

Supplementary Materials

Valorization of Hazardous Organic Solid Wastes towards Fuels and Chemicals via Fast (Catalytic) Pyrolysis

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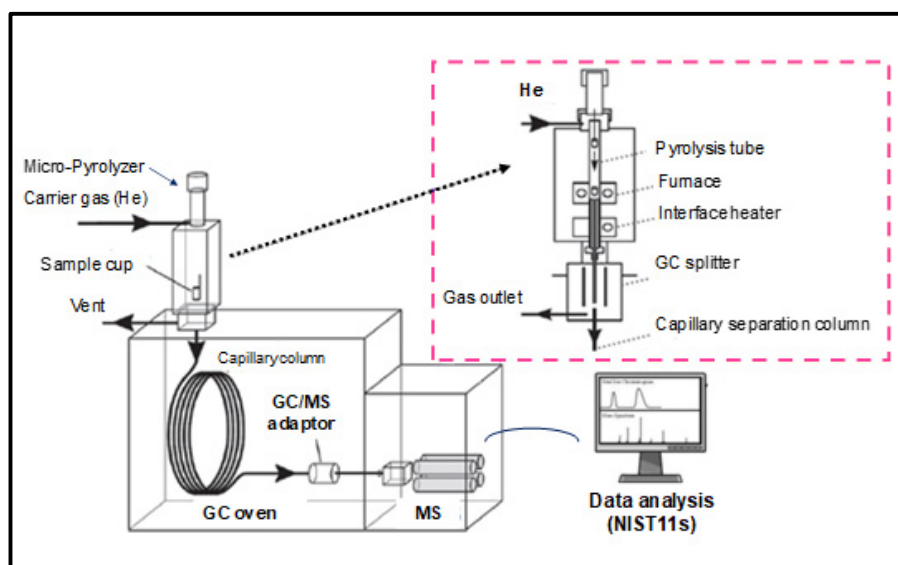


Figure S1. Schematic representation of the Py/GC-MS system.

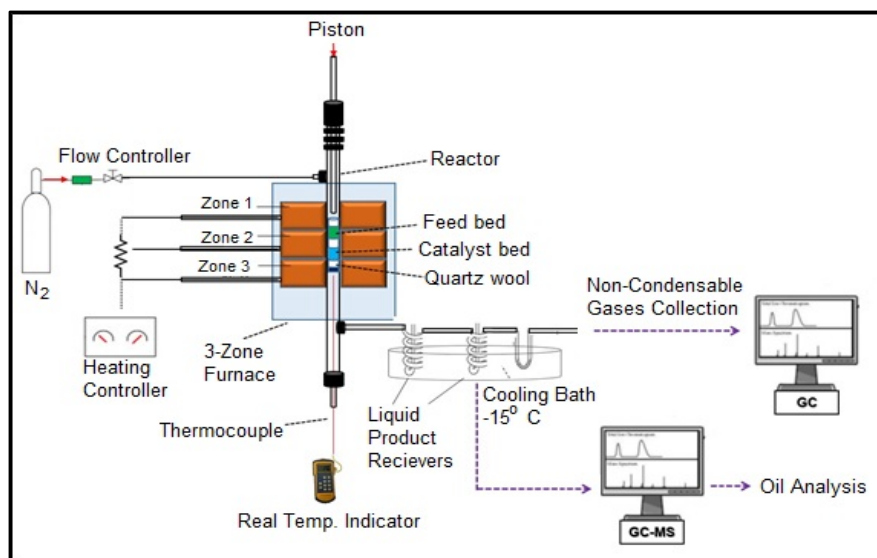


Figure S2. Schematic representation of bench-scale fixed bed reactor for fast pyrolysis of solid wastes.

