SUPPORTING INFORMATION

Inhalable constituents of thirdhand tobacco smoke:

Chemical characterization and health impact considerations

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S1. Methods

S1.1. Operation of the smoking machine

Since real-world SHS includes about 15% exhaled mainstream smoke, the diluted sidestream smoke used in this study is best described as simulated SHS. The brands of cigarettes chosen for the laboratory study were Marlboro Special Blend (gold pack) and filtered Camel 99s, to match those used by the smoker whose home was monitored. Three cigarettes of each brand were mounted in alternating order into a laboratory-built multi-port programmable ignition system that was connected to a smoking machine (ADL/II, AD Little, Cambridge, MA). The cigarettes were smoked sequentially with one 35-mL puff every minute, until the burning cone was 1.0 cm from the filter. The sidestream smoke dispersed quickly into the chamber air, and the mainstream smoke was vented to the outdoors.

S1.2. Chromatographic conditions

Peaks were identified using authentic standards (for benzene, ethylbenzene, *m*-ethyl toluene, *p*-xylene, propylbenzene, *o*-xylene, *p*-ethyltoluene, styrene, toluene, acetone, chloromethane, dichloromethane and isoprene). Others were tentatively identified by searching the NIST mass spectral library. Concentrations of tentatively identified compounds were estimated based on peak areas relative to the internal standard (toluene-d_8). Quantification was carried out using fluorobenzene and toluene-d_8 as internal standards. Total VOC concentrations were calculated for all compounds with equivalent toluene-d_8 concentrations higher than 0.1 μg m^{-3}. The chromatographic conditions used in this study are summarized in Table 1S.

TABLE 1S: Summary of the GCxGC-TOF MS conditions used in this study

Parameter	Condition
ATD cartridge desorption	300 °C for 10 min at 45 mL/min with inlet split of 10 mL/min
Secondary trap	Perkin-Elmer “air toxics trap”; outlet split was off
Trap temperature	10 °C
Trap higher temperature	325 °C for 3 min
ATD transfer line temperature	225 °C
GC Injector	225 °C; split 8:1
Column flow	1.6 mL/min
Primary column	DB-VRX, 60 m,0.25 mm I.D., 1.4 µm film (Agilent, Santa Clara, CA)
Secondary column	Stabilwax, 2 m, 0.25 mm I.D., 0.25 µm film (Restek, Bellefonte, PA)
Primary GC oven program	50 °C hold for 5 min, 4 °C/min to 190 °C, then 5 °C/min to 235 °C and hold at 235 °C for 8 min
Secondary GC oven program	5 °C above the primary GC oven
GCxGC modulator	15 °C above the primary GC oven
GCxGC modulation	4 s period, 0.8 s hot pulse
GC transfer line temperature	240 °C
MS source	200 °C, electron impact (70 eV)
MS detector	1600 V
MS data acquisition	200 spectra/s; 34 to 400 amu

S1.3. Gravimetric determination of PM2.5.

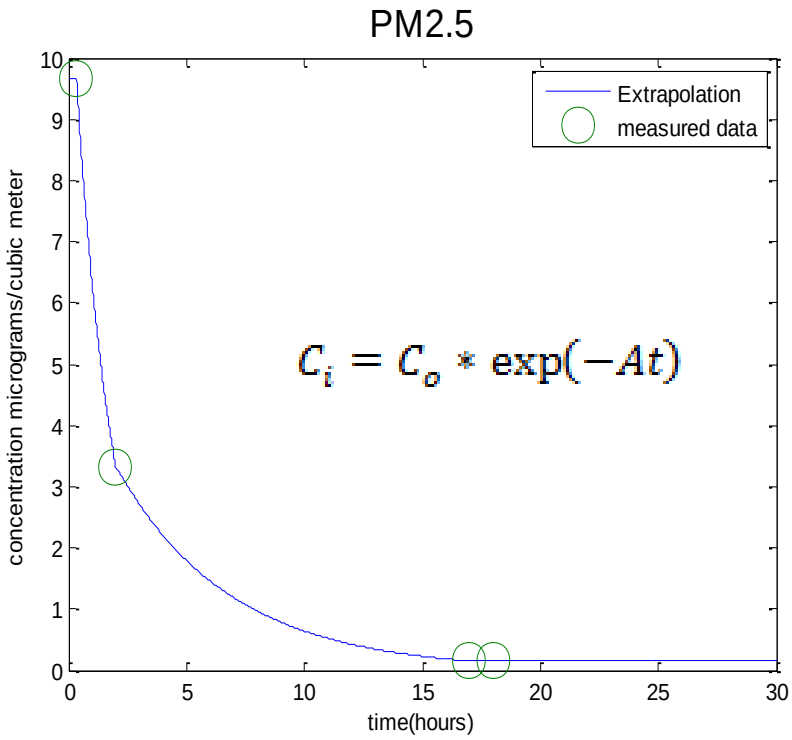
Teflon-coated fiberglass filters chosen because their Teflon coating is less susceptible to adsorption artifacts than uncoated fiberglass, and its mesh of coated fibers leads to smaller pressure drops than most types of Teflon membrane filters. Before and after use, the filters were equilibrated at 23 ^{o}C and 35 % RH for 24 h in moving air, then weighed on a microbalance (Sartorius SE-2F, Goettingen, Germany). Mass amounts on the downstream filters were about 5% of those on the upstream filters which is consistent with the manufacturer’s measurements of particle breakthrough of

