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Levels, trends and health concerns of atmospheric PAHs in Europe

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HIGHLIGHTS

- Atmospheric PAHs generally have higher levels in the Central/Baltic areas.
- Marked seasonality of PAH concentration, with winter highs and summer lows.
- Downward temporal trends are perceived as we approach the higher latitudes.
- Air quality guidelines for BaP have been surpassed in several areas in the last years.

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ABSTRACT

Changes in climate can affect the concentration patterns of polycyclic aromatic hydrocarbons (PAHs) by altering the dispersion (wind speed, mixing layer height, convective fronts), deposition by precipitation, dry deposition, photochemistry, natural emissions and background concentrations. This means the evolution trends of these pollutants have to be studied under a multi-scale perspective, allowing the establishment of transport patterns and distribution of PAHs. In this sense, this work tries to unveil the atmospheric behaviour of these pollutants using temporal data series collected in different stations from the European Monitoring and Evaluation Programme (EMEP) air sampling network. These sites are thought to avoid the direct influence of emitting areas (background stations), allowing the study of long-range transport effects, intra- and trans-annual variability, relationships between concentrations patterns and meteorological variables and latitudinal gradients of PAH levels in Europe. Overall, a typical high concentration pattern was found for the colder months (and an opposite behaviour is found for summertime). Negative trends were detected over high latitudes, for instance, in Svalbard (Norway), whereas for the United Kingdom the pattern is the inverse. Also, negative latitudinal gradients were observed in 4 of the 15 PAHs studied. Finally, air quality parameters revealed concern over human health issues, given the recent increase of BaP levels in Europe.

1. Introduction

As a result of the industrial development occurred in Europe during the last century, environmental pollution levels have been increasing exponentially. For example, the use of fossil fuels such as coal and petroleum generated increases in the emission of different pollutants into the atmosphere, such as nitrogen oxides, sulphur dioxide, carbon monoxide or organic compounds of different volatilities. The latter group includes the polycyclic aromatic hydrocarbons (PAHs), produced by natural and anthropogenic sources (Srogi, 2007), that can be found not only in the atmosphere associated with the gas or the particulate phase (Kiss et al., 2001; Lammel et al., 2009), but also in other environmental matrices like soil, sediment or vegetation (Wild and Jones, 1995; Chen et al., 2004; Navarro-Ortega et al., 2012). These compounds are widely studied due to their carcinogenic and mutagenic properties. Their high lipid solubility makes them prone to absorption in the lung tissue, skin, breasts or intestines, carrying risks to the human health (Maron and Ames, 1982; Howard et al., 1995; Kim et al., 2013).

However, existing literature on PAHs report that their atmospheric content is reduced comparing to the other environmental matrices. Maliszewska-Kordybach (1999) mentioned an overall presence of only 0.5%, while Wild and Jones (1995) found that the airborne PAHs in the British territory represent only 4% of total anthropogenic emissions in a year. This is evidence that the atmosphere is not a repository for these compounds but rather a pathway for their transport, dilution and transformation (Wild and Jones, 1995).

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An important aspect in that in most studies, the areas covered are small (form cities to 50 km wide regions), tendency profiles are often missing and the temporal trends use datasets of less that 10 years (Dimashki et al., 2001; Prevedouros et al., 2004). For instance, Hwang and Wade (2008) and Amador-Muñoz et al. (2013) had time series of 4 and 2 years to obtain the PAHs levels in specific locations of the USA and Mexico, respectively. Hwang and Wade (2008) used pine needles as biomonitors of the airborne PAHs. Also, no detailed analysis was found integrating the entire European continent, essential to assess trans-boundary pollution, except for the information yielded by the EMEP network, a programme which derives from the Geneva Convention of 1977 on transboundary pollution (http://www.emep.int). However, even the EMEP network in its 2012 report on particulate matter applying temporal series, relies on time-frames of less than 10 years and only distinguishes between PM2.5 and PM10, despite having information on individual contaminants (Torseth et al., 2012).