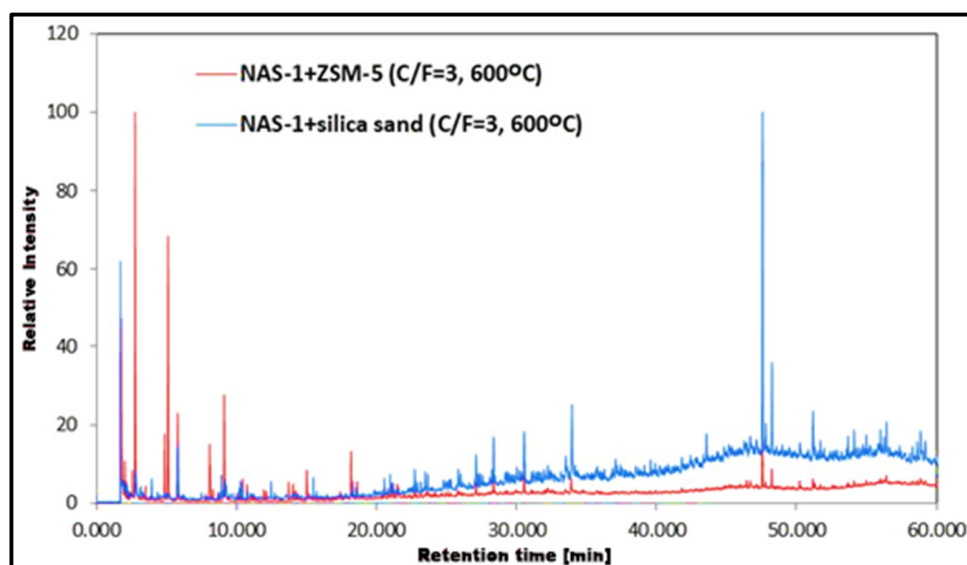


Figure S3. Representative Py/GC-MS spectra of the non-catalytic (thermal) and catalytic fast pyrolysis of NAS-1 at 600°C .

Table S1. Waste materials physicochemical properties.

Waste material type	Water Content (% wt.)	Ash (% wt.) [on dry base]
Petroleum containing sludges (NAS-1)	44.5	73.6
Residual paint on scrap metals (NAS-2)	3.6	30.4
Creosote treated wood waste (NAS-3)	9	1.5

Table S2. Composition of the ash of the three waste feeds.

Composition (% elemental ratio)	NAS-1	NAS-2	NAS-3	Composition (% elemental ratio)	NAS-1	NAS-2	NAS-3
Na	1.4	2.3	3.2	S	1.3	0.6	16.4
Mg	4.9	-	6	K	2.2	3.3	5.7
Al	10.1	6.3	7.6	Ca	31.9	67.6	46.3
Si	28.7	13.6	9.6	Ti	0.9	6.3	-
P	1.2	-	-	Fe	17.6	-	5.2

Table S3. Elemental analysis of waste materials (normalized values⁽¹⁾).

Waste	C (% w.t.)	H (% w.t.)	N (% w.t.)	S (% w.t.)	O (% w.t.)
NAS-1	57.85	5.08	1.35	4.23	31.50 ⁽²⁾
NAS-2	69.80	8.86	0.14	0.65	21.12
NAS-3	51.14	5.73	0.96	5.69	36.49

(1) Normalized values (Vn) have been estimated from the following equation considering the ash content (wt.%) of each solid waste: $Vn = a \times [100 / ((100 - y))]$, where a is the initial value from elemental analysis and y is the ash content (wt.%).

(2) The O content values for the petroleum sludge might be overestimated due to the very high content of ash (hydroxy-oxides).

Table S4. FT-IR peaks identification for the solid waste materials.

Wavenumber (cm ⁻¹)	Chemical bond type	Waste material
3622	O-H, alcohol or O-H humidity	NAS-1
3450	O-H, Alcohol	NAS-2
3325	O-H, Alcohol	NAS-3
3300	carboxylic acid	NAS-2
3050	Alcohol	NAS-3
2960	CH ₃ , asym. stretching, alkane	NAS-2
2925	CH ₂ , asym. stretching, alkane	NAS-1, NAS-3
2875	CH ₃ , sym. stretching, alkane	NAS-2
2850	CH ₂ sym. stretching, alkane or O-H alcohol intramolecular bonding	NAS-1, NAS-3
2510	S-H stretching thiol	NAS-1, NAS-2
2350	C=O from atmospheric CO ₂	NAS-2, NAS-3
1800	acid halide or C-H bending, aromatic compound	NAS-1
1730	C=O, stretching, aldehyde or ester	NAS-2
1660	C=C, stretching, alkenes	NAS-2
1600	C=O stretching, coniferyl/sinapyl aldehydes in lignin	NAS-3
1600	Aromatic C=C sym. stretching in lignin	NAS-3
1515	Aromatic C=C asym. stretching in lignin	NAS-3
1450	CH ₂ , bending, alkane	NAS-2, NAS-3
1450	Asymmetric C-H deformation, lignin	NAS-3
1425	CH ₂ +CH ₃ bending, alkane	NAS-1
1380	Sulfate	NAS-3
1275	C-O stretching in (aromatic) esters	NAS-2, NAS-3
1150	C-O, stretching, ether or alcohol	NAS-2
1065	primary alcohol	NAS-2
1025	C-O-C stretching of primary alcohol in cellulose and hemicellulose	NAS-3
1025	Si-O-Si stretching, clays	NAS-1
875	C-H bending, 1,2,4 trisubstituted	NAS-1, NAS-2

