

## Season variations of B(a)P concentrations

[Caricchia, Anna Maria, Salvatore Chiavarini, Massimo Pezza (1999): Polycyclic aromatic hydrocarbons in the urban atmospheric particulate matter in the city of Naples (Italy), *Atmospheric Environment*, 33 (1999), 3731-3738, doi:10.1016/S1352-2310(99)00199-5]

Data relative to the summer campaign (III) are lower than those relative to the autumn and winter campaigns, in all the three sites (Table 1). The ratios of autumn and winter to summer total PAH are in the range 1.5-4.5. This is in accordance with results reported in literature: winter levels, as stated in several investigations carried out in Europe and in the USA, are generally higher by a factor of 2-10 (Baek *et al.*, 1991a; Greenberg *et al.*, 1985; Harrison *et al.*, 1996; Menichini, 1992; Smith and Harrison, 1996). The higher urban PAH in winter with respect to warmer seasons are due to the contribution of domestic heating and to the different meteorological conditions that, during summer, cause PAH photodegradation and an easier atmospheric dispersion of pollutants.

**Table 1**

Site 3. Median (Med), average (Avg), minimum (Min) and maximum (Max) values of the compounds concentrations ( $\text{ng m}^{-3}$ ) in the urban atmospheric particulate

	I Sampling 24 September 1996–30 September 1996				II Sampling 26 February 1997–04 March 1997				III Sampling 25 June 1997–01 July 1997			
	Med	Avg	Min	Max	Med	Avg	Min	Max	Med	Avg	Min	Max
PHE	0.93	0.96	0.45	1.68	1.22	1.14	0.34	2.13	0.30	0.32	0.10	0.54
AN	0.09	0.09	0.05	0.16	0.22	0.19	0.06	0.32	0.09	0.17	0.04	0.66
FA	0.83	0.87	0.25	1.57	1.18	3.68	0.21	9.93	0.61	0.86	0.16	1.78
PY	1.11	1.17	0.32	2.12	1.38	5.07	0.24	12.96	0.81	1.12	0.20	2.37
BNT	0.04	0.04	0.02	0.06	0.20	0.20	0.01	0.51	0.01	0.02	0.01	0.04
CPP	0.01	0.02	0.01	0.05	5.98	4.63	0.01	19.94	0.01	0.39	0.01	1.38
BaA	1.59	1.34	0.44	2.04	1.90	3.17	0.13	8.01	0.76	0.96	0.10	2.11
CRY	3.24	2.67	1.15	3.83	4.19	5.21	0.30	12.75	1.56	1.78	0.23	3.34
B(b + j + k)F	7.78	7.98	3.64	12.80	7.52	8.96	0.99	20.06	5.99	4.73	0.66	7.93
BeP	6.10	5.95	2.97	8.34	5.22	6.59	0.77	13.84	4.11	3.38	0.53	5.79
BaP	1.53	1.38	0.41	2.14	3.13	5.13	0.09	12.28	1.23	1.75	0.09	4.02
IP	4.45	4.32	2.18	6.18	3.80	4.76	0.53	10.25	3.37	2.93	0.45	5.11
DBahA	0.46	0.47	0.21	0.78	0.46	0.50	0.05	1.20	0.42	0.34	0.04	0.60
BghiP	5.68	5.65	2.91	7.69	4.51	6.49	0.82	13.39	3.61	3.31	0.69	5.72
COR	2.21	2.25	1.26	3.19	2.26	3.69	0.41	8.85	1.67	1.86	0.41	3.54
Total	37.13	35.14	17.56	50.14	36.35	59.40	5.30	130.46	28.89	23.90	3.70	41.48

[Gianelle V., Cristina Colombi, Stefano Caserini, Senem Ozgen, Silvia Galante, Alessandro Marongiu, Guido Lanzani ( 2013) Benzo(a)pyrene air concentrations and emission inventory in Lombardy region, Italy, *Atmospheric Pollution Research*, 4, (2013), 257-266 doi:10.5094/APR.2013.028]

Residential wood combustion (RWC) (Belis *et al.*, 2011; Silibello *et al.*, 2012) and vehicular traffic (Slezakova *et al.*, 2010) has been found to be a significant source of B(a)P. Incomplete combustion in industrial processes (Ravindra *et al.*, 2008) and open burning of agricultural biomass (Hays *et al.*, 2005) are also responsible for B(a)P emissions. In addition to local sources, B(a)P may also reach monitoring area through long range transport of polluted air masses (Sato *et al.*, 2008). Many authors used marker compounds, diagnostic ratios, and statistical analyses to identify possible major B(a)P and PAH sources in the area of investigation (Ravindra *et al.*, 2008).

Although up to now no B(a)P emission inventory has been realized for Italy and Lombardy, a total-PAH emission inventory for Lombardy region has been compiled by ISPRA (ISPRA, 2011) for the years 1990, 1995, 2000 and 2005, through a top-down disaggregation of the national emission inventory. Dominant

sources result to be biomass and agricultural waste burning and metallurgical industries. The disadvantage of these inventories is that it is not always clear which PAH species has been included in emission data or emission factor (EF) during the compilation of the inventory (i.e., the four PAHs considered in CLRTAP, more complex and detailed mixtures, all PAH species or PAH with the exception of the 2-ring compounds similar to naphthalene). B(a)P emissions in the residential sector are difficult to assess because of the high uncertainty level connected with both the amount of wood combusted per appliance type at the local level and the corresponding emission factor. Furthermore, differences in measurement methods (i.e. consideration of condensable fraction) could lead to significantly different emission results (*Nussbaumer et al.*, 2008). Wood stoves constitute the most thoroughly examined appliance type (*US-EPA*, 1998; *Hedberg et al.*, 2002; *Kakareka et al.*, 2005; *Mc Donald et al.*, 2000; *Pettersson et al.*, 2011) whereas for closed (*US-EPA*, 1998; *Schauer et al.*, 2001; *Mc Donald et al.*, 2000) and open (*Houck and Eagle*, 2006; *Goncalves et al.*, 2011) fireplaces few studies considering only B(a)P in the particulate phase are available. Fuel type results to be an important factor influencing the emission levels to vary up to an order of magnitude between similar type of appliances fed with different fuels (e.g., pellets vs. wood) (*Boman et al.*, 2011; *Lamberg et al.*, 2011).

Fuel combustion in motor vehicles is recognized as being one of the major contributors to the atmospheric emissions of B(a)P in urban areas (*Slezakova et al.*, 2010). Many data on emissions and emission factors of B(a)P are available for gasoline- and dieselpowered passenger cars and light- and heavy-duty vehicles (*Eiguren-Fernandez and Miguel*, 2012) while only little published data are available on B(a)P emissions from two-wheelers (*Spezzano et al.*, 2008).

Open burning encompasses a large number of combustions in open air which determine toxics emissions, not only of B(a)P but also of other PAH and PCDD/F (*Lemieux et al.*, 2004; *US-EPA*, 1998).

Recent literature studies report average ambient B(a)P levels ranging between 10-2 and 101 ng m<sup>-3</sup> for urban environments with different local characteristics (see Table S1). **In spite of different sampling methods and reference PM fraction (TSP, PM<sub>10</sub> or PM<sub>2.5</sub>)**, it emerges from these studies that B(a)P generally follows the pattern of seasonal variations of ambient particulate matter, being higher in the winter and lower in the summer. B(a)P are reported to be more associated with fine particles than with coarse (*Saarnio et al.*, 2008).