Fuelled by the relatively small amount of related literature (considering the existing databases on atmospheric PAHs), four main objectives are intended for the current study: to conduct a comparative analysis of the concentration of 15 PAHs in the atmosphere throughout the European continent, characterising the temporal variability of the series, including intra-annual patterns, using the Mann–Kendall test to determine statistical significance; to establish correlations with meteorological parameters, in order to explain the influence of climate on the variability of PAH levels, with significance calculated by the Pearson test; to determine possible latitudinal gradients of concentration for each PAH under study; and finally to evaluate if the PAH incidence in the time series considered poses problems concerning human health quality guidelines. With these objectives it is also an aim to estimate the influence of atmospheric transport from the source to remote areas in Europe.

2. Methodology

2.1. Target pollutants

The study will focus on the dataset available from the EMEP network concerning 15 of the 16 EPA (Environmental Protection Agency) PAHs: acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fluo), phenanthrene (Phen), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA) and benzo[ghi]perylene (BghiP). Naphthalene was not included since there were no data available to constitute valid time series. EMEP air samples were collected by high-volume sampling devices.

2.2. Sampling stations

The EMEP network consists of 315 stations spread throughout Europe from Portugal to the Norwegian territory of Svalbard, but only 47 measured persistent organic pollutants (POPs) and other related contaminants, not covering all countries (for instance France and Italy are not included, see http://www.emep.int). Moreover, only 15 of the 47 stations measure the air + aerosol levels of PAHs (see Fig. 1 and Table 1 for details), and even so with variable data ranges, as indicated in Table 1. In any case, only series with at least one year of monthly mean results for each individual PAH were considered in this study. Table 2 indicates the number of valid measurements for each PAH in the sampling stations used.

2.3. Meteorological data

The meteorological data used in this study were mainly obtained from the European Climate Assessment & Dataset project (ECA&D), a database which is part of the agency for international climate assessment (http://eca.knmi.nl). The exception was the Kosetice station (Czech Republic), where the Weather Underground website (http://www.wunderground.com) was used. The parameters available and considered for the correlation analysis were: mean and maximum temperature, precipitation, cloud cover, solar radiation (hours of sunlight) and relative humidity. Nevertheless, not all of them were measured in all sampling sites considered in this work.

2.4. Data processing

The first task of this study was to extract, review and compile data available from the EMEP network on the air concentrations of PAHs, building the respective time series for each location. The data collected are averages of each pollutant's monthly concentrations, expressed in ng m^{-3}. Subsequently, these results were treated using the methodologies detailed below.

2.4.1. Basic treatment of the data

A basic statistical processing has been applied to the available air concentrations of each PAH in each sampling station, which comprised the following procedure: average the monthly concentration values - building the “type year”; average the seasonal concentrations (winter, spring, summer and autumn) and calculate the standard deviation between seasons; average the total concentration of each series, with the respective standard deviations.

2.4.2. Trend analysis: Mann–Kendall test

In a second step, an analysis of trends over time was performed, normalising the results by subtracting the “type year” and applying the statistical test of Mann–Kendall, one of the most widely used to verify the significance of the tendencies in temporal data series (Hipel and McLeod, 1994; Hamed, 2008). This test combines the development of the nonparametric trend test first proposed by Mann (1945) and further developments by Kendall (1955). In this work, the first approach, commonly known as “pre-whitening” of the data (Von Storch, 1995), was applied, namely by subtracting the aforementioned “year type” to each year of the time series before running the test on the uncorrelated residuals. In the current study, the test was used for a level of confidence of 95%.

2.4.3. Correlation with meteorological variables and latitude

The process to obtain the meteorological data involved finding the closest ECA&D stations to the air sampling site, and relying in factors such as proximity (giving more importance to the latitudinal component), the existence of records in the years in which data on PAH concentration was available and finally those in which additional atmospheric variables are recorded such as humidity, solar radiation, cloud cover, and discarding the stations which had not at least of the two parameters that were considered the most important: temperature and precipitation.

Then, the PAHs levels from the stations where a significant temporal trend was observed were correlated with meteorological series in order to determine whether those patterns can be attributed to climatic factors. The significance of such correlations was assessed using the Pearson test (with a significance of at least $p < 0.05$) to determine the consistency of the correlation.

Finally, the possibility of latitudinal trends was also evaluated, correlating the mean total concentration of each PAH with the

Fig. 1. Map of EMEP sampling stations measuring air + aerosol levels of PAHs.

respective coordinates of each station. Again the Pