Supplementary material for:

Source apportionment of elevated wintertime PAHs by compound-specific radiocarbon analysis

Rebecca J. Sheesley*[*]*, Martin Kruså, Patricia Krecl, Christer Johansson and Örjan Gustafsson

Department of Applied Environmental Science (ITM), Stockholm University, 10691 Stockholm, Sweden

Contents:

5 pages

Table S1. Scheme of harvested PAH from particulate (GFF) and gas phase (PUF) extracts combined for CSRA.

	GFF			**PUF**		
Substance/sample	**23 Jan - 6 Feb**	**6 Feb -20 Feb**	**22 Feb -8 Mar**	**23 Jan- 31 Jan**	**31 Jan -6 Feb**	**µg C for CSRA**
Phenantrene PUF				**X**	**X**	**44.6**
Phenanthrene GFF	**X**		**X**			**29.3**
Fluoranthene	**X**	**X**	**X**			**65.8**
Pyrene	**X**	**X**	**X**			**63.4**
Benzo[b+k]fluoranthene composite	**X**	**X**	**X**			**57.3**
B[b]fluoranthene	X	X	X			
B[k]fluoranthene	X	X	X			
Indeno[cd]pyrene + benzo[ghi]perylene composite	**X**	**X**	**X**			**22.7**
Indeno[cd]pyrene		X				
Benzo[ghi]perylene	X	X	X			
Sum PAHs	**X**	**X**	**X**	**X**	**X**	**196.7**
Phenanthrene		X				
Anthracene	X	X	X	X	X	
2-Methylanthracene	X	X	X	X	X	
4H-Cyclopenta[def]-phenantrene plus 9-metylphenantrene	X	X	X	X	X	
1-Methylanthracene	X	X	X	X	X	
1-Metylphenantrene	X	X	X	X	X	
2-Phenylnaphtalene	X	X	X	X	X	
Fluoranthene				X	X	
Pyrene				X	X	
Benzo(a)flourene		X				
Benzo(e)flourene	X	X	X			
2-Methylpyrene	X	X	X			
X-Methylpyrene	X	X	X			
1-Metylpyrene	X	X	X			
Benzo[ghi]fluoranthene	X	X	X			
Benz[a]anthracene	X	X	X			
Chrysene	X	X	X			
Benzo[e]pyrene	X	X	X			
Benzo[a]pyrene	X	X	X			
Perylene	X	X	X			
Indeno[cd]pyrene	X		X			
Coronene	X	X	x			

Compounds in bold were harvested for CSRA, while compounds in normal type have been pooled for compound class-specific radiocarbon analysis (CCSRA).
GFF is glass fiber filter which collects particle-phase material.
PUF is polyurethane foam which collects gas-phase organic components.

Table S2. PAH concentrations (ng m^{-3}) in the particulate (GFF) and gas (PUF) phases measured in Lycksele in winter 2006.

PAH	GFF[a]	PUF[b]	GFF+PUF
Phenanthrene	2.26	8.97	11.3
Anthracene	0.15	1.02	1.17
2-Methylanthracene	0.03	0.05	0.08
1-Methylphenanthrene	0.34	0.33	0.68
2-Phenylnaphthalene	0.44	0.37	0.83
1,7-Dimethylphenanthrene	0.10	0.12	0.22
Fluoranthene	3.87	1.27	5.22
Pyrene	3.67	0.86	4.60
Retene	1.41	0.36	1.77
Benzo[a]fluorene	0.83	0.09	0.93
Benzo[b]fluorene	0.35	0.03	0.38
2-Methylpyrene	0.16	0.02	0.18
1-Methylpyrene	0.20	0.02	0.22
Benzo[ghi]fluoranthene	1.13	0.06	1.18
Cyclopenta[cd]pyrene	0.53	0.01	0.54
Benzo[a]anthracene	0.95	ND	0.95
Chrysene	2.79	0.07	2.83
Benzo[b]fluoranthene	2.72	ND	2.71
Benzo[k]fluoranthene	3.29	ND	3.25
Benzo[e]pyrene	1.16	ND	1.15
Benzo[a]pyrene	1.13	0.02	1.15
Perylene	0.13	ND	0.13
Indeno[1,2,3-cd]pyrene	4.19	ND	4.15
Pentacene	0.31	ND	0.31
Dibenz[a,h]anthracene	0.95	ND	0.94
Benzo[ghi]perylene	6.38	ND	6.32
Coronene	1.26	ND	1.26

[a] Mean concentration normalized by sample duration, 23 Jan. – 8 Mar. [b] Mean concentration normalized by sample duration, 23 Jan. – 6 Feb. ND: not determined.

Discussion of fossil contributions in Lycksele.

PAH/OC ratios from published gasoline and diesel emissions analysis (Riddle et al., 2007;Lough et al., 2007;Rogge et al., 1993) were used to calculate OC contributions based on the indeno[cd]pyrene plus benzo[ghi]perylene ambient concentrations (Table S3). These calculations illustrate two key points: PAH/OC ratios are quite variable and dependent upon vehicle conditions and the two gasoline references are the only ones which come close to the fossil TOC contribution as estimated by isotopic mass balance modeling. It should be noted that the ratios give an estimate of ambient OC not TOC, so these values would be expected to be biased low not high. Vehicle registration for Lycksele kommun, which includes both the city and surrounding area, indicates 86% gasoline 14% diesel (Granlund, H. and Fjällstedt, K., personal communication, 2009). These values give an indication of relative importance but still require input of source profiles in order to scale from number of vehicles to potential emission impacts. The combination of additional emission ratios for PAHs and vehicle registration numbers gives greater credence to the assertion that gasoline vehicle exhaust is dominant for fossil carbon in Lycksele, but does not prove that gasoline is the sole source of fossil PAH in Lycksele.