Table S1. Literature review for PM-bound B(a)P concentrations (ng m<sup>-3</sup>) in ambient air

Location, Country	Type of site	PM fraction	Season			Reference
			annual	cold	warm	
Piacenza, Italy	urban background	PM <sub>2.5</sub>	0.1			ISPRA, 2011
Venezia, Italy	urban background	PM <sub>2.5</sub>	1.1			
Padova, Italy	urban traffic	PM <sub>2.5</sub>	1.3			
Milano, Italy	urban traffic	PM <sub>2.5</sub>	0.2			
Perugia, Italy	suburban traffic	PM <sub>2.5</sub>	0.6			
Taranto 1, Italy	suburban industrial	PM <sub>2.5</sub>	0.4			
Taranto 2, Italy	suburban industrial	PM <sub>2.5</sub>	1.3			
Trieste, Italy	urban industrial	PM <sub>2.5</sub>	0.7			
Verona, Italy	rural background	PM <sub>2.5</sub>	0.6			
Northern Italy	urban and rural	PM <sub>10</sub>		0.4-4		Belis et al., 2011
Milan, Italy	urban station	PM <sub>10</sub>		2.2		
Kozani, Greece	urban industrial	PM <sub>2.5</sub>	0.38			Evangelopoulos et al., 2010
Kozani, Greece	urban industrial	PM <sub>10</sub>	0.49			
Cantabria, Spain	urban	PM <sub>10</sub>	0.04-0.13			Arruti et al., 2012
Madrid, Spain	urban traffic	PM <sub>10</sub>	0.1			
Dettenhausen, Germany	urban background	PM <sub>10</sub>		1.62		Bari et al., 2009
Pais Vasco, Spain	urban	PM <sub>10</sub>	0.5			Barrero and Canton, 2007
Zaragoza, Spain	urban	PM <sub>10</sub>	0.5			Callen et al., 2008
London, UK	urban	PM <sub>10</sub>	1.4			Kendall et al., 2001
Rome, Italy	urban	PM <sub>10</sub>	1.1			Menichini et al. 2007
Duisburg, Germany	urban background	PM <sub>10</sub>		1.1		Saarnio et al., 2008
Prague, Czech Republic	urban background	PM <sub>10</sub>		3.15		
Amsterdam, Netherlands	urban background	PM <sub>10</sub>		0.35		
Helsinki, Finland	urban background	PM <sub>10</sub>			0.15	
Barcelona, Spain	urban background	PM <sub>10</sub>			0.08	
Athens, Greece	urban background	PM <sub>10</sub>			0.05	
Zagreb, Croatia	urban	PM <sub>10</sub>	1.2			Sisovic et al., 2008
Vinohrady, Finland	rural-coastal	PM <sub>10</sub>	0.21			Vestenius et al., 2011
Madrid, Spain	urban	PM <sub>10</sub>	0.2			Viana et al., 2008
Niembro, Spain	rural	PM <sub>10</sub>	0.014			MARM, 2010
Lazio, Italy	rural	PM <sub>10</sub>	0.02			Menichini et al. 2007
Athens, Greece	rural	TSP	0.02		0.02	Mandalakis et al., 2002
Zonguldak, Turkey	urban industrial	TSP		16.8	0.5	Akyuz and Cabuk, 2010
Belgrade 1, Serbia	urban traffic	TSP		6.53	1.18	Ovetkovic et al., 2010
Belgrade 2, Serbia	urban industrial	TSP		5.06	0.41	
Belgrade 3, Serbia	urban traffic	TSP		8.87	0.97	
Belgrade 4, Serbia	urban industrial	TSP		8.62	0.9	
Belgrade 5, Serbia	urban traffic	TSP		2.73	0.78	
Belgrade 6, Serbia	suburban traffic	TSP		4.09	0.73	
Canarias, Spain	urban traffic	TSP	0.3			Lopez et al., 2002
Athens, Greece	urban	TSP	0.2			Mandalakis et al., 2002
Montana, USA	rural	TSP	1 - 6.3			Ward et al., 2009
Zelzate, Belgium	industrial	n.d.		0.95	0.37	Vercauteren et al., 2011

Table S4. Monthly average concentrations for B(a)P and PM<sub>10</sub>

Name and ID of sampling site (see Figure 1 and Table 1)													
B(a)P (ng/m <sup>3</sup> )	Brescia	Casirate d'Adda	Darfo	Magenta	Mantova	Meda	Milano-Pascal	Milano-Senato	Moggio	Schivenoglia	Sondrio	Soresina	Varese
	UB1	RB1	UB2	UB3	UB4	UT1	UB5	UT2	RB2	RB3	UB6	ST1	UT3
January	2.70	2.40	6.20	1.10	1.47	3.94	0.51	0.43	0.24	1.26	3.53	1.61	1.55
February	1.21	1.61	3.41	0.76	0.99	2.63	0.29	0.36	0.19	0.92	1.75	1.36	0.75
March	0.40	0.48	1.16	0.18	0.36	0.59	0.11	0.11	0.16	0.36	0.41	0.34	0.25
April	0.15	0.11	0.28	0.07	0.15	0.23	0.05	0.05	0.04	0.06	0.08	0.09	0.10
May	0.07	0.14	0.07	0.03	0.04	0.10	0.03	0.03	0.07	0.23	0.04	0.03	0.04
June	0.29	0.03	0.04	0.02	0.02	0.03	0.02	0.03	0.12	0.02	0.03	0.04	0.02
July	0.02	0.06	0.02	0.03	0.04	0.03	0.02	0.08	0.03	0.03	0.02	0.04	0.04
August	0.04	0.05	0.02	0.02	0.04	0.03	0.01	0.04	0.04	0.03	0.03	0.05	0.03
September	0.04	0.08	0.22	0.06	0.08	0.09	0.03	0.05	0.07	0.06	0.06	0.06	0.03
October	0.40	0.50	1.05	0.24	0.31	0.70	0.14	0.15	0.07	0.32	0.53	0.42	0.14
November	0.90	1.30	3.80	0.76	0.66	2.33	0.44	3.22	0.08	0.53	2.39	1.16	0.56
December	2.87	1.52	6.85	0.99	1.20	3.94	0.75	0.55	0.25	1.22	2.60	1.91	1.22
Year	0.78	0.68	1.94	0.36	0.43	1.23	0.20	0.46	0.10	0.39	0.99	0.59	0.36

Name and ID of sampling site (see Figure 1 and Table 1)													
PM <sub>10</sub> (µg/m <sup>3</sup> )	Brescia	Casirate d'Adda	Darfo	Magenta	Mantova	Meda	Milano-Pascal	Milano-Senato	Moggio	Schivenoglia	Sondrio	Soresina	Varese
	UB1	RB1	UB2	UB3	UB4	UT1	UB5	UT2	RB2	RB3	UB6	ST1	UT3
January	74	63	70	69	50	85	67	72	10	49	60	56	44
February	66	65	62	71	60	76	62	68	15	53	51	64	46
March	47	42	49	49	41	48	40	43	17	37	41	45	36
April	30	31	31	29	31	30	30	33	18	26	22	32	22
May	28	27	27	28	27	26	27	28	15	26	21	32	20
June	26	27	23	27	24	23	24	25	21	28	22	23	18
July	24	22	24	30	25	23	28	34	21	33	22	25	19
August	24	21	22	24	23	19	22	25	16	21	19	26	14
September	25	25	25	30	29	25	36	33	19	29	17	31	21
October	42	56	47	60	46	49	61	59	22	43	27	56	33
November	46	42	45	54	35	51	51	50	7	33	43	44	31
December	55	50	54	56	40	66	56	58	8	38	45	50	35
Year	41	38	40	44	36	44	42	46	16	35	33	40	28