0.3 μm diameter particles. That size is close to the mass median diameter estimated for SHS, 0.15 to 0.25 μm [11], depending on the aging time [12].

S1.4. Mass balance modeling

Concentration profiles such as those illustrated in Figure 1S (Supp. Info.) were generated for each pollutant in each scenario.

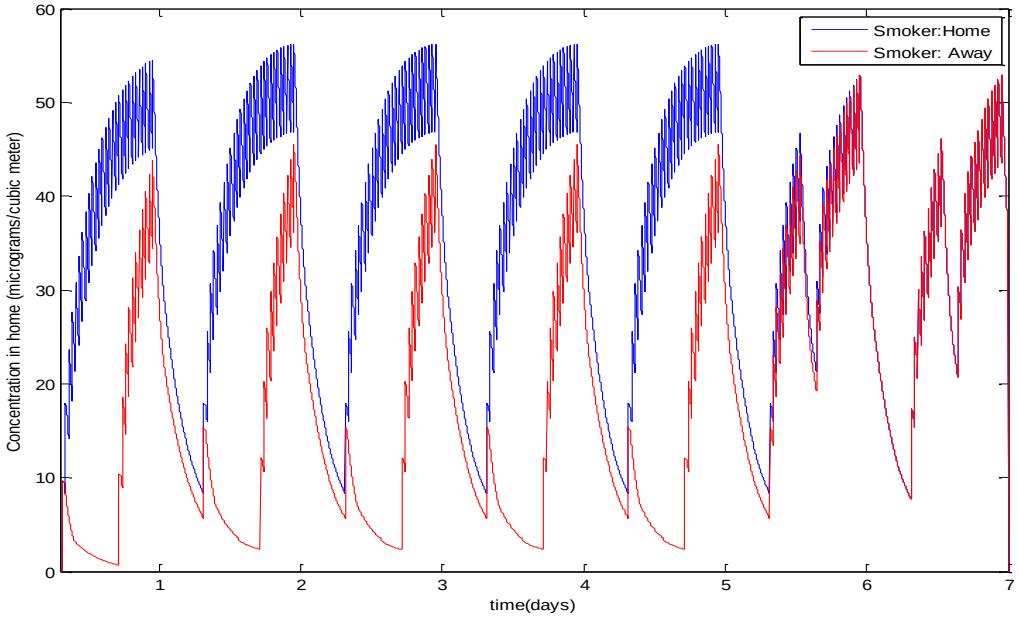
FIGURE 1S: An example of model extrapolation for PM 2.5. Concentrations correspond to emissions by a single cigarette, diluted into a 380 m^3 home.



Using the concentration vs. time profiles for one cigarette, we used superposition to determine the impact of smoking 28 cigarettes daily at regular time intervals [13]. Since

deposition and desorption rates in the chamber are a function of surface-to-volume ratio and air velocity in the room, the predicted concentrations constitute first-order approximations. Typical predicted weekly concentration profiles for scenarios in which the smoker is home and away are shown in Figure 2S (Supp. Info).

