OA spectra by positive matrix factorisation analysis.
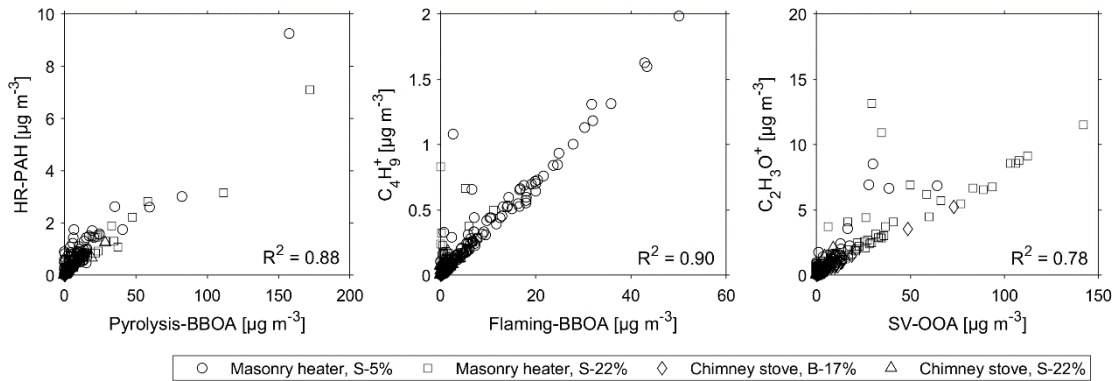

Figure S18: Correlations between (a) pyrolysis-BBOA and PAC concentration, (b) flaming-BBOA and $C_4H_9^+$, and (c) SV-OOA to $C_2H_3O^+$.

S5.2 HR-PAH analysis

Table S8: Ions included in the HR-PAH analysis, based on the grouping by Herring et al. (2015). Analysis was adapted for RWC exhaust with an addition of polycyclic aromatic compounds typically released from RWC (Avagyan et al., 2016; Bertrand et al., 2018; Bruns et al., 2015; Czech et al., 2018; Miersch et al., 2019).

Unsubstituted PAHs (UnSubPAH)	Oxygenated PAHs (OPAH)	Methylated PAHs (MPAH)	Nitrogen-substituted PAHs (NPAH)	Amino PAHs (APAH)
C10H8	C9H8O	C11H10	C10H7NO2	C16H11N
C12H8	C11H8O	C12H12	C12H9NO2	C18H13NO
C12H10	**C11H8O2***	C13H12	C13H9NO2	C20H13N
C13H10	C10H6O2	C14H12	C14H9NO2	C21H13N
C14H8	C12H8O	C15H12	C16H9NO2	
C14H10	C13H8O	C15H14	C13H8N2O4	
C15H10	C13H10O	C16H14	C18H11NO2	
C16H10	**C12H6O3***	C17H16		
C17H12	C12H6O2	**C18H14***		
C18H10	C12H8O2	C18H18		
C18H12	**C12H8O3***	C19H14		
C20H10	C14H10O	C20H16		
C20H12	C13H8O2	C21H16		
C22H12	C15H8O			
C22H14	C14H8O2			
C24H12*	**C15H8O2***			
	C16H8O2			
	C16H10O*			
	C17H10O*			
	C19H10O			
	C18H10O2			

* added for RWC

150 **S6 In-situ derivatisation and thermal desorption - gas-chromatography - time-of-flight mass spectrometry**

In-situ derivatisation and thermal desorption - gas-chromatography - time-of-flight mass spectrometry (IDTD-GC-ToFMS, (Orasche et al. 2011) was utilised in analysis of semi volatile organic target analytes in particulate phase. Subsamples were taken from the collected quartz fibre filters and prepared in GC-liners for the analyses. 10 µl Methyl-trimethylsilyl-trifluoroacetamid (MSTFA, Macherey-Nagel, Germany) was added automatically in each liner by the sampling robot (PAL

155 Focus, Atas GL, Netherlands) before samples were placed into a direct thermal desorption unit (Linex and Optic 3, Atas GL,

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Netherlands). Furthermore, the samples were spiked with mixtures of isotope labelled internal standards and calibration standards for the purpose of calibration and quantification of the non-polar (Table S9) and polar (Table S10) compounds.