## Wood burning as one of the most significant source of B(a)P

[Gianelle V., Cristina Colombi, Stefano Caserini, Senem Ozgen, Silvia Galante, Alessandro Marongiu, Guido Lanzani ( 2013) Benzo(a)pyrene air concentrations and emission inventory in Lombardy region, Italy, Atmospheric Pollution Research, 4, (2013), 257-266 doi:10.5094/APR.2013.028]

B(a)P emissions estimated for 2008 in the regional emission inventory of Lombardy are listed in Table 3 for the main emission sources. RWC in small appliances results to be the major source of B(a)P in the region accounting for the 77% (2.9t y<sup>-1</sup>) of the total emissions (3.8 t y<sup>-1</sup>). In particular, traditional wood stoves, closed fireplaces and open fireplaces are the major contributors accounting respectively for 32%, 23% and 19%. Other important sources are the combustion of wood and lignocellulosic biomass in small industrial boilers (8% of the total) and uncontrolled open burning of agricultural residues (5%). B(a)P emission from diesel vehicles is of secondary importance (2% of total emissions).

Table 3. Emission of B(a)P in Lombardy, year 2008

SNAP code	Source description	B(a)P (kg year <sup>-1</sup> )	(%)	Cumulative (%)
2.2.7	Residential heating, traditional stove-wood	1 185	32%	32%
2.2.8	Residential heating, closed fireplaces-wood	853	23%	54%
2.2.6	Residential heating, open fireplaces-wood	708	19%	73%
3.1	Industrial combustion-biomass	282	7.5%	81%
10.3.1	Open burning of agriculture residue	204	5.4%	86%
2.01	Institutional and commercial heating, pizza oven-wood	110	2.9%	89%
7	Road transport-diesel	87	2.3%	91%
2.2.10	Residential heating, automatic pellets stove-wood	75	2.0%	93%
2.2.9	Residential heating, innovative stove-wood	58	1.6%	95%
3.1.3	Industrial combustion-coal	58	1.5%	96%
3.3.10	Secondary aluminum smelting	35	0.9%	97%
11.3.1	Forest fires	31	0.8%	98%
3.1.3	Industrial combustion-fuel oil	23	0.6%	99%
8	Other machinery-diesel	11	0.30%	99%
4.1.2	Fluid catalytic cracking	10	0.26%	99%
7	Road transport-gasoline	8.2	0.22%	100%
7	Tire and break wear	7.6	0.20%	100%
4.2	Steel production	4.9	0.13%	100%
4.2	Other metallurgical processes	1.9	0.05%	100%
9.2.2	Industrial waste incineration	1.4	0.04%	100%
2.2.2	Residential heating, small boilers-gas oil	1.2	0.03%	100%
1.2.3	Energy production, district heating-biomass	0.56	0.01%	100%
8	Off road transport-diesel	0.34	0.01%	100%
2.1.3	Institutional and commercial heating, small boilers-gas oil	0.20	0.01%	100%
1	Energy production-natural gas	0.16	0.004%	100%
3.3.7	Secondary lead smelting	0.12	0.003%	100%
2.2.2	Residential heating, small boilers-natural gas	0.12	0.003%	100%
2.1.3	Institutional and commercial heating, small boilers-natural gas	0.035	0.001%	100%
9.7.0	Agriculture waste incineration	0.016	0.000%	100%
7	Road transport-LPG	0.0079	0.000%	100%
9.2.1	Municipal solid waste incineration	0.0076	0.000%	100%
7	Road transport-natural gas	0.00023	0.000%	100%
	TOTAL	3 758	100%	

Regarding the spatial distribution of the emissions, Table 4 shows B(a)P and PM10 emission estimates for 2008 together with the contribution of different sources, the pro-capita emissions and emission densities for a 10x10 km<sup>2</sup> area centered on every monitoring site, as well as B(a)P/PM10 ratios. The contribution of RWC is more than 80% in 8 monitoring sites out of 13. In Mantova, a forest fire has been

registered by the INEMAR Emission Inventory in 2008 determining 6% of the total emission of B(a)P. This type of uncontrolled and unpredictable events can cause higher level of uncertainties in the emission inventory at the local scale.

The most relevant contribution of industrial sources (25%) was assessed for the station of UB2–Darfo, due to the presence of industrial biomass combustion (10%) and of the aluminium industry (15%). Two sites of Milan (UB5 and UT2) presented a completely different pattern, since RWC is less common, and therefore the pro-capita emission diminishes by a factor of ten. For this reason, in Milan the contribution of other emission sources increases, in particular the biomass burning in pizzerias (52–54%) and road transport (19%).

Table 4. B(a)P, PM<sub>10</sub> emissions and BaP/PM<sub>10</sub> emission ratios in 2008 in the municipalities interested by a 10 x 10 km<sup>2</sup> area centered on the sampling site

Sampling site	Inhabit.	Num. of municipalities	Total emission B(a)P (kg)	Total emission PM <sub>10</sub> (t)	Residential heating - wood B(a)P	Residential heating - wood PM <sub>10</sub>	Residential heating (excluding wood) B(a)P	Residential heating (excluding wood) PM <sub>10</sub>	Road transport B(a)P	Road transport PM <sub>10</sub>
UB1 Brescia	220 067	5	33	494	79%	21%	0.0%	0.1%	8.3%	35%
RB1 Casirate d'Adda	77 244	9	18	138	87%	40%	0.0%	0.5%	3.8%	33%
UB2 Darfo	35 212	7	12	80	66%	37%	0.0%	0.5%	3.8%	35%
UB3 Magenta	86 175	12	21	185	82%	35%	0.0%	0.4%	7.8%	51%
UB4 Mantova	81 500	4	13	229	76%	18%	0.1%	0.3%	7.7%	28%
UT1 Meda	258 669	15	44	337	85%	42%	0.1%	0.8%	4.5%	36%
UB5 Milano - via Pascal	1 377 482	4	32	850	20%	3%	1.5%	4.1%	19%	55%
UT2 Milano - via Senato	1 308 735	1	29	774	17%	3%	1.7%	4.4%	19%	55%
RB2 Moggio	9 895	9	22	92	96%	85%	0.1%	1.2%	0.6%	9%
RB3 Schivenoglia	16 837	8	11	74	92%	56%	0.0%	0.4%	1.5%	14%
UB6 Sondrio	33 075	8	29	140	94%	76%	0.1%	1.5%	1.0%	13%
ST1 Soresina	20 373	9	15	93	94%	58%	0.0%	0.2%	1.7%	17%
UT3 Varese	156 428	16	56	331	92%	58%	0.0%	0.6%	2.6%	29%

## SPAIN

### Season variations of B(a)P concentrations

[Villanueva 2015 Characterization of particulate polycyclic aromatic hydrocarbons in an urban atmosphere of central-southern Spain]

Benzo[a]-pyrene concentrations ranged from 2.4 to 110 pg/m<sup>3</sup>, these values are lower than the target value proposed by the European legislation, 1 ng/m<sup>3</sup>.

The results fit with a seasonal pattern with the maximum concentration in winter and the lowest in summer, due to the increment of domestic heating PAH emissions in the former and the temperature and solar intensity characteristics of the cold seasons. These data are corroborated from the analysis of the diurnal variations in PAH concentrations. In spring, the highest concentrations are found at 7:00–10:30 h, which reveals a significant contribution of traffic emissions. In winter, the most elevated concentrations are found in the 19:00–23:00-h period, due to domestic heating emissions and climatic conditions.