FIGURE 2S: Typical PM 2.5 weekly concentration profiles for scenarios in which the smoker is home and away.



S1.5. Computation of DALYs

Damage factors for PM2.5 are estimated from epidemiology-based concentration-response functions and published estimates of the DALYs lost per disease. Damage factors have high uncertainties largely due to uncertainties in concentration response functions. Confidence intervals are larger for VOCs because of uncertainties in extrapolating from animal-based toxicology results to human health impacts.

TABLE 2S: SHS/THS constituents included in the DALY calculation

SHS/THS constituent	CAS number	Health Effect
1,3-Butadiene	106-99-0	C&NC
1-Hexanol, 2-ethyl-	104-76-7	C
1-Propene, 2-methyl-	115-11-7	C
2-Butanone	78-93-3	NC
2-Butenal, (E)-	123-73-9	C
Acrylonitrile	107-13-1	C&NC
Acetaldehyde	75-07-0	C&NC
Acetonitrile	75-05-8	NC
Acrolein	107-02-8	NC
Benzaldehyde	100-52-7	C
Benzene	71-43-2	C&NC
Benzene, (1-methylethyl)-	98-82-8	NC
Ethyl alcohol	64–17–5	C
Ethyl benzene	100-41-4	C&NC
Furan	110-00-9	C
Furan, tetrahydro-	109-99-9	C
Hexane	110-54-3	NC
isoprene	78-79-5	C
Methane, chloro-	74-87-3	NC
Methylene Chloride	75-09-2	C
PM2.5	N/A	NC
Styrene	100-42-5	C&NC
Toluene	108-88-3	C&NC

C = cancer

NC = non-cancer

S2. Results and Discussion

S2.1. VOCs and $PM_{2.5}$ in aged tobacco smoke

Table 3S summarizes concentrations of VOCs measured in the LBNL chamber

TABLE 3S: Concentrations of VOCs measured in the LBNL chamber

Nitrogenated Compounds				
Code	**Name**	**20 min**	**2h**	**18h**
NC1	3-Ethenylpyridine	6.3	1.0	nd
NC2	2-Ethenylpyridine	2.4	nd	nd
NC3	2- & 3-Picoline	78.0	nd	nd
NC4	3-Ethylpyridine	7.1	nd	nd
NC5	Myosmine	1.5	nd	nd
NC7	Nicotine	48.9	nd	nd
NC8	Pyridine	119.2	nd	nd
NC10	Pyrrole	61.4	nd	nd
NL1	Acetonitrile	73.0	278.0	19.5
NL2	Acrylonitrile	100.2	4.2	2.2
NL3	Propanenitrile	57.9	0.8	0.9

Aromatics (AR)				
Code	**Name**	**20 min**	**2h**	**18h**
AR5	2,5-Dimethylfuran	95.8	13.5	3.8
AR7	Benzene	263.5	77.0	14.5
AR8	Ethylbenzene	108.5	10.1	4.7
AR9	Furan	261.3	27.6	5.6
AR10	Furan, 2-methyl	227.1	23.9	4.5
AR11	Iso-Propylbenzene	9.0	1.1	0.3
AR12	m-Ethyltoluene	80.2	4.5	3.2
AR13	m,p-Xylene	288.5	37.8	14.8
AR14	Naphthalene	3.8	0.6	0.3
AR16	n-Propylbenzene	18.9	1.1	0.8
AR17	o-Ethyltoluene	15.8	2.9	2.6
AR18	o-Xylene	81.1	32.0	3.4
AR19	p-Ethyltoluene	23.0	2.9	0.6
AR21	Styrene	129.0	7.5	0.2
AR23	Toluene	226.7	125.4	15.7

Carbonyls (CB) and Chlorinated VOCs (CL)

Code	Name	20 min	2h	18h
CB1	2,3-Butanedione	68.5	2.7	0.6
CB2	2-Butanone	19.3	5.7	3.6
CB3	Acetaldehyde	230.0	21.2	3.4
CB4	Acetone	244.5	49.1	25.0
CB5	Acetophenone	4.3	nd	0.1
CB6	Acrolein	127.9	7.0	2.4
CB7	Benzaldehyde	26.1	1.8	1.2
CB9	Methacrolein	90.7	6.4	nd
CB10	MEK	116.9	3.8	2.1
CL1	Chloromethane	28.1	7.1	0.4
CL2	Dichloromethane	32.7	24.3	6.4