	CaCO ₃	NAS-2
800	C-H bending, 1,2,3,4 tetrasubstituted	NAS-1
750	monosubstituted	NAS-3
715	C-H bending, aromatic	NAS-2

Table S5. Composition of bio-oil derived from non-catalytic fast pyrolysis of petroleum sludge (NAS-1) in the Py/GC-MS system, at 600°C (GC-MS peak area, %).

<i>Compound</i>	<i>Group</i>	<i>%</i>	<i>Compound</i>	<i>Group</i>	<i>%</i>
1-Butanol	AL	0.32	2-methylhexacosane	ALI	0.82
Methyl methacrylate	EST	0.72	9-Hexadecenoic acid, eicosyl ester, (Z)-	EST	0.44
Toluene	AR	4.79	Bicyclo[4.2.0]octa-1,3,5-triene	AR	3.39
Styrene	AR	7.87	1,6-Octadiene, 2,7-dimethyl-	ALI	0.44
1-Nonene	ALI	1.97	Cyclohexene, 1-methyl-5-(1-methylethenyl)-	ALI	0.55
.alpha.-Methylstyrene	AR	1.35	2-Pentene, 2,4,4-trimethyl-	ALI	0.50
2-Butenoic acid, butyl ester	EST	1.98	Tridecane	ALI	0.41
Phenol	PH	0.66	Dodecane, 2,6,10-trimethyl-	ALI	2.17
1-Decene	ALI	2.30	1-Tetradecene	ALI	0.96
D-Limonene	ALI	1.66	Tetradecane	ALI	0.46
2,4,4,6,6,8,8-Heptamethyl-2-nonene	ALI	0.62	2-Ethyl-1-dodecanol	AL	0.53
1-Undecene	ALI	1.56	Pentadecane	ALI	0.82
1-Dodecene	ALI	2.19	Pentadecane, 3-methyl-	ALI	0.44
1-Tridecene	ALI	2.46	Cetene	ALI	0.88
1-Pentadecene	ALI	1.42	Hexacosane	ALI	0.73
1-Heptadecene	ALI	0.71	Pentadecane, 2,6,10-trimethyl-	ALI	2.19
Phenol, 2,6-dimethoxy-4-(2-propenyl)-	OxyPH	0.86	Tridecane, 4-cyclohexyl-	ALI	0.57
n-Dodecyl methacrylate	ALI	0.53	Hexadecane, 4-methyl-	ALI	0.69
1-Heneicosanol	AL	0.53	Tetratetracontane	ALI	0.60
Heneicosane	ALI	3.35	Heptadecane	ALI	0.76
Heptadecanal	ALD	0.54	Pentadecane, 2,6,10,14-tetramethyl-	ALI	3.51
Octacosane	ALI	1.39	Hexadecane, 2,6,10,14-tetramethyl-	ALI	4.74
n-Nonadecanol-1	AL	1.29	Carbonic acid, propargyl 2-ethylhexyl ester	EST	0.72
1-Decanol, 2-hexyl-	AL	2.64	Octadecane, 3-methyl-	ALI	0.61
Nonadecane, 4-methyl-	ALI	0.34	Octadecanoic acid, 2-propenyl ester	EST	1.10
Heneicosane, 3-methyl-	ALI	0.91	Cyclohexane, (1-hexyltetradecyl)-	ALI	0.56
Docosane	ALI	1.31	2-methyltetracosane	ALI	0.56