MSTFA was continuously added to the helium carrier gas on a 4 ml min^{-1} stream, throughout the 16 minutes of thermal extraction at 300 °C. After the thermal extraction and derivatisation procedure, the pure helium flow was set back to 0.7 ml min^{-1}, with a split flow of 50 ml min^{-1}. IDTD was done at an isothermal GC temperature of 50° C in order to trap desorbed compounds at the front of the separation column before starting the GC-MS run. The column in use was an almost non-polar BPX5, 25 m, 0.22 mm ID, 0.25 µm film thicknesses (SGE, Australia), used in an Agilent 6890 gas chromatograph (Agilent, USA). Mass spectrometric detection in the range 35 to 500 m/z was carried out on a Pegasus III ToFMS (LECO, USA) using an acquisition frequency of 25 spectra per second. Evaluation of mass spectra was done with the ChromaTOF software package (LECO, USA).

Table S9: Isotope labelled internal standards for nonpolar compounds used in the IDTD-GC-ToFMS analyses.

Isotope labelled internal standard mixture	Calibration standard mixture	
9,10-Anthracenedione 13C6	1(2H)-Acenaphthylenone	Benz[a]pyrene
Acenaphthene d10	1,2,4-Trimethylanthracene	Benz[e]pyrene
Acenaphthylene d8	1,2-Dimethylnaphthalene	Benzo[a]anthracene-7,12-dione
Anthracene d10	1,3,6,8-Tetramethylpyrene	Benzo[b]fluoranthene
Benz[a]anthracene d12	1,3,7-Trimethylnaphthalene	Benzo[b]fluoranthene
Benz[a]pyrene d12	1,4-Naphthoquinone	Benzo[b]naphtho[1,2-d]thiophene
Benz[e]pyrene d12	1,8-Naphthalic anhydride	Benzo[b]naphtho[2,1-d]furan
Benzo[a]anthracene-7,12-dione d10	11H-Benzo[a]fluoren-11-one	Benzo[b]naphtho[2,1-d]thiophene
Benzo[b]fluoranthene d12	11H-Benzo[b]fluoren-11-one	Benzo[b]naphtho[2,3-d]furan
Benzo[bfluoranthene d12	1-Ethylpyrene	Benzo[c]phenanthrene
Benzo[ghi]perylene d12	1-Methyl-benz[a]anthracene	Benzo[ghi]perylene
Benzo[k]fluoranthene d12	1-Methylfluorene	Benzo[k]fluoranthene
Benzo[k]fluoranthene d12	1-Methylpyrene	Benzo[k]fluoranthene
Biphenyl d10	1-Naphthaldehyde	Biphenyl
Chrysene d12	2,2'-Binaphthalene	Chrysene
Coronene d12	2,3,6,7-Tetramethylanthracene	Coronene
Dibenz[ah]anthracene d14	2,6-Dimethoxybenzoquinone	Cyclopenta(def)phenanthrenone
Dibenzothiophene d8	2-Phenylnaphthalene	Dibenz[ac]anthracene
Docosane d46	3,6-Dimethylphenanthrene	Dibenz[ah]anthracene
Eicosane d42	3-Methylfluorene	Dibenzothiophene
Fluoranthene d10	4,5-Dimethylpyrene	Fluoranthene
Fluorene d10	5,12-Naphthacenedione	Fluorene
Hexadecane d34	6-Ethylchrysene	Indeno[1,2,3-cd]pyrene
Indeno[1,2,3-cd]pyrene d12	7,12-Dimethyl-Benz[a]anthracene	Methyleicosanoate
Naphthalene d8	9,10-Anthracenedione	n-Alkanes C12-C40
Octadecane d38	9,10-Dimethylphenanthrene	Naphthalene
Perylene d12	9H-Fluoren-9-one	Naphtho[2,1,8,7-klmn]xanthene
Phenanthrene d10	9-Methylphenanthrene	Perylene
Pyrene d10	Acenaphthene	Phenanthrene
Tetracosane d50	Acenaphthonquinone	Picene
Triacontane d62	Acenaphthylene	Pyrene
	Anthracene	Retene
	Benz[a]anthracene	Xanthone