**Table 1** Carcinogenic and mutagenic potencies, mean concentrations ( $\text{pg m}^{-3}$ ) of PAHs and meteorological parameters at Ciudad Real city  $\pm$  relative standard deviations over the whole period and each season

Compound	TEFs <sup>a</sup>	MEFs <sup>b</sup>	Autumn	Winter	Spring	Summer
Phenanthrene	0.001	-	22 $\pm$ 11	51 $\pm$ 10	5 $\pm$ 3	31 $\pm$ 19
Anthracene	0.01	-	-	33 $\pm$ 46	27 $\pm$ 19	-
Fluoranthene	0.00	-	33 $\pm$ 8	74 $\pm$ 25	36 $\pm$ 5	26 $\pm$ 13
Pyrene	0.001	-	50 $\pm$ 19	126 $\pm$ 12	50 $\pm$ 19	12.5 $\pm$ 0.9
Benzo[a]anthracene	0.1	0.082	22 $\pm$ 6	54 $\pm$ 30	23 $\pm$ 3	23 $\pm$ 18
Chrysene	0.01	0.017	48 $\pm$ 14	108 $\pm$ 24	32 $\pm$ 6	26 $\pm$ 14
Benzo[b]fluoranthene	0.1	0.25	48 $\pm$ 17	97 $\pm$ 24	41 $\pm$ 11	21 $\pm$ 11
Benzo[k]fluoranthene	0.1	0.11	18 $\pm$ 5	45 $\pm$ 22	19 $\pm$ 10	8 $\pm$ 5
Benzo[a]pyrene	1	1.00	14 $\pm$ 7	35 $\pm$ 50	16 $\pm$ 12	14 $\pm$ 11
Dibenzo[a,h]anthracene	1	0.29	49 $\pm$ 22	122 $\pm$ 54	25 $\pm$ 8	30 $\pm$ 19
Benzo[g,h,i]perylene	0.01	0.19	48 $\pm$ 32	80 $\pm$ 27	72 $\pm$ 8	38 $\pm$ 20
Indeno[1,2,3-c,d]pyrene	0.1	0.31	30 $\pm$ 13	64 $\pm$ 22	24 $\pm$ 9	30 $\pm$ 19
<i>PAH</i> <sup>d</sup>			35 $\pm$ 14	74 $\pm$ 33	31 $\pm$ 18	24 $\pm$ 9
$\Sigma$ PAH			382	888	368	259
$\Sigma$ B[a]P <sub>TEQ</sub>			75.7	185.5	53.0	53.6
$\Sigma$ B[a]P <sub>MEQ</sub>			63.31	141.08	59.02	48.10
Temperature/ $^{\circ}\text{C}$			6 $\pm$ 2	8 $\pm$ 1	18 $\pm$ 3	28.9 $\pm$ 0.5
Rainfall/ $\text{L m}^{-2}$			0.01 $\pm$ 0.02	0.07 $\pm$ 0.1	0.0 $\pm$ 0.0	0.0 $\pm$ 0.0
Wind speed/ $\text{m s}^{-1}$			1.4 $\pm$ 0.6	3 $\pm$ 1	1.6 $\pm$ 0.7	1.6 $\pm$ 0.4
Humidity/%			85 $\pm$ 6	86 $\pm$ 8	51 $\pm$ 6	28 $\pm$ 6
Solar intensity/ $\text{W m}^{-2}$			68 $\pm$ 27	104 $\pm$ 57	285 $\pm$ 26	316 $\pm$ 8

<sup>a</sup>Toxic equivalency factors for cancer potency relative to BaP (Nisbet and LaGoy 1992)

<sup>b</sup>Mutagenic equivalency factors from Durant et al. (1996)

<sup>c</sup>Concentrations under the LOQ values

<sup>d</sup>Anthracene concentrations have not been included

[San José, Roberto, Juan Luis Pérez, María Soledad Callén, José Manuel López, Ana Mastral (2013): B(a)P (PAH) air quality modelling exercise over Zaragoza (Spain) using an adapted version of WRF-CMAQ model, Environmental Pollution xxx (2013), 1-8, <http://dx.doi.org/10.1016/j.envpol.2013.02.025>]

Table 2 shows the experimental B(a)P concentrations obtained for the sampling campaign carried out in Zaragoza city during 12 weeks.

The study reproduces the degree of seasonality of the B(a)P, with higher concentrations in the winter months. This is particularly prevalent at urban locations, where domestic combustion is the major source. In the rest of areas, for example industrial zones, concentrations are affected by other meteorological parameters, temperature, boundary layer, wind speed and direction.

**Table 2**

Mean and standard deviations (SD) of the PM10 ( $\mu\text{g}/\text{m}^3$ ), BaP ( $\text{ng}/\text{m}^3$ ) and meteorological conditions for the whole sampling campaign, the cold and the warm seasons ( $N$  = number of samples; cold season = 21st September–20th March; warm season = 21st March–20th September).

	Whole period	SD	Cold season	SD	Warm season	SD
PM10	34.37	28.98	38.17	37.40	30.56	16.48
BaP	0.090	0.110	0.150	0.125	0.030	0.036
Wind direction ( $^{\circ}$ )	216.2	73.8	212.5	81.8	220.0	65.6
Rainfall (mm)	0.05	0.17	0.04	0.14	0.06	0.19
Temperature ( $^{\circ}\text{C}$ )	13.7	7.9	8.3	5.6	19.1	6.0
Relative humidity (%)	65.2	13.7	71.1	14.4	59.3	10.0
Solar radiation ( $\text{W}/\text{m}^2$ )	196.5	97.0	129.1	61.3	263.8	77.5
Wind speed (m/s)	3.4	2.5	3.4	2.4	3.4	2.7
$N$	84		42		42	

[Callen, M.S., J.M. Lypez, A.M. Mastral (2010) Seasonal variation of benzo(a)pyrene in the Spanish airborne PM<sub>10</sub>. Multivariate linear regression model applied to estimate B(a)P concentrations, *Journal of Hazardous Materials* 180 (2010) 648–655, doi:10.1016/j.jhazmat.2010.04.085]

One of the main objectives of this work is to assess the B(a)P concentrations in different localizations in Aragyn with regard to the fulfilling of the Directive 2004/107/EC [11] related to PAH in air. Independently of the sampling point, the highest B(a)P concentrations were obtained in the cold season. This trend was already observed in different campaigns carried out in ZGZ where the low temperatures favoured the PAH accumulation [12,13,17]. In addition, there are additional anthropogenic activities which show seasonality, one of them is the residential heating that during warm season does not contribute to PAH formation.

Zaragoza was the sampling point with the highest B(a)P concentrations in both, warm (BaPmean = 0.089 ng/m<sup>3</sup>) and cold seasons (BaPmean = 0.500 ng/m<sup>3</sup>), exceeding the guideline value of B(a)P (1.0 ng/m<sup>3</sup>) established by the Directive 2004/107/EC [11] twice in the cold season (13%) as well as the upper (0.6 ng/m<sup>3</sup>; 27%) and the lower assessment thresholds (0.4 ng/m<sup>3</sup>; 33%). In fact, the mean B(a)P concentration during the cold season also exceeded the lower assessment threshold of B(a)P. Most of these B(a)P exceedances were produced during San Antyn (bonfire festival), obtaining the maximum B(a)P concentrations for those dates.