Pentatriacontane	ALI	5.10	Hexadecanoic acid, 1-(hydroxymethyl)-1,2-ethanediyl ester	EST	0.73
Nonacosane	ALI	1.11	13-Octadecenal, (Z)-	ALD	0.29
Eicosane	ALI	1.14	1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl) ester	EST	1.08
17-Pentatriacontene	ALI	0.58	Nonahexacontanoic acid	AC	0.98
Dodecane, 4-cyclohexyl-	ALI	0.58	1,1,3,6-tetramethyl-2-(3,6,10,13,14-pentamethyl-3-ethyl-pentadecyl)cyclohexane	ALI	0.23
Tetracosane	ALI	0.74	17-(1,5-Dimethylhexyl)-10,13-dimethyl-	ALI	4.47
Benzeneacetic acid, 4-tridecyl ester	EST	0.73	2,3,4,7,8,9,10,11,12,13,14,15,16,17-tetradecahydro-1H-cyclopenta[a]phenanthren-3-ol		
Tetracontane	ALI	1.07	Triacontane, 1-bromo-	ALI	0.75
Tetrapentacontane	ALI	1.32	Tetracontane-1,40-diol	AL	0.87
Disulfide, di-tert-dodecyl	SUL	0.21	13-Methyl-Z-14-nonacosene	ALI	0.46

Table S6. Composition of bio-oil derived from catalytic fast pyrolysis (with ZSM-5 zeolite as catalysts) of petroleum sludge (NAS-1) in the Py/GC-MS system, at 600°C (GC-MS peak area, %).

<i>Compound</i>	<i>Group</i>	<i>%</i>	<i>Compound</i>	<i>Group</i>	<i>%</i>
Cyclopentene, 4-methyl-	ALI	1.06	Tetradecanoic acid	AC	0.24
Benzene	AR	9.79	Dotriacontyl heptafluorobutyrate	ALI	0.49
Cyclopentane, 1,3-dimethyl-	ALI	0.67	Tetrapentacontane, 1,54-dibromo-	ALI	0.07
Cyclopentene, 4,4-dimethyl-	ALI	2.29	n-Tetracosanol-1	AL	0.15
Cyclohexane, methyl-	ALI	0.40	Oxalic acid, allyl octadecyl ester	EST	0.16
Cyclobutane, (1-methylethylidene)-	ALI	1.20	Heptafluorobutyric acid, n-tetradecyl ester	EST	0.41
Toluene	AR	18.13	2-Octadecyl-propane-1,3-diol	AL	1.48
Cyclopentene, 1,2,3-trimethyl-	ALI	0.53	Sulfurous acid, octadecyl 2-propyl ester	EST	2.74
Ethylbenzene	AR	3.35	Cyclohexene	ALI	0.58
Benzene, 1,3-dimethyl-	AR	18.38	1-Hexene, 5-methyl-	ALI	0.51
o-Xylene	AR	2.49	Cyclopentene, 1,5-dimethyl-	ALI	0.40
Benzene, propyl-	AR	0.34	Cyclopentane, ethyl-	ALI	0.07
Benzene, 1-ethyl-2-methyl-	AR	2.30	Cyclohexene, 4-methyl-	ALI	0.12
Benzene, 1,2,4-trimethyl-	AR	2.92	1,3,5-Hexatriene, 3-methyl-, (Z)-	ALI	0.08
Benzonitrile	NIT	0.38	1,3-Dimethyl-1-cyclohexene	ALI	0.30
Indane	AR	1.24	Octane, 3-methyl-	ALI	0.11