Table S10: Isotope labelled internal standards used for polar compounds in the IDTD-GC-ToFMS analyses.

| Isotope labelled internal standard mixture	Calibration standard mixture	
4-Nitrophenol d4	1-Hydroxypyrene	Glutaric acid
Adipic acid d10	2-Hydroxy-1-naphthaldehyde	Hexanedioic acid
Cholesterol d6	2-Ketoglutaric acid	Hexanoic acid
Dodecanol d25	2-Methyl-4-Nitrophenol	Isopimaric acid
Fumaric acid d2	2-Methylerythritol	Levoglucosan
Glucose 13C6	2-Naphthanol	Linoleic acid
Glycerol d8	3-Hydroglutaric acid	Maleic acid
Levoglucosan 13C6	3-Hydroxyphenanthrene	Malic acid
Palmitic acid d31	4-Hydroxyphenanthrene	Malonic acid
Vanillin 13C6	4-Nitrocatechol	Mannosan
	4-Nitrophenol	Methyl vanillate
	9-Hydroxyphenanthrene	Oleic acid
	Abietic acid	Palmitic acid
	Acetosyringone	Phthalic acid
	Adipic Acid	Salicylic acid
	Azelaic acid	Sorbit
	Butanedioic acid	Stearic acid
	Cholesterol	Syringic Acid
	Coniferaldehyde	Tartaric acid
	Ergosterol	Threitol
	Erythritol	Trimellitic acid
	Fumaric acid	Vanillic acid
	Galactosan	Vanillin

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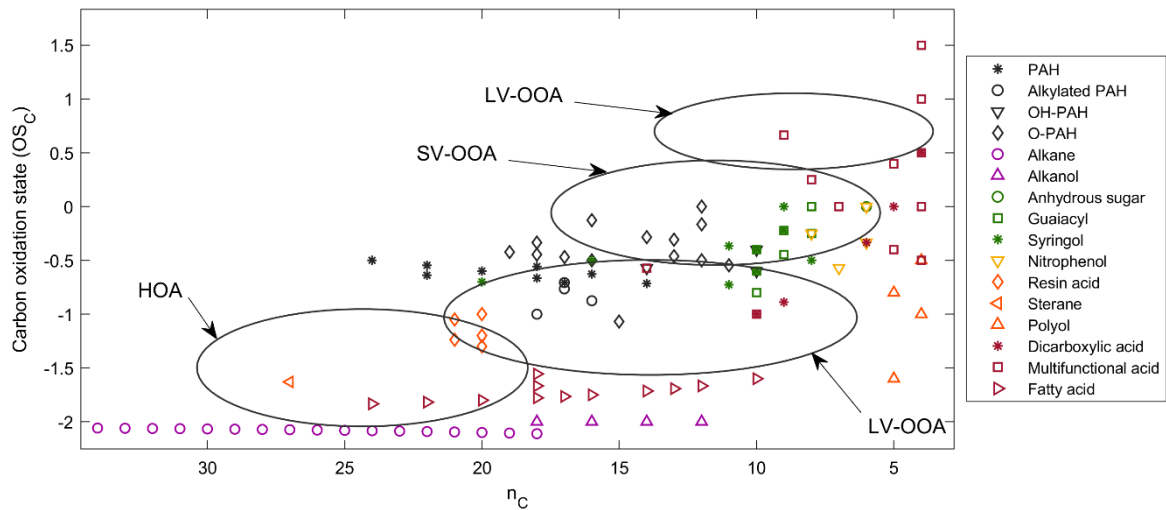
Figure S19: Compounds analysed by IDTD-GC-ToFMS displayed in the OS_C-n_C space. The ambient organic aerosol classes identified by Kroll et al. (2011) are presented as an approximate reference for average hydrocarbon-like OA (HOA), primary biomass burning OA (BBOA), semi-volatile oxidised OA (SV-OOA) and low-volatility oxidised OA (LV-OOA).