Table S3. Estimates of ambient fossil organic carbon based on published PAH/OC ratios for motor vehicle exhaust.

Ambient fossil concentration	**ng m^{-3}**	**µg m^{-3}**
I[cd]P+B[ghi]P	3.1	
TOC		0.7
Fossil sources		
	(I[cd]P+B[ghi]P)/OC	**Estimated OC [a]**
Diesel 2001	6.34E-06	489
HDD 1987	1.20E-05	258
Gasoline-2001	8.45E-04	4
Gasoline-1991	9.30E-04	3
idle/creep HDD 1999	3.30E-05	94
HDD-1998 ave	1.21E-05	257

[a] µg m^{-3}

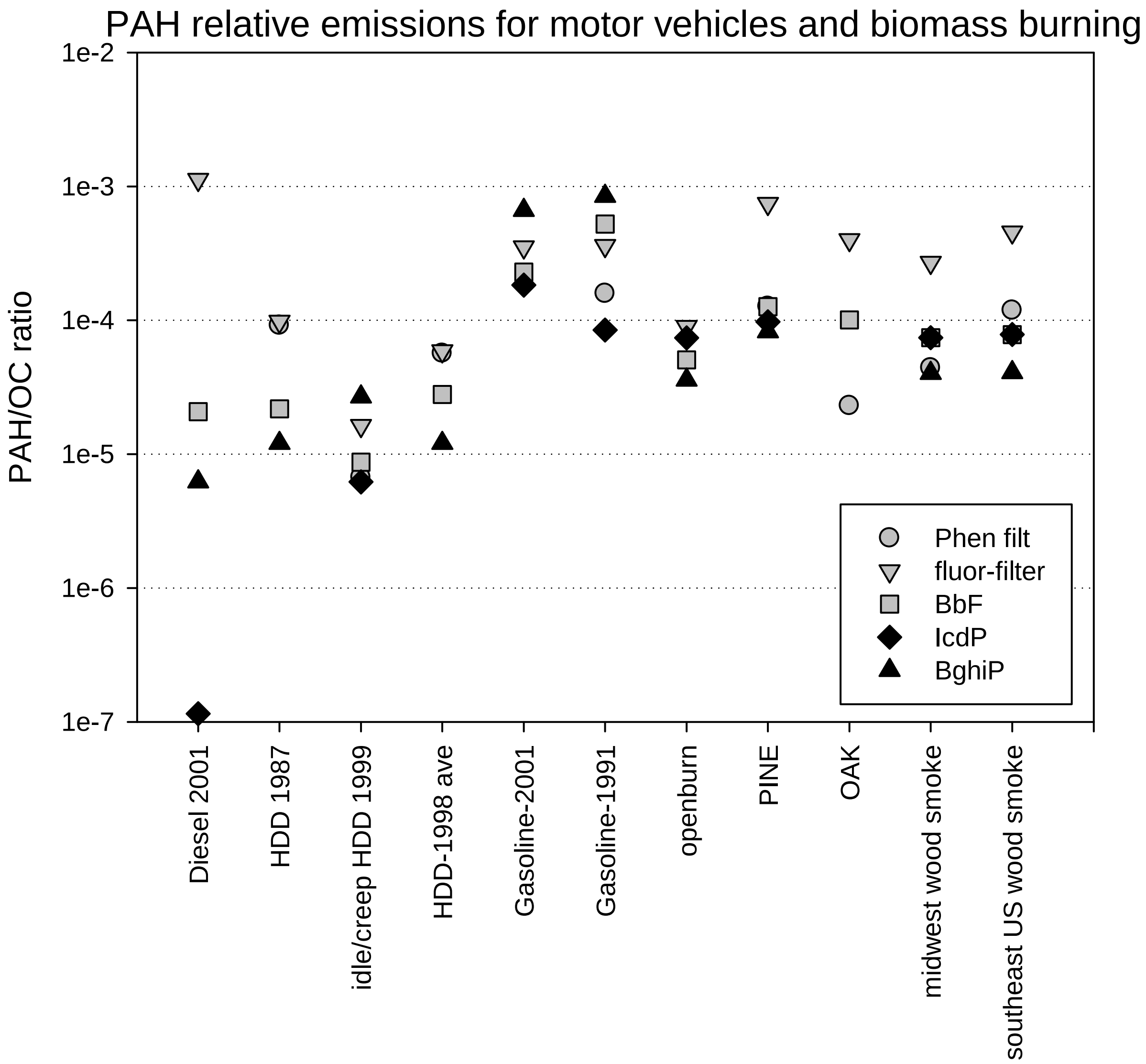

Figure S1. PAH/OC ratio for selected PAHs emitted by motor vehicles and biomass burning (OC is organic carbon as defined by the NIOSH 5040 method using the Sunset Labs thermal-optical instrument). For motor vehicle exhaust the date represents the vehicle year unless otherwise noted: diesel 2001 and gasoline 2001 (test year for averaged profile) exhaust values are from Lough et al. (2007); HDD 1987 (heavy duty diesel) and gasoline 1991 (test year) from Rogge et al (1993); HDD idle/creep 1999 and HDD 1998 are from Riddle et al (2007). It should be noted that for Riddle et al (2007), 3 other HDD vehicles emitted no detectable IcdP or BhiP. (For biomass burning and residential wood smoke openburn values represent average results from a prescribed field burn in Georgia, USA (Lee et al., 2005); pine and oak wood smoke profiles are from Schauer et al. (2001); Midwest and Southeast wood ratios are based on average profiles (Sheesley et al., 2007) for wood indigenous to the Midwest and Southeast regions in the USA (Fine et al., 2001, 2004, 2002).

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