Indene	AR	1.09	Azulene	ALI	1.05
1-Phenyl-1-butene	PH	0.25	Dodecane, 2,6,10-trimethyl-	ALI	0.28
2,4-Dimethylstyrene	AR	0.36	Hexadecane	ALI	0.27
1H-Indene, 2,3-dihydro-4-methyl-	AR	0.55	Benzene, 1-ethyl-3-methyl-	AR	2.41
1H-Indene, 3-methyl-	AR	1.28	Benzene, 1,2,3-trimethyl-	AR	3.28
1H-Indene, 1-methyl-	AR	0.37	Benzene, 1,4-diethyl-	AR	0.12
Naphthalene, 1,2,3,4-tetrahydro-	PAH	0.29	Benzene, 1-methyl-4-propyl-	AR	0.14
Naphthalene	PAH	1.34	Benzene, 1-methyl-2-(2-propenyl)-	AR	0.23
1H-Indene, 1,1-dimethyl-	AR	0.10	Benzene, 1-ethenyl-4-ethyl-	AR	0.38
Naphthalene, 2-methyl-	PAH	2.41	1H-Indene, 2,3-dihydro-5-methyl-	AR	0.53
Dodecane, 2,6,11-trimethyl-	ALI	0.20	2-Methylindene	AR	0.35
Naphthalene, 1-ethyl-	PAH	0.54	1H-Indene, 2,3-dihydro-4,7-dimethyl-	AR	0.12
Naphthalene, 1,7-dimethyl-	PAH	1.21	Biphenyl	AR	0.12
Naphthalene, 2,3-dimethyl-	PAH	0.36	1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl) ester	EST	0.20
Naphthalene, 1,5-dimethyl-	PAH	0.23	Tricyclo[3.1.0.0(2,4)]hex-3-ene-3-carbonitrile	NIT	0.37
Hexadecane, 2,6,10,14-tetramethyl-	ALI	1.78	Naphthalene, 1-methyl-	PAH	1.35
Eicosanoic acid	AC	0.18	Naphthalene, 1,8-dimethyl-	PAH	0.39
Pentadecane, 2,6,10-trimethyl-	ALI	0.78	Naphthalene, 2,6-dimethyl-	PAH	0.25
Pentadecane, 2,6,10,14-tetramethyl-	ALI	1.35	Naphthalene, 2,3,6-trimethyl-	PAH	0.12

Table S7. Composition of bio-oil derived from non-catalytic fast pyrolysis of creosote-impregnated wood (NAS-3) in the Py/GC-MS system, at 600°C (GC-MS peak area, %).

<i>Compound</i>	<i>Group</i>	<i>%</i>	<i>Compound</i>	<i>Group</i>	<i>%</i>
1,3,5-Hexatriene	ALI	0.11	11H-Benzo[a]carbazole	NIT	0.41
1-Hydroxy-2-butanone	KET	0.09	1-Phenyldibenzofuran	OxyAR	0.12
Bicyclo[4.2.0]octa-1,3,5-triene	AR	0.29	Naphtho[2,1,8,7-klmn]xanthene	OxyAR	0.31
Phenol	PH	0.51	1-Aminopyrene	NIT	0.56
Indene	AR	0.21	Benz[a]anthracene, 7-methyl-	PAH	1.01
Phenol, 2-methyl-	PH	0.13	Chrysene, 1-methyl-	PAH	0.24
Phenol, 3-methyl-	PH	0.26	5,12-Naphthacenedione	PAH	0.19
Phenol, 2-methoxy-	OxyPH	1.75	10,11-Dihydro-10-hydroxy-2,3-dimethoxydibenz(b,f)oxepin	AL	0.38
2-Methylindene	AR	0.14	Benz(a)anthracene-7,12-dione	PAH	0.50
Creosol	OxyPH	3.02	Benz[e]acephenanthrylene	PAH	5.26
4-Hydroxy-3-methylacetophenone	OxyPH	1.61	Perylene	PAH	1.27
Eugenol	OxyPH	1.15	Acetic acid	AC	0.69