Alkanes (KA), Alkenes (KE) and Terpenes (TP)

Code	Name	20 min	2h	18h
KA1	Butane	62.7	180.4	13.7
KA2	Isobutane	82.7	46.6	7.3
KA3	n-Decane	23.0	1.7	2.7
KA5	n-Nonane	22.0	2.1	2.0
KE1	1,3-Butadiene	122.4	34.0	3.9
KE2	1,3-Pentadiene	100.2	13.0	2.5
KE4	1-Butene	109.9	73.3	4.0
KE5	1-Nonene	51.1	3.2	2.3
KE6	1-Octene	60.0	5.2	2.4
TP1	d-Limonene	110.9	15.4	nd
TP2	Isoprene	273.3	265.5	32.2

Table 4S summarizes concentrations and emission factors of VOCs and other pollutants present in SHS, as measured in laboratory chambers.

TABLE 4S: Tobacco smoke VOC chamber levels reported in the literature

CODE	ANALYTE	MW	GAS PHASE CONCENTRATIONS (μg/m^3)					EMISSION FACTORS (μg/cigarette)				
			Mean	range	SD	RSD (%)	Ref	mean	range	SD	RSD (%)	Ref
CRITERIA POLLUTANTS												
	PM2.5		12130	7420–16000		23.5	[1]	17900			2	[2]
			2530				[3]					
			900	400-1400		111	[3]					
	RSP		1439		42		[4]	13674		411.0		[4]
	CO		11 ppm	9–12 ppm		11	[1]					
			5845		113		[4]	55101		1064		[4]
	NOx		195		3.2		[4]	1801		32		[4]
AMINES												
NC1	3-ethenylpyridine	105.4	10.5	8–14		20.8	[1]	174.4			33.6	[2]
				2-16			[5]	380.0	272-428	30		[5]
								660	450-890		23.0	[6]
			20.7				[3]					
			11.2	9-13		39	[3]					
			37.1		2		[4]	333		7.8		[4]
NC8	Nicotine	162.3	25.0	8.5–46		49.4	[1]	241			85.0	[2]
				4.9-64			[5]	689	396-1270	50.0		[5]
			982				[3]					
			nd				[3]					
			90.8		0.11		[4]	1585		42.0		[4]
NC10	pyridine	79		1.6-16			[5]	348	292-364	22		[5]
								430	340-670		29.0	[6]
			28.8				[3]					

			4.9	5		4	[3]					
			29.8		0.68		[4]	278.0		6.9		[4]
NC7	myosmine	146		0.3-3.1			[5]	46.0	25-83	5.0		[5]
			13.6				[3]					
			2.6	1.0-4.1		120	[3]					
			5.4		0.8		[4]	49		1.2		[4]
NC11	pyrrole	67						400	330-570		22	[6]
			56.3				[3]	231	157-230	23		[5]
			16.3	10-22.6		75	[3]					
NC6	N-methylformamide	59	28.6				[3]					
			10.8	6-15.5		81	[3]					
NC9	ammonia	17	453		11.3		[4]	4148		107		[4]
NC3,5	picoline (2- and 3-)	93	34.5		1.12		[4]	321		11		[4]
								338	260-390	20		[5]
NC2	ethenylpyridine	103	59.8		2.13		[4]	539		19		[4]
NC4	ethylpyridine	107	16.9		0.47		[4]	154		4.6		[4]
AROMATIC HYDROCARBONS												
AR7	Benzene	78	45.3	23–79		42	[1]	422			32	[2]
				1.7-23			[5]	435	429-435	29.0		[5]
								410	320-530		18	[6]
			29		0.47		[4]	280		4.9		[4]
AR8	Ethylbenzene	106	9.7	1.0–16		49	[1]	114			10	[2]
								101	83-142		22	[6]
								132	128-132	7.0		[5]
			8.5		0.13		[4]	79.6		1.4		[4]
AR13	m,p-Xylene	106	25.9	7–35		35	[1]	365			23	[2]
								300	260-400		17	[6]
								383	371-383	20.0		[5]
			25.6		0.45		[4]	238		4.8		[4]
AR18	o-Xylene	106	6.9	1.2–10		41	[1]	73.4			5.9	[2]

