# **Supporting Information**



Figure S1. Collecting samples from a sealed parking lot (left) and a sealed test plot (right).



**Figure S2.** Abraded particles from a coal-tar emulsion sealed parking lot, observed by light (left) and electron (right) microscropy. The particles abrade in a wide range of sizes. Particles of quartz sand, which are added to the sealant for added traction, are also visible. A single spherical particle (left figure) is probably from atmospheric deposition of combustion particles, which have a typically spherical shape (*S1*, *S2*), quite distinct from the parking lot particles. (Images courtesy of Irène Zimmerlin, Université de Rouen, France.)

#### Table S1. Concentrations of PAHs analysed in commercially-available sealant

products.

[All concentrations in mg/kg, dry weight; nd, non-detection; CT, coal-tar based emulsion; AS, asphalt-based emulsion.]

	Acenaph- thene	Acenaph- thylene	Anthra- cene	Benzo[a]- anthracene	Benzo[a]- pyrene	Benzo[b]- fluoranthene	Benzo[g,h,i]- perylene	Benzo[k]- fluoranthene
CT <sub>P</sub> 1	4000	nd	5150	4160	4600	4060	3190	2760
CT <sub>P</sub> 2	5380	nd	6770	5270	5980	5020	4040	3690
CT <sub>P</sub> 3	9230	nd	11800	9950	11100	9260	7660	6880
CT <sub>P</sub> 4	1590	nd	2020	1710	1840	1650	1280	1120
CT <sub>P</sub> 5	2050	nd	3050	2930	3080	2800	1830	1980
CT <sub>P</sub> 6	2560	nd	3810	3610	3690	2970	2250	2550
AS <sub>P</sub> 1	12.1	0.23	17	21.3	16.2	35.2	nd	nd
AS <sub>P</sub> 2	222	nd	364	422	347	314	140	297
AS <sub>P</sub> 3	53.9	nd	63.9	68	70.4	72.2	39.2	43.5

	Chrysene	Dibenz[a,h]- anthracene	Fluoran- thene	Fluorene	Indeno[1,2,3- cd]pyrene	Naphthalene	Phenan- threne	Pyrene
CT <sub>P</sub> 1	4160	nd	17500	3310	2470	8280	21200	13900
CT <sub>P</sub> 2	5310	nd	22900	4470	3170	10800	28600	17800
CT <sub>P</sub> 3	10100	670	41300	7860	5920	18500	49600	32200
CT <sub>P</sub> 4	1710	nd	6830	1340	1000	3350	8420	5240
CT <sub>P</sub> 5	3040	nd	8470	2010	1490	7630	10100	6630
CT <sub>P</sub> 6	3690	nd	10500	2510	1800	9630	12500	8120
AS <sub>P</sub> 1	22.2	nd	50.4	13	nd	39.9	60.7	49.6
AS <sub>P</sub> 2	427	nd	1510	266	135	241	1700	1100
AS <sub>P</sub> 3	71.5	nd	258	49.3	35.3	98.1	328	200

# **Table S2.** Yields of $\Sigma$ PAH and individual PAHs.

[All PAH yields in micrograms per square meter. nd, non-detection; CT, coal-tar emulsion sealant; AS, asphalt-emulsion sealant; UNSAS, unsealed asphalt pavement; UNSCON, unsealed concrete pavement. See manuscript Table 1 for additional site information.]