Phenol, 2-methoxy-4-(1-propenyl)-, (Z)-	OxyPH	0.68	N-Nitrosodimethylamine	NIT	0.32
Acenaphthylene	PAH	0.41	Furan, 2-methyl-	FUR	0.14
trans-Isoeugenol	OxyPH	4.06	Toluene	AR	0.36
Acenaphthene	PAH	0.24	Ethanol, 2-nitro-, propionate (ester)	NIT	0.21
Fluorene	PAH	0.47	Pentanal	ALD	0.21
2-Propenal, 3-(4-hydroxy-3-methoxyphenyl)-	OxyPH	1.14	Furfural	ALD	0.24
4-((1E)-3-Hydroxy-1-propenyl)-2-methoxyphenol	OxyPH	2.34	Styrene	AR	0.25
Phenanthrene	PAH	1.83	1,2-Cyclopentanedione	KET	0.49
Anthracene	PAH	1.08	Oxazolidine, 2,2-diethyl-3-methyl-	NIT	0.33
p-Phenylbenzonitrile	NIT	0.25	1H-Indene, 1-chloro-2,3-dihydro-	AR	0.13
5H-Indeno[1,2-b]pyridine	NIT	0.58	1,2-Cyclopentanedione, 3-methyl-	KET	0.64
1H-Cyclopropa[1]phenanthrene, 1a,9b-dihydro-	AR	0.11	Phenol, 4-ethyl-2-methoxy-	OxyPH	0.27
Phenanthrene, 3-methyl-	PAH	0.14	2-Methoxy-4-vinylphenol	OxyPH	1.95
4H-Cyclopenta[def]phenanthrene	PAH	0.48	Vanillin	OxyPH	0.44
9,10-Anthracenedione	PAH	0.38	Biphenylene	AR	0.17
Cyclopenta(def)phenanthrenone	OxyAR	0.52	Apocynin	OxyPH	0.36
Pyrene	PAH	15.45	2-Propanone, 1-(4-hydroxy-3-methoxyphenyl)-	OxyPH	0.18
Phenaleno[1,9-bc]thiophene	SUL	0.16	Dibenzo[b,e]7,8-diazabicyclo[2.2.2]octa-2,5-diene	NIT	0.69
Benzo[b]naphtho[2,3-d]furan	FUR	3.58	Benzo[h]quinoline	NIT	0.27
Indeno(1,2,3-ij)isoquinoline	NIT	0.81	Anthracene, 9-methyl-	PAH	0.42
11H-Benzo[a]fluorene	PAH	0.66	Naphthalene, 2-phenyl-	PAH	0.32
Pyrene, 1-methyl-	PAH	3.22	n-Hexadecanoic acid	AC	0.20
11H-Benzo[b]fluorene	PAH	2.81	Fluoranthene	PAH	6.16
1H-Benzimidazole, 1-(1H-inden-2-yl)-	NIT	0.07	9-Anthracenecarbonitrile	NIT	0.33
4H-Benz[de]anthracene, 5,6-dihydro-	PAH	0.12	1-benzyl-3-methylnaphthalene	PAH	0.26
1,2,3,4-Tetrahydro-6-methylthio-9-acridinol	SUL	0.14	Pyrene, 4-methyl-	PAH	0.24
11H-Benzo[a]fluoren-11-one	KET	1.81	7H-Benz[de]anthracen-7-one	KET	0.55
Benzo[b]naphtho[2,3-d]thiophene	SUL	2.29	Triphenylene	PAH	2.69
Benzo[ghi]fluoranthene	PAH	0.81	11H-Indeno(1,2-b)quinoline	PAH	0.55
Benzo[c]phenanthrene	PAH	1.79	9H-Cyclopenta[a]pyrene	PAH	0.17
Benz[c]acridine	PAH	0.91	Benzo(a)pyrene-4,5-dione	PAH	0.70
Benz[a]anthracene	PAH	10.06	Benzo[ghi]perylene	PAH	0.13

Table S8. Composition of bio-oil derived from catalytic fast pyrolysis (with ZSM-5 zeolite as catalysts) of creosote-impregnated wood (NAS-3) in the Py/GC-MS system, at 600°C (GC-MS peak area, %).