With regard to MON, this place also showed B(a)P concentrations exceeding the quantification limit for all samples, with higher concentrations during the cold season (BaPmean = 0.223 ng/m<sup>3</sup>) versus the warm season (BaPmean = 0.050 ng/m<sup>3</sup>) (Fig. 2c and d). Nevertheless, the guideline value of 1.0 ng/m<sup>3</sup> and the upper and lower assessment thresholds established by the Directive 2004/107/EC [11] were not exceeded.

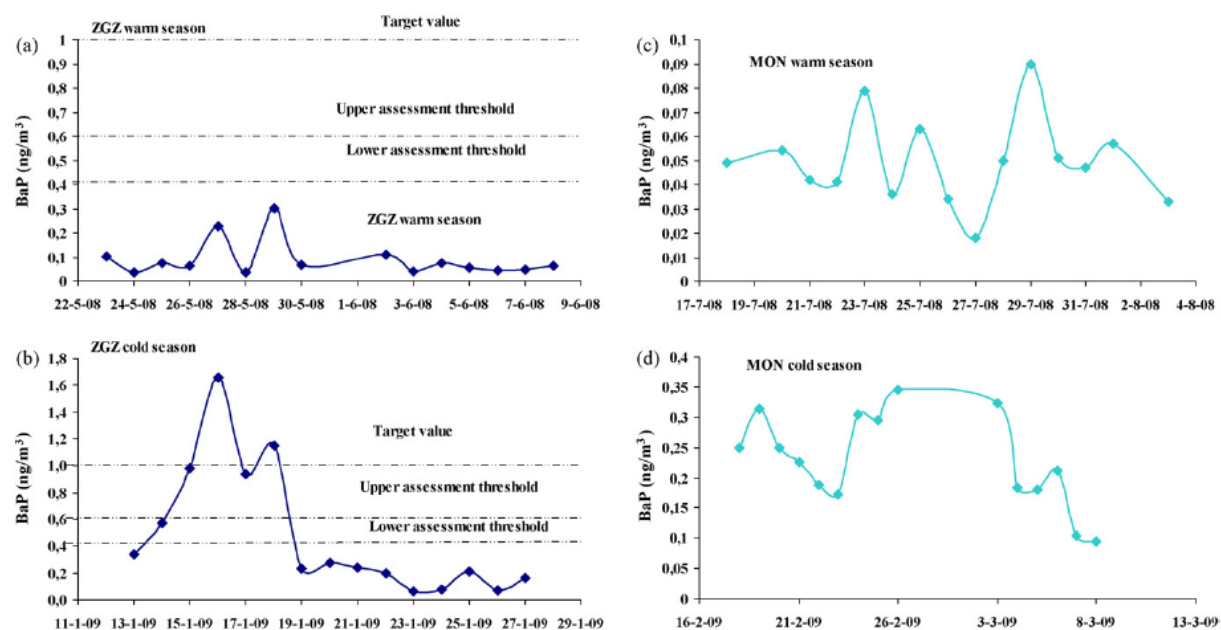


Fig. 2. Temporal evolution of BaP for ZGZ during the a) warm and b) cold seasons and for MON during the c) warm and d) cold seasons.

In PIR and MNG, the B(a)P concentrations were almost negligible with values lower than the detection and/or quantification limits for most of the samples (PIR: BaPmeancold = 0.020 ng/m<sup>3</sup>, BaPmeanwarm = 0.013 ng/m<sup>3</sup>; MNG: BaPmeancold = 0.021 ng/m<sup>3</sup>, BaPmeanwarm = 0.024 ng/m<sup>3</sup>). These minimum concentrations are predictable by considering that PIR is localised in the Pyrenees Mountain where the anthropogenic contributions are minimum. Regarding MNG, a rural background area, minimum B(a)P concentrations were also detected despite the possible contribution of anthropogenic sources related to a power station and to long-range transport. For the four sampled places, ZGZ showed a higher pollution level than MON.

Both localizations showed urban profiles with higher B(a)P concentrations during cold season in which local anthropogenic activities were responsible for most of this pollutant.

This is expected by considering that the main PAH pollution sources are related to big cities involving different anthropogenic activities such as transport, industry, residential heating, etc.

However, the mean B(a)P concentration for the two periods was  $0.284 \text{ ng/m}^3$ , lower than the lower assessment threshold of B(a)P,  $0.4 \text{ ng/m}^3$ .

## Wood burning as one of the most significant source of B(a)P

[San José, Roberto, Juan Luis Pérez, María Soledad Callén, José Manuel López, Ana Mastral (2013): B(a)P (PAH) air quality modelling exercise over Zaragoza (Spain) using an adapted version of WRF-CMAQ model Environ Pollut. 2013 Dec;183:151-8. doi: 10.1016/j.envpol.2013.02.025]

The B(a)P emissions are broken down into the SNAP 12 categories, Table 1. Road transport and Non-Industrial combustion are the dominant source. In case of high resolution domains (1 km, 3 km and 9 km), a top-down process scheme has been used to go from the coarse resolution to the desired resolution. The downscaling process is based on surrogates to allocate the emission into the model grid.

Different surrogates are used for different SNAP's activities, Table 1.

**Table 1**  
Weights and surrogates for the emission snap activities.

Snap activities	Weight	Surrogates
Combustion in energy	15.0%	Industrial areas
Non-industrial combustion	23.0%	Buildings
Combustion in manufacturing	15.0%	Industrial areas
Production processes	5.10%	Industrial areas
Extraction of fossil fuels	8.40%	Mineral extraction
Solvent use	0.50%	Industrial areas
Road transport	29.7%	Roads and streets
Other mobile sources	0.10%	Roads
Waste treatment	3.00%	Waste areas
Agriculture	0.20%	Natural areas

## POLAND, GDYNIA, (SOUTHERN BALTIC SEA)

### Season variations of B(a)P concentrations

[Staniszewska et.al, 2013 Factors controlling benzo(a)pyrene concentration in aerosols in the urbanized coastal zone. A case study: Gdynia, Poland (Southern Baltic Sea)]

An annual average concentration of benzo(a)pyrene in aerosols in the studied area exceeded the value ( $1 \text{ ng/m}^3$ ) acceptable for the EU countries (Directive 2004/107/WE; Table 2). At the same time, a clear seasonality (Table 2) and large intra-monthly variability of benzo(a)pyrene concentration were observed. During the heating season (17.XII.07–30.IV.08; 1.X.08–17.XII.08) both average and median concentrations clearly exceeded the values observed in the non-heating season (1.V.08–30.IX.08) (Table 2). Over the most of the heating season, the benzo(a)pyrene concentration was higher than  $1 \text{ ng/m}^3$ . But actually there is no reference point to our results. The ambient air quality standards determine only the value of the acceptable annual average benzo(a)pyrene concentration. The utility of such an indicator seems to be questionable considering continuous exposure to high concentrations within a few months of the year. Therefore, determination of the acceptable daily or monthly average concentration of benzo(a)pyrene would be very valuable.



**Table 2** Basic statistics of BaP concentrations ( $\text{ng m}^{-3}$ ) in aerosols during measurements performed in Gdynia (17.XII.2007–17.XII.2008)

Variable	Sampling period				
	(17.XII.07–17.XII.08)	(17.XII.07–30.IV.08; 1.X.08–17.XII.08) heating season	(1.V.08–30.IX.08) non-heating season	Weekends	Workdays
Number of measurements	153	89	64	58	94
Average concentrations $\pm$ standard deviation	$1.29 \pm 2.75$	$2.18 \pm 3.33$	$0.05 \pm 0.09$	$1.94 \pm 4.72$	$0.91 \pm 1.51$
Min.-max.	n.d.– 25.20	0.11–25.20	n.d.–0.36	n.d.– 25.20	n.d.– 8.26
Median concentrations	0.34	1.10	< LOD	0.52	0.30
(Lower–upper quartile)	< LOD– 1.25	0.52–2.36	< LOD– 0.07	0.06–2.05	< LOD– 1.22

LOD limit of detection

Combustion processes, especially domestic heating of both local and regional origin, were identified as the main sources of benzo(a)pyrene in study area. As a result, B(a)P concentrations during the heating season were significantly higher than those observed in the non-heating season. Strong seasonal variability suggests that the establishing of monthly acceptable level of concentration of benzo(a)pyrene would be valuable.

[Barbara Kozielska,\*, Wioletta Rogula-Kozłowska and Krzysztof Klejnowski [2015] Seasonal Variations in Health Hazards from Polycyclic Aromatic Hydrocarbons Bound to Submicrometer Particles at Three Characteristic Sites in the Heavily Polluted Polish Region, Atmosphere, 2015, 6, 1-20; doi:10.3390/atmos6010001]

The ambient concentration of the PM<sub>10</sub>-bound B(a)P (B(a)P bound to particles not greater than 10  $\mu\text{m}$ ) has the limit that should not be exceeded; the yearly permissible ambient concentration of PM<sub>10</sub>-bound B(a)P is 1  $\text{ng/m}^3$  [24,25].

Consequently, the mean PM<sub>1</sub>-bound B(a)P concentrations in RB, UB and UT were very high. The values were 3.24  $\text{ng}\cdot\text{m}^{-3}$  (4.03  $\text{ng/m}^3$  - Heating season; 2.46  $\text{ng/m}^3$  - Non-heating season), 7.72  $\text{ng/m}^3$  (12.48  $\text{ng/m}^3$  - Heating season; 2.97  $\text{ng/m}^3$  - Non-heating season) and 9.5  $\text{ng/m}^3$  (14.27  $\text{ng/m}^3$  - Heating season; 4.73  $\text{ng/m}^3$  - Non-heating season) in RB, UB and UT, respectively. Such a finding shows that even though RB was located far away from the anthropogenic emission sources, the B(a)P and PM<sub>1</sub>-bound B(a)P concentrations exceeded the permissible level more than three times there. In UB in the densely developed and populated area, the permissible B(a)P value was exceeded nearly eight times, whereas the value was almost 10 times higher in UT. High B(a)P concentrations were observed both in the heating season and the remaining part of the year. Such a situation was not observed in other European regions even though the permissible mean yearly values were also exceeded in the Czech cities. Nonetheless, the PM<sub>1</sub>-bound B(a)P concentrations did not exceed 0.39  $\text{ng/m}^3$  [14].

The research conducted at the roadside and urban background in Madrid (Spain) showed that the B(a)P concentration in winter was 0.24  $\text{ng/m}^3$  and 0.054  $\text{ng/m}^3$ , respectively. In summer, the B(a)P concentrations observed in Madrid (Spain) were even lower [26]. On the other hand, the B(a)P concentrations as high as those observed in RB, UB and UT are typical for cities in southern Poland and are observed every year. For example, in the urban background in Zabrze (a city 15 km away from Katowice), the mean concentration of the PM<sub>1</sub>-bound B(a)P was 16  $\text{ng/m}^3$  in winter 2007 [27].

**Table 1.** The statistics of 24-h concentrations of PM1 ( $\mu\text{g}/\text{m}^3$ ) and PM1-bound PAHs ( $\text{ng}/\text{m}^3$ ) at three locations in two measurement periods.

	Regional Background (RB)						Urban Background (UB)						Traffic Point (UT)					
	Heating Season (N = 20)			Non-Heating Season (N = 20)			Heating Season (N = 20)			Non-Heating Season (N = 20)			Heating Season (N = 20)			Non-Heating season (N = 20)		
	Min Max	Avg $\pm$ SD	50% *	Min Max	Avg $\pm$ SD	50%	Min Max	Avg $\pm$ SD	50%	Min Max	Avg $\pm$ SD	50%	Min Max	Avg $\pm$ SD	50%	Min Max	Avg $\pm$ SD	50%
BkF	0 3.09	1.24 $\pm$ 0.92	1.18	0 2.98	0.68 $\pm$ 0.96	0.22	3.22 26.09	10.74 $\pm$ 5.21	9.51	0 7.13	1.87 $\pm$ 1.58	1.56	1.79 34.51	13.80 $\pm$ 7.77	11.97	0 6.01	0.97 $\pm$ 1.38	0.68
BaP	0.80 28.23	4.03 $\pm$ 5.97	2.40	0 9.52	2.46 $\pm$ 2.61	1.51	4.52 32.28	12.48 $\pm$ 6.36	11.29	0.41 5.14	2.97 $\pm$ 1.39	3.15	1.48 46.70	14.27 $\pm$ 11.27	11.87	0 19.45	4.73 $\pm$ 3.92	4.13

- In the typical urban background area, mainly municipal emissions (burning coal, biomass, waste and rubbish in home furnaces) and energy production (mainly based on hard and brown coal combustion) influence PAH concentrations in the air; in the urban site located near highway, the same sources were active as in the urban background area; additionally, there was a strong influence of the traffic emission. In RB, mainly local and dispersed sources and the inflow of pollutants from other, more polluted regions, affected the air pollution with PAHs in both seasons;
- 5 high percentage of B(a)P in the PAH sum (9%–13%) and very high ambient concentrations of the PM1-bound B(a)P, particularly in the heating season (4–14  $\text{ng}/\text{m}^3$ ), may pose a serious threat to the Silesia inhabitants; the risk does not only concern the residents of large cities and regions located close to important traffic emission sources, it also involves people who dwell in the “clean” areas far away from large urban agglomerations (regional/rural background);
- it may be suspected that in Central, Central-East and East Europe traffic is not the primary PAH source; the PM-bound PAHs come mainly from the fossil fuel combustion for heat and power production.

## LITHUANIA

### Season variations of B(a)P concentrations

[Milukaite ( 2006): Long-term trends of benzo(a)pyrene concentration on the eastern coast of the Baltic Sea, Atmospheric Environment 40 (2006) 2046–2057, doi:10.1016/j.atmosenv.2005.11.045]

The data of benzo(a)pyrene investigation at Preila background station show that daily concentration of benzo(a)pyrene varied by three orders of magnitude from 0.02 to 25.08  $\text{ng}/\text{m}^3$  at this rural site, with high frequency of 0.02–0.5  $\text{ng}/\text{m}^3$  concentration in warm season (May–September) and with high frequency of 0.2–1.0  $\text{ng}/\text{m}^3$  in cold season (October–April). The ratio of benzo(a)pyrene monthly concentration in cold season to warm season reaching 4.2 has indicated that benzo(a)pyrene is attributed to the pollutants which are generated by fuel combustion.

Table 2

Ranges of benzo(a)pyrene monthly concentration ( $\text{ng}/\text{m}^3$ ) and seasonal averages at Preila background station in different periods of investigation

Period	Cold season (October–April)			Warm season (May–September)			
	Min.	Max.	Mean $\pm$ $\sigma$	Min.	Max.	Mean $\pm$ $\sigma$	Ratio, $C_{\text{cold}}/C_{\text{warm}}$
1980–1982	0.72	3.03	1.88 $\pm$ 0.72	0.68	1.72	1.01 $\pm$ 0.28	1.86
1983–1984	0.54	2.62	1.42 $\pm$ 0.74	0.25	0.83	0.46 $\pm$ 0.16	3.09
1985–1989	0.24	3.30	1.00 $\pm$ 0.70	0.02	0.56	0.22 $\pm$ 0.14	4.54
1990–1994	0.23	2.22	0.92 $\pm$ 0.44	0.04	0.79	0.27 $\pm$ 0.22	3.41
1995–1999	0.33	3.24	1.56 $\pm$ 0.80	0.14	1.38	0.35 $\pm$ 0.26	4.46
2000–2002	0.18	1.54	0.97 $\pm$ 0.38	0.12	0.73	0.33 $\pm$ 0.18	2.94

## HUNGARY

### Season variations of B(a)P concentrations

[Csanádi, Zs., A. Szabó Nagy, J. Szabó, J. Erdős (2015): Temporal Variation of PM10-Bound Benzo(a)pyrene Concentration in an Urban and a Rural Site of Northwestern Hungary, International Journal of Environmental, Chemical, Ecological, Geological and Geophysical Engineering Vol:9, No:8, 2015]

The comparison of the B(a)P concentration data for the heating and non-heating periods for both sampling site shows the same tendency. These results are summarized in Table II. The B(a)P concentration measured in Győr exceeded the Hungarian daily limit value of  $1 \text{ ng/m}^3$  in 31 % of the samples collected in periods I. and IV. Concentrations in all samples collected in other seasons were under this limit value. Similar concentration trend was observed in Sarród where B(a)P concentrations in 18 % of the samples were higher than the daily limit value.

TABLE II  
CONCENTRATION RANGES, MEAN VALUES AND STANDARD DEVIATIONS OF  
BAP IN PM10 IN HEATING AND NON-HEATING SEASONS AT THE URBAN SITE  
OF GYŐR AND RURAL SITE OF SARRÓD, HUNGARY ( $\text{ng/m}^3$ )

Sampling site	Year	Heating season	Non-heating season
Győr	2008	0.50–8.00	ND–0.44
		2.18±1.92	0.05±0.08
	2009	ND–3.54	0.02–0.16
		0.99±0.80	0.06±0.04
	2010	0.43–3.63	ND–0.07
		1.32±0.77	0.02±0.02
	2011	0.41–6.92	0.02–0.43
		2.94±2.14	0.10±0.11
	2012	0.37–5.22	0.02–0.33
		2.26±1.37	0.05±0.06
	Sarród	0.02–3.30	ND–0.17
		0.68±0.89	0.04±0.04
	2009	0.18–2.90	ND–0.29
		1.00±0.78	0.09±0.07
	2010	0.19–1.93	ND–0.04
		0.67±0.43	0.01±0.01
	2011	0.13–3.92	ND–0.34
		1.52±1.13	0.07±0.10
	2012	0.05–4.07	ND–0.11
		1.05±1.08	0.03±0.03

ND: not detected

## UK

### Wood burning as one of the most significant source of B(a)P

Report to the Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive, and the Department of the Environment for Northern Ireland  
Assessment of benzo[a]pyrene concentrations in the United Kingdom in the period 2003- 2020 (2006)

In 2002, the largest source was the non-industrial combustion plants (SNAP sector 02) with about 40% of the national total. This consists principally of the estimates of emissions from domestic wood and coal combustion together with emissions from office and commercial heating systems.

**Table 2.3 A comparison of emissions for each of the area source sectors in Great Britain and Northern Ireland (2002).**

SNAP code	Source description	Great Britain		Northern Ireland	
		BAP emission (kg)	Percentage of GB area emission	BAP emission (kg)	Percentage of NI area emission
01	Combustion in industrial	7	0.1%	0	0.0%
02	Non-industrial combustion plants ( <b>Domestic Home heating</b> )	2282	40.0%	611	88.1%
03	Combustion in manufacturing	18	0.3%	1	0.1%
04	Production processes	52	0.9%	0	0.1%
06	Solvent and other product use	38	0.7%	2	0.3%
07	Road transport	492	8.6%	15	2.1%
08	Other mobile sources	26	0.5%	1	0.2%
09	Waste treatment and disposal ( <b>Waste fires</b> )	1921	33.7%	39	5.6%
11	Other sources and sinks ( <b>Natural fires</b> )	863	15.1%	25	3.6%
	Total	5699	100.0%	694	100.0%

## Population exposure to ambient air B(a)P in Europe

**UK: Report to the Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive, and the Department of the Environment for Northern Ireland**  
**Assessment of benzo[a]pyrene concentrations in the United Kingdom in the period 2003- 2020 (2006)**

Nationally the number of people exposed to the air quality objective concentration of  $0.25 \text{ ng/m}^3$  decreased from 2.4 million in 2003 to 1.6 million by 2010. For the target value of  $1.0 \text{ ng/m}^3$  the proportional decrease is even larger- from 17000 exposed in 2003 to 5000 in 2010.

There are no agglomeration zones for which the target value of  $1 \text{ ng/m}^3$  was predicted to be exceeded. For the regional zones in 2003 there are small number of exceedences in Yorkshire and Humberside and Northern Ireland. By 2010, only a small number of people in Yorkshire and Humberside were predicted to be exposed to concentrations greater than the target concentration.

## Population exposure to ambient air B(a)P in Europe

**[Guerreiro C.B.B, Valentin Foltescu, Frank de Leeuw (2014): Air quality status and trends in Europe, Atmospheric Environment 98 (2014), 376-384, <http://dx.doi.org/10.1016/j.atmosenv.2014.09.017>]**

The European directive (EU, 2004) sets a target value (TV) for ambient air concentration of B(a)P in order to avoid, prevent and reduce harmful effects of PAHs on human health and the environment as a whole. Around 90% or more of B(a)P in ambient air is adsorbed onto aerosols and around 10% or less is in the gas phase (Yamasaki *et al.*, 1982; Yagishita *et al.*, 2015; Gao *et al.*, 2015). The TV for B(a)P (measured in PM<sub>10</sub>) was set to  $1 \text{ ng/m}^3$  as an annual mean, to be met by 2013.

Benzo(a)pyrene measurements in 2011 were above the target value threshold ( $1 \text{ ng/m}^3$  annual average to be met by 2013) at 35% of the monitoring stations. This was the case mainly at urban and suburban background stations (50% exceeded the target value) and, to a lesser extent, at rural, traffic and industrial stations. Exceedences are most predominant in central and eastern Europe. Exposure of the European population to B(a)P concentrations above the target value is significant and widespread, especially in

central and eastern Europe. Between 22% and 31% of the urban population in the EU-27 was exposed to B(a)P concentrations above the target value ( $1 \text{ ng/m}^3$ ) in the period 2009 to 2011, and in this time a tendency of increasing exposure can be observed. As much as 94% of the EU-27 urban population was exposed to B(a)P concentrations above the WHO reference level ( $0.12 \text{ ng/m}^3$ ) over the same period. In large parts of Europe the concentrations are expected to be below the lower assessment threshold ( $0.4 \text{ ng/m}^3$ ), and consequently the air quality directive (EU, 2004) does not require monitoring of B(a)P. As the WHO reference level is below the lower assessment threshold the estimate of the population exposure is highly uncertain. Emissions of B(a)P in the EU-27 have increased by 11% between 2002 and 2011, due to the increase in emissions from the 'commercial, institutional and household fuel combustion' sector of 24%. In Europe, B(a)P pollution is an increasing problem, especially in areas where domestic coal and wood burning is common.

[Guerreiro, C.B.B, J. Horalek b, F. de Leeuw c, F. Couvidat., (2016): Benzo(a)pyrene in Europe: Ambient air concentrations, population exposure and health effects, *Environmental Pollution* 214 (2016) 657-667, <http://dx.doi.org/10.1016/j.envpol.2016.04.081>]

About 20% of the European population living in regions with an acceptable uncertainty has been exposed to B(a)P annual mean concentrations above the TV ( $1 \text{ ng/m}^3$ ) in 2012 and only about 7% live in areas with concentrations under the estimated reference level of  $0.12 \text{ ng/m}^3$  (Table 2). Both the percentage of inhabitants living in the areas above the TV and the population-weighted concentration are quite high. The reason for this is the fact that the most of the European population live in urban areas, in general more polluted than the rural areas.

**Table 2**

Population exposure and population-weighted concentration for BaP annual mean in 2012, based on the interpolated concentration map using the EMEP and CHIMERE modelled data in areas with an acceptable relative uncertainty (equal or below 0.60).

Map	Population [inhbs. 1000]	BaP, annual average, exposed population [%]						Population-weighted conc. [ng m <sup>-3</sup> ]
		<TV			>TV			
		<0.12 ng m <sup>-3</sup>	0.12–0.4 ng m <sup>-3</sup>	0.4–0.6 ng m <sup>-3</sup>	0.6–1 ng m <sup>-3</sup>	1–1.5 ng m <sup>-3</sup>	>1.5 ng m <sup>-3</sup>	
Using EMEP	311,873	7.5	53.0	9.5	10.0	5.4	14.7	0.93
		79.9				20.1		
Using CHIMERE	317,679	7.3	52.9	9.8	9.8	5.4	14.7	0.95
		79.9				20.1		

Exposure to these B(a)P concentrations lead to an estimated increase in the number of lung cancer incidence within the modelled area (with an acceptable uncertainty) of 370 (95% CI: 120-630) and 380 (95% CI: 130-660) using the EMEP and CHIMERE model, respectively. The largest health impacts can be found in the central eastern European countries. These results are most probably underestimated, and a sensitivity test indicates the number of incidents may be underestimated by 20% or more. In addition, it is important to bear in mind that B(a)P only represents part of the overall carcinogenicity amongst the group of PAH compounds for which data are available (Holland et al., 2001; Pufetele et al., 2004).

Furthermore, long-term exposure to PAHs has been associated, in addition to lung cancer, to other health endpoints such as increased incidence of skin and bladder cancer; genotoxicity (Topinka et al., 2011); prenatal and early-life exposure to PAHs may affect children's cognitive development and be associated with behaviour problems as ADHD (Edwards et al., 2010; Perera et al., 2014). WHO (2013) has found new evidence linking PAH exposure to cardiovascular morbidity and mortality, although at present the effects of PAH exposure cannot be easily separated from those of particles.

[EEA Report No 5/2015: Air quality in Europe — 2015 report, doi:10.2800/62459]

Exposure to benzo[a]pyrene (B(a)P) pollution is quite significant and widespread, in particular in central and eastern Europe. Approximately half of the B(a)P measurement stations in Europe were in exceedance of the EU target value in 2013, mostly in urban areas. About 20% of the total European population was



exposed to B(a)P annual mean concentrations above the European target value in 2012 and about 88% lives in areas with concentrations above the estimated reference level (3).

Considering only urban populations, in 2013 25% of the EU-28 urban population was exposed to B(a)P concentrations above the target value, and as much as 91% was exposed to B(a)P concentrations above the estimated reference level (Table ES.1).

**Table ES.1** Percentage of the urban population in the EU-28 exposed to air pollutant concentrations above certain EU and WHO reference concentrations (2011–2013)

Pollutant	EU reference value	Exposure estimate	WHO AQG	Exposure estimate
PM <sub>2.5</sub>	Year (25)	9–14	Year (10)	87–93
PM <sub>10</sub>	Day (50)	17–30	Year (20)	61–83
O <sub>3</sub>	8-hour (120)	14–15	8-hour (100)	97–98
NO <sub>2</sub>	Year (40)	8–12	Year (40)	8–12
BaP	Year (1 ng/m <sup>3</sup> )	25–28	Year (RL, 0.12 ng/m <sup>3</sup> )	85–91
SO <sub>2</sub>	Day (125)	<1	Day (120)	36–37

Key:	< 5%	5–50%	50–75%	> 75%
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**Notes:** The estimated range in exposures refers to a recent three year period (2011–2013, except for SO<sub>2</sub> WHO AQG, 2011–2012) and includes variations due to meteorology, as dispersion and atmospheric conditions differ from year to year.

The reference concentrations include EU limit or target levels, WHO air quality guidelines (AQG) and estimated reference levels.

The reference concentrations in brackets are in µg/m<sup>3</sup> except for BaP in ng/m<sup>3</sup>.

For some pollutants EU legislation allows a limited number of exceedances. This aspect is considered in the compilation of exposure in relation to EU air quality limit and target values. The comparison is made for the most stringent EU limit or target values set for the protection of human health. For PM<sub>10</sub> the most stringent limit value is for 24-hour mean concentration and for NO<sub>2</sub> it is the annual mean limit value.

As the WHO has not set AQG for BaP, the reference level in the table was estimated assuming WHO unit risk for lung cancer for PAH mixtures, and an acceptable risk of additional lifetime cancer risk of approximately 1 x 10<sup>-6</sup> (ETC/ACM, 2011).

**Source:** Based on EEA, 2015d.

## Exceedances of the target value

Ambient air concentrations of B(a)P are high across large parts of Europe, mostly as a result of emissions from the domestic combustion of coal and wood. About half of the B(a)P measurement stations in Europe continue measuring concentrations above the target value threshold (1 ng/m<sup>3</sup> annual average, to be met by 2013) in 2013. Exceedances were measured mainly at urban and suburban stations, with 97% of all stations in exceedance located in urban and suburban locations, and 87% of all exceedances measured at suburban and urban background stations. As in previous years, exceedances are most predominant in central and eastern Europe (Austria, Bulgaria, Croatia, the Czech Republic, Hungary, Italy, Lithuania, Poland and Slovenia), although there are also exceedances in France, Germany, Spain and the United Kingdom. Figure 6.1 shows the annual mean B(a)P values for 2013 for all EU Member States. It shows that average annual concentrations of B(a)P exceeded the target value in the 13 countries mentioned above. The average concentration measured at Polish stations is 4.6 times higher than the target value. Only 24 of the EU-28 Member States reported B(a)P data with sufficient data coverage (13) for 2013. Reported monitored data are missing from a large part of south-eastern Europe. For example, no measurements are reported from Romania, a country with high estimated B(a)P concentrations.

**Table 10.1** Population exposure and population-weighted concentration for BaP annual mean in 2012, based on the interpolated concentration map

BaP: annual mean, exposed population (%)						BaP population-weighted concentration
< 0.12 ng/m <sup>3</sup>	0.12–0.4 ng/m <sup>3</sup>	0.4–0.6 ng/m <sup>3</sup>	0.6–1.0 ng/m <sup>3</sup>	1.0–1.5 ng/m <sup>3</sup>	> 1.5 ng/m <sup>3</sup>	
11.7%	46.7%	10.4%	10.7%	6.8%	13.6%	0.84 ng/m <sup>3</sup>

**Source:** ETC/ACM, 2015b.