								75.0	68-75	5.0		[5]
			6.1		0.15		[4]	58.6		1.6		[4]
AR1	1.2.4-Trimethylbenzene	120.2						48.0			3.4	[2]
			4.2	1.9–5.4		26	[1]	64.0	57-74	6.0		[5]
AR2	1.3.5-Trimethylbenzene	120.2	0.85	0.17–1.59		55.0	[1]	12.9			0.7	[2]
			1.38		0.05		[4]	13.8		0.6		[4]
AR3	1,2,3-trimethylbenzene	120.2						43.3			2.4	[2]
			3.36		0.11		[4]	32.7		1.2		[4]
AR16	n-Propylbenzene	120.2	1.88	1.14–2.28		19.4	[1]	14.8			1.3	[2]
			1.04		0.08		[4]	8.5		0.9		[4]
AR11	iso-Propylbenzene	120.2	0.53	0.17–0.71		32.4	[1]	7.1			0.5	[2]
			0.48		0.05		[4]	5.1		0.7		[4]
AR4	tert-Butylbenzene	134.2	0.63	0.25–0.79		28.4	[1]					
AR6	n-Butylbenzene	134.2	0.82	0.54–1.04		23.1	[1]	5.7			0.5	[2]
AR23	Toluene	92	44.7	10.7–77.7		47.6	[1]	792			61.3	[2]
				3.4-46			[5]	863	821-863	43		[5]
								660	570-860		16	[6]
			54.5		0.97		[4]	498		10.8		[4]
AR17	o-ethyltoluene	120.2						21.2			2.1	[2]
AR12	m-Ethyltoluene	120.2	3.2	1.44–4.20		27.9	[1]					
AR19	p-ethyltoluene	120.2						63.0			4.4	[2]
AR20	p-isopropyltoluene (p-cymene)	134.2						30.1			1.9	[2]
AR21	Styrene	104	15.8	9.99–18.9		20.2	[1]	134			14	[2]
								147	122-191		17	[6]
								178	144-178	18.0		[5]
			10.2		0.16		[4]	94.3		1.8		[4]
AR14	naphthalene	128.2		0.14-1.2			[5]	25.0	17-34	2		[5]
								13.2			0.9	[2]
AR15	2-methyl naphthalene	142		0.09-0.72			[5]	14.0	8-21	1.2		[5]
AR22	1-methyl naphthalene	142						13.0	8-20	1.1		[5]

AR5	2,5-Dimethylfuran	96	21.0	8.58–30.5		36.5	(1)	211			15	[2]
ALKANES												
KA4	n-heptane	100.2						27			2.4	[2]
KA6	n-octane	114.2						19			1.4	[2]
KA5	n-nonane	128.2						21			1.7	[2]
KA3	n-decane	142.2						11			0.8	[2]
KA7	n-undecane	156.2						6.2			0.6	[2]
ALKENES												
KE1	1,3-butadiene	54		1.1-17			[5]	394	281-394	43.0		[5]
			40		1.26		[4]	373		12.6		[4]
KE6	1-octene	112.2						53.9			5.4	[2]
KE5	1-nonene	126.2						39.3			4.9	[2]
TERPENES												
TP2	isoprene	68		7.6-167			[5]	2420	1990-2830	330		[5]
			6570		12.5		[4]	6158		123		[4]
TP1	d-Limonene	136	34.2	19.2–43.9		22.8	[1]	475			50.3	[2]
								410	340-480		11.0	[6]
			29.1		0.49		[4]	269		5.3		[4]
NITRILES												
NL1	acetonitrile	41		3.6-4			[5]	850	812-858	140		[5]
			118		4.1		[4]	1145		45		[4]
NL2	acrylonitrile	53						207	161-207	13		[5]
PHENOLS												
PH1	phenol							47			9.7	[2]
								98	49-175	12		[5]
PH2	o-cresol	108.4		0.08-0.69			[5]	12	7-21	0.9		[5]
PH3	m,p-cresol	108.4						22	12-41	1.0		[5]
PH4	catechol	110	1.24		0.04		[4]	11		0.5		[4]
CARBONYLS												
CB6	acrolein	56						557	404-557	95		[5]