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Table S11: Pearson correlation coefficients between the particulate organic aerosol constituents measured by AMS (PMF and HR-PAH) and IDTD-GC-TOF-MS.

		PMF factors				HR-PAH					
		Pyrolysis-BBOA	Flaming-BBOA	SV-OOA	LV-OOA	Total	Unsub-PAH	OPAH	MPAH	NPAH	APAH
PMF factors	Pyrolysis-BBOA	1.00	0.17	0.40	-0.02	**0.96**	**0.96**	**0.87**	**0.97**	**0.86**	**0.89**
	Flaming-BBOA	0.17	1.00	-0.07	-0.21	0.19	0.21	0.17	0.14	0.17	0.21
	SV-OOA	0.40	-0.07	1.00	0.35	0.48	0.44	**0.70**	0.38	**0.68**	0.57
	LV-OOA	-0.02	-0.21	0.35	1.00	-0.02	-0.04	0.09	-0.04	0.06	0.03
HR-PAH	Total	**0.96**	0.19	0.48	-0.02	1.00	**1.00**	**0.94**	**0.99**	**0.94**	**0.96**
	UnsubPAH	**0.96**	0.21	0.44	-0.04	**1.00**	1.00	**0.93**	**0.99**	**0.93**	**0.95**
	OPAH	**0.87**	0.17	**0.70**	0.09	**0.94**	**0.93**	1.00	**0.88**	**0.98**	**0.94**
	MPAH	**0.97**	0.14	0.38	-0.04	**0.99**	**0.99**	**0.88**	1.00	**0.88**	**0.92**
	NPAH	**0.86**	0.17	**0.68**	0.06	**0.94**	**0.93**	**0.98**	**0.88**	1.00	**0.97**
	APAH	**0.89**	0.21	0.57	0.03	**0.96**	**0.95**	**0.94**	**0.92**	**0.97**	1.00
IDTD-GC-ToF-MS	PAH	0.01	**0.64**	-0.22	-0.17	0.19	0.27	0.04	0.15	0.09	0.09
	OH-PAHs	0.31	-0.05	**0.88**	**0.65**	**0.75**	**0.65**	**0.92**	0.54	**0.88**	**0.87**
	Carbonyl-PAHs	0.50	-0.12	0.49	0.35	**0.69**	**0.67**	**0.74**	0.52	**0.65**	**0.58**
	alkylated PAHs	0.01	**0.75**	-0.16	-0.26	0.21	0.29	0.01	0.24	0.09	0.14
	alkanes	-0.10	0.31	-0.19	-0.38	-0.17	-0.12	-0.25	-0.10	-0.22	-0.17
	alkanols	-0.11	0.26	0.43	**0.70**	0.22	0.17	0.36	0.06	0.35	0.38
	anhydrous sugars	0.53	-0.24	**0.70**	0.35	**0.68**	**0.61**	**0.71**	**0.65**	**0.78**	**0.76**
	guaiacyls	0.10	0.26	0.10	-0.09	0.29	0.30	0.21	0.26	0.28	0.28
	nitro phenols	-0.17	-0.14	**0.60**	**0.76**	0.09	-0.03	0.37	-0.08	0.32	0.31
	resin acids	**0.63**	0.23	-0.24	-0.42	0.45	0.54	0.09	**0.66**	0.25	0.24
	polyols	0.27	-0.09	**0.93**	0.63	**0.71**	**0.61**	**0.89**	0.51	**0.87**	**0.87**
	dicarboxylic acids	0.12	-0.01	**0.87**	**0.74**	**0.63**	0.53	**0.83**	0.40	**0.80**	**0.81**
	multifunctional acids/ anhydrids	-0.10	-0.16	**0.93**	**0.89**	0.40	0.26	**0.71**	0.15	**0.66**	**0.67**
	fatty acids	-0.05	0.50	-0.40	-0.34	-0.08	0.01	-0.26	-0.01	-0.21	-0.15

Table S12: Secondary concentrations (ng m^{-3}) of compounds measured by IDTD-GC-ToFMS. Normalised to dry, 13 % O$_2$ flue gas conditions. Attached as a .xlsx file.

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