Site	Yield of sediment (g/m <sup>2</sup> )	ΣPAH yield	Naph- thalene	Fluorene	Phenan- threne	Anthra- cene	Fluoran- thene	Pyrene	Benz[ <i>a</i> ]- anthracene
CT <sub>TP</sub> 1	0.062	110	0.75	1.2	19	3.1	32	21	6.9
CT <sub>TP</sub> 2	0.051	61	0.33	.40	7.6	1.5	16	12	5.1
ASTP	0.067	6.4	nd	nd	.87	0.34	1.6	1.1	.62
$UNS_{TP}$	0.061	25	0.14	0.12	2.0	0.39	6.8	5.2	2.7
CT <sub>TP</sub> 1	0.026	14	.15	0.15	1.7	.26	4.2	2.9	1.0
CT <sub>TP</sub> 2	0.036	140	.86	1.2	17	2.8	40	31	13
ASTP	0.092	3.7	.05	0.06	.49	.09	1.0	0.64	0.17
UNS <sub>TP</sub>	0.069	1.7	nd	0.03	.15	.04	0.42	0.33	0.19
CT⊤⊳1	0.013	6.0	04	03	0.87	06	21	13	0.21
CT⊤⊳2	0.013	37	.0 <del>4</del> nd	.03	0.07	.00. 06	10	0.76	0.21
ASTP	0.020	1.6	nd	.0 <del>4</del> 00	0.01	.00 nd	0.35	0.70	0.37
UNS <sub>TP</sub>	0.036	0.50	nd	nd	0.05	nd	0.11	0.09	0.08
AS <sub>PL</sub> 1	0.30	77	1.4	1.1	8.3	1.6	22	12	4.4
$AS_{PL}1 dup$	0.30	71	1.2	.98	7.1	1.4	20	12	4.4
AS <sub>PL</sub> 2	0.32	270	1.5	.94	11	2.3	87	58	16
AS <sub>PL</sub> 3	1.2	920	nd	9.5	110	14	310	190	50
CT <sub>PL</sub> 1	0.11	220	1.9	1.6	31	2.3	63	50	12
CT <sub>PL</sub> 2	0.2	1,800	2.8	6.4	190	15	540	400	120
CT <sub>PL</sub> 3	0.97	1,900	6.5	4.5	67	14	510	410	180
CT <sub>PL</sub> 4	0.087	110	0.57	0.31	8.2	1.0	31	24	7.5
CT <sub>PL</sub> 5	0.24	130	0.41	0.54	5.1	1.2	34	24	10
CT <sub>PL</sub> 6	0.20	1,500	2.6	1.7	63	6.9	450	370	140
CT <sub>PL</sub> 6 dup	0.20	880	1.8	1.1	37	4.1	260	200	73
$UNSAS_{PL}1$	0.41	26	1.2	0.91	2.7	.87	5.8	5.0	3.0
UNSAS <sub>PL</sub> 1	0.46	3.3	nd	nd	0.33	nd	0.78	0.74	nd
UNSCON <sub>PL</sub> 1	0.21	16	0.32	0.28	1.2	0.32	3.8	3.2	1.4
UNSCON <sub>PL</sub> 2	0.18	12	0.20	0.18	0.98	0.23	3.0	2.5	1.0

### Supporting Information Table S2, cont.

		Benzo[ <i>a</i> ]-	Dibenz- [ <i>a,h</i> ]-	_
Site name	Chrysene	pyrene	anthracene	Coronene
CT <sub>TP</sub> 1	12	6.2	1.1	1.2
CT <sub>TP</sub> 2	9.1	5.1	1.1	1.3
AS <sub>TP</sub>	1.1	0.81	nd	0.49
$UNS_{TP}$	3.7	3.2	0.68	0.80
CT <sub>TD</sub> 1	18	11	0.37	0.31
	21	14	3.0	34
AS	0.55	0.35	0.15	0.7
AGTP	0.00	0.55	0.15	0.20
UNSTP	0.20	0.24	0.09	0.10
CT <sub>TP</sub> 1	1.0	0.37	0.08	0.13
CT <sub>TP</sub> 2	0.66	0.45	0.13	0.13
ASTP	0.22	0.36	nd	0.23
$UNS_{TP}$	0.07	0.12	nd	0.07
AS <sub>PI</sub> 1	15	6.2	3.0	4.4
AS <sub>PI</sub> 1 dup	14	6.5	2.9	3.6
AS <sub>PI</sub> 2	62	22	6.2	7.8
AS <sub>PL</sub> 3	140	66	28	34
	33	18	34	56
	300	160	34	40
	450	240	55	70
	-30 24	12	24	31
	24	12	2.4	J. <del>4</del> 4.4
	23	170	J.7 45	4. <del>4</del> 12
	270	100	40	40
CTPLO dup	100	100	10	22
$UNSAS_{PL}1$	2.8	2.9	nd	1.8
UNSAS <sub>PL</sub> 1	0.46	1.0	nd	0.69
UNSCON <sub>PL</sub> 1	2.1	1.6	0.76	0.68
UNSCON <sub>PL</sub> 2	1.7	1.3	0.50	0.61

# Supporting Information Table S3. Watershed characteristics.

			Percentage of Watershed						
Watershed	Latitude and longitude (deg min sec)	Area (km <sup>2</sup> )	Total urban	Resi- dential	Commer- cial	Indus -trial	Trans- portation (includes parking lots)	Parking lots (all types)	Sealed parking lot
Lake Como Inflow	N 32 43 45 W 97 24 08	1.86	93.3	51.7	10.5	2.21	28.9	7.58	1.19
Echo Lake Inflow	N 32 42 03 W 97 19 09	2.13	98.3	32.6	9.23	25.4	31.3	6.35	2.10
Fosdic Lake Inflow	N 32 45 10 W 97 15 27	0.84	96.7	61.2	16.1	0	19.4	2.87	1.03
Williamson Creek	N 30 13 16 W 97 47 36	78.0	63.4	38.6	8.0	2.2	14.6	4.5	1.9

#### References

- (S1) Griffen, J.J.; Goldberg, E.D. Sphericity as a characteristic of solids from fossil fuel burning in a Lake Michigan sediment. *Geochim. Cosmochim. Acta* 1981, 45, 763-769.
- (S2) Motelay-Massei, A. Contribution du compartiment atmosphérique au transfert des HAP dans le basin versant aval de la Seine: Bilan à l'échelle de deux basins versants expérimentaux. Ph.D. Dissertation, Université Paris VI, 2003.