<i>Compound</i>	<i>Group</i>	<i>%</i>	<i>Compound</i>	<i>Group</i>	<i>%</i>
1,3-Cyclohexadiene	ALI	0.06	Fluoranthene, 2-methyl-	PAH	0.29
Benzene	AR	3.20	11H-Benzo[b]fluorene	PAH	2.30
Toluene	AR	8.28	Pyrene, 2-methyl-	PAH	0.40
Ethylbenzene	AR	1.47	11H-Benzo[a]fluoren-11-one	KET	1.12
Benzene, 1,3-dimethyl-	AR	9.09	Benzo[b]naphtho[2,3-d]thiophene	SUL	1.19
p-Xylene	AR	0.96	Benzo[c]phenanthrene	PAH	1.11
Benzene, 1-ethyl-2-methyl-	AR	0.75	Benz[c]acridine	PAH	0.56
Benzene, 1,2,3-trimethyl-	AR	2.31	Benz[a]anthracene	PAH	4.75
3-Hydroxyphenylacetylene	PH	0.44	Benz[e]acephenanthrylene	PAH	1.59
Benzene, 1-ethenyl-2-methyl-	AR	0.29	Benzo[k]fluoranthene	PAH	0.23
Indane	AR	1.73	Benzo[j]fluoranthene	AR	0.22
Indene	AR	0.99	1H-Indenol	AL	0.16
1H-Indene, 2,3-dihydro-2-methyl-	AR	0.23	Phenanthrene, 9-methyl-	ALI	0.34
Phenol, 2-methoxy-	OxyPH	0.39	o-Xylene	AR	1.20
Benzofuran, 2-methyl-	FUR	0.41	Benzene, 1-ethyl-3-methyl-	AR	0.93
1H-Indene, 2,3-dihydro-5-methyl-	AR	0.94	1H-Indene, 1-chloro-2,3-dihydro-	AR	1.01
Phenol, 3-methyl-	PH	0.15	2-Methylindene	AR	1.60
1H-Indene, 3-methyl-	AR	1.38	1,1'-Biphenyl, 4-ethenyl-	AR	0.26
1H-Indene, 1-methyl-	AR	0.71	Benzofuran	FUR	0.46
Azulene	ALI	1.37	Benzofuran, 7-methyl-	FUR	0.35
Creosol	OxyPH	0.57	Benzofuran, 4,7-dimethyl-	FUR	0.17
1H-Indene, 1,3-dimethyl-	AR	0.78	6-Methoxy-3-methylbenzofuran	FUR	0.07
Naphthalene, 2-methyl-	PAH	4.39	Dibenzofuran	FUR	0.09
2-Methoxy-4-vinylphenol	OxyPH	0.56	9-Anthracenecarbonitrile	NIT	0.20
Naphthalene, 1-ethyl-	PAH	0.13	11H-Benzo[a]carbazole	NIT	0.14
Naphthalene, 1,3-dimethyl-	PAH	1.05	Benzene, 1-(1-butenyl)-4-methoxy-, trans-	OxyAR	0.12
Phenol, 2-methoxy-5-(1-propenyl)-, (E)-	OxyPH	0.15	Phenol, 2-methoxy-4-(1-propenyl)-, (Z)-	OxyPH	0.26
Biphenylene	AR	0.09	Naphthalene	PAH	1.56
Phenol, 2-methoxy-4-(1-propenyl)-	OxyPH	0.30	Naphthalene, 2-ethyl-	PAH	0.23
1-(4-Hydroxybenzylidene)acetone	OxyPH	0.14	Naphthalene, 2,6-dimethyl-	PAH	1.06
3(2H)-Benzofuranone, 2,4-dimethyl-	FUR	0.16	Naphthalene, 2,3-dimethyl-	PAH	0.12
Naphthalene, 1,4,5-trimethyl-	PAH	0.17	Acenaphthylene	PAH	0.10
Fluorene	PAH	1.01	Acenaphthene	PAH	0.09

9H-Fluoren-9-one	KET	0.23	Naphthalene, 2-(1-methylethyl)-	PAH	0.15
Anthracene	PAH	4.15	Phenanthrene, 3-methyl-	PAH	0.27
1H-Cyclopropa[1]phenanthrene, 1a,9b-dihydro-	AR	0.23	Naphthalene, 2-phenyl-	PAH	0.26
Anthracene, 9-methyl-	PAH	0.43	Fluoranthene	PAH	4.29
Phenanthrene, 2-methyl-	PAH	0.26	Pyrene, 1-methyl-	PAH	0.41
4H-Cyclopenta[def]phenanthrene	PAH	0.69	Pyrene, 4-methyl-	PAH	0.12
Anthracene, 1-methyl-	PAH	0.45	Triphenylene	PAH	1.60
Phenanthrene, 1-methyl-	PAH	0.10	Benz[a]anthracene, 7-methyl-	PAH	0.28
Indeno[2,1-a]indene, 5,10-dihydro-	AR	0.48	9H-Cyclopenta[a]pyrene	PAH	0.08
Cyclopenta(def)phenanthrenone	OxyAR	0.25	Perylene	PAH	0.32
Pyrene	PAH	12.14	1-Phenyl-1-butene	PH	0.28
Phenaleno[1,9-bc]thiophene	SUL	0.24	Phenol, 2-methyl-	PH	0.21
Benzo[b]naphtho[2,3-d]furan	FUR	2.28	p-Cresol	PH	0.09
Indeno(1,2,3-ij)isoquinoline	NIT	0.30	Phenol, 2,6-dimethyl-	PH	0.36
Indeno[2,1-b]chromene,	AR	0.10			