CB7	benzaldehyde	106				88			3.5	[2]
CB5	acetophenone	120				65			3.4	[2]
CB8	formaldehyde	30				1310	960-1880		27	[6]
			143	3	[4]	1333		34		[4]
CB3	acetaldehyde	44	268	6	[4]	2496		55		[4]
CB4	acetone	58				1190	930-1560		18	[6]
			116	5	[4]	1069		49		[4]
CB2	2-butanone	72				290	240-390		19	[6]
						315	275-327	25		[5]

Table 5S summarizes concentrations of VOCs and other pollutants present in SHS, determined in field measurements.

TABLE 5S: Tobacco smoke VOC field measurements reported in the literature

			SMOKING (μg/m³)				NON SMOKING (μg/m³)					
CODE	**Analyte**	**MW**	**mean**	**median**	**range**	**SD**	**mean**	**median**	**range**	**SD**	**Conditions**	**Ref.**
CRITERIA POLLUTANTS												
	PM2.5		146	148	63-311	67	14.9	8.6	0.4-73	19.4	(a)	[7]
	CO (ppm)		5.7	5.5	2.1-9.6	2.3	2.6	2.1	0.4-7.9	1.9	(a)	[7]
AMINES												
NC1	3-ethenylpyridine	105.4			nd - 13.3						(b)	[6]
			0.8	0.3	nd - 5.9	1.4	nd	nd	nd	nd	(c)	[8]
NC8	nicotine	162.3	1.86		1.3 - 2.4		0.02		0.01-0.03		(h)	[9]
NC10	pyridine	79			nd - 7.1						(b)	[6]
			0.9	0.1	nd - 6.6	1.6	nd	nd	nd	nd	(c)	[8]
NC11	pyrrole	67			nd - 9.4						(b)	[6]
AROMATIC HYDROCARBONS												
AR7	Benzene	78	1.65	1.39	0.4 - 5.4	1.11					(d)	[10]
			5.79	5.71	1.7 - 11.8	3.04					(e)	[9]
			2.64			1.94	1.98			1.1	(f)	[2]
					4.7 - 14.8						(b)	[6]
			16.30	11.4	4.2 - 63.7	15.3	11.5	6.6	3.4 - 51	11.8	(c)	[8]
			31.10	31.7	30.0 - 31.7	0.98	nd	nd	nd	nd	(g)	[7]
AR8	Ethylbenzene	106	1.94	1.54	1.01 - 7.95	1.51					(d)	[9]
			2.77	2.12	0.54 - 10.8	2.41					(e)	[9]
					1.3 - 8.7						(b)	[6]
			1.90	1.5	0.6 - 5.9	1.2	2.7	2.4	1.1 - 6.5	1.2	(c)	[8]
AR13	m,p-Xylene	106	2.56	2.15	0.73 - 6.78	1.42					(d)	[9]
			8.87	6.68	1.74 - 38.6	8.65					(e)	[9]
					3.9 - 32						(b)	[6]

			5.70	4.7	1.8 - 21.6	3.8	8.7	7.6	3.4 - 20.9	4.1	(c)	[8]
			25.5	25	25 - 27	0.9	nd	nd	nd	nd	(g)	[7]
AR18	o-Xylene	106	1.39	1.19	0.34 - 4.1	0.8					(d)	[9]
			2.48	2.04	0.71 - 9.7	2.0					(e)	[9]
					1.3 - 10						(b)	[6]
			1.40	1.2	0.4 - 6.3	1.1	2.5	2.2	1.0 - 6.0	1.3	(c)	[8]
AR1	1.2.4-Trimethylbenzene	120.2	2.41			2.6	2.0			1.6	(f)	[2]
			1.2	0.6	0.3 - 5.1	1.4	2.2	1.4	0.6 - 8.6	2.1	(c)	[8]
			6.1	8.3	nd - 10	5.4	nd	nd	nd	nd	(g)	[7]
AR2	1.3.5-Trimethylbenzene	120.2	0.8			0.8	0.7			0.55	(f)	[2]
			0.3	0.2	0.1 - 1.4	0.4	0.6	0.4	0.2 - 2.3	0.60	(c)	[8]
AR3	1,2,3-trimethylbenzene	120.2	0.77			0.7	0.7			0.88	(f)	[2]
AR23	Toluene	92	5.67	3.63	1.8 – 22	4.6					(d)	[9]
			19.2	14.5	2.3 - 67	17.7					(e)	[9]
					7.6 - 49						(b)	[6]
			29.20	28.4	9.6 - 78	17.4	47	45	8.8 - 99	22	(c)	[8]
			57.20	58.3	53 - 60	3.5	nd	nd	nd	nd	(g)	[7]
AR17	o-ethyltoluene	120.2	1.12	1.14	0.4 - 1.7	0.2					(d)	[9]
			1.50	0.98	0.34 - 6.9	1.5					(e)	[9]
			0.62			0.57	0.61			0.58	(f)	[2]
AR21	Styrene	104	1.28	1.24	1.1 - 1.7	1.6					(d)	[9]
			4.05	3.17	0.8 - 12	2.9					(e)	[9]
			0.60			0.49	0.54			0.46	(f)	[2]
					1.0 - 5.6						(b)	[6]
			0.80	0.5	0.2 - 3.4	0.8	0.7	0.4	0.3 - 3.2	0.7	(c)	[8]
AR14	naphthalene	128.2	1.13			0.77	3.3			8.4	(f)	[2]
			0.5	0.5	0.2 - 1.4	0.3	1	0.5	0.2 - 6.0	1.3	(c)	[8]
AR5	2,5-Dimethylfuran	96	0.54	0.44	nd - 1.4	0.39					(d)	[9]
			2.49	1.27	0.4 - 9.3	2.47					(e)	[9]
			0.20			0.31	0.02			0.09	(f)	[2]
ALKANES												

KA4	n-heptane	100.2	7.2	8.3	nd - 13.3	6.7	nd	nd	nd	nd	(g)	[7]
KA5	n-nonane	128.2	1.0			1.0	1.9			3.1	(f)	[2]
KA3	n-decane	142.2	2.0			3.5	3.5			6.7	(f)	[2]
KA7	n-undecane	156.2	2.2			4.8	2.3			4.4	(f)	[2]
ALKENE												
KE1	1,3-butadiene	54	1.7	0.7	0.3 - 10	2.5	0.5	0.4	0.1 - 1.1	0.3	(c)	[8]
TERPENES												
TP1	d-Limonene	136	32			38	38			50	(f)	[2]
					7.0 - 82						(b)	[6]
			189	217	67 - 283	111	123	85	83 - 200	66	(g)	[7]
TP3	alpha-pinene	136	3.3	0	nd - 10	5.7	7.2	10	nd - 11	6.3	(g)	[7]
CARBONYLS												
CB7	benzaldehyde	106	0.83	0.53	0.43 - 2.5	0.63					(d)	[9]
			7.6	8.7	nd - 15	5.9					(e)	[9]
CB8	formaldehyde	30			11.7 - 42						(b)	[6]
CB4	acetone	58			16 - 111						(b)	[6]
			114	47	45 - 250	118	18	25	nd - 31	16	(g)	[7]
CB2	2-butanone	72			nd - 10						(b)	[6]
CB11	methyl ethyl ketone	72	16	16	15 - 18	1.6	nd	nd	nd	nd	(g)	[7]
OTHER												
	1,4-dichlorobenzene	147	3.9	0	nd - 11	6.8	44	0	nd - 132	76	(g)	[7]
	2-propanol	60	21	18	nd - 45	22	54	60	28 - 73	23	(g)	[7]
	ethyl acetate	88	64	55	51 - 86	19	57	47	38 - 87	25	(g)	[7]
	iso-butanol	74	13	13	10 - 16	3.5	5	nd	nd - 15	8.7	(g)	[7]
	tetrachloroethylene	165	11	nd	nd - 11	6.8	nd	nd	nd	nd	(g)	[7]

(a) pre- and post-smoking ban (n = 17); (b) smoking rooms (n = 5); (c) 6 smoking homes vs. 6 non-smoking homes; (d) measured in the fall (n=21); (e) measured in the winter (n=20); (f) 19 smoking vs. 28 non-smoking; (g) pre- and post-smoking ban (n = 3); (h) 81 smoking homes vs. 50 non-smoking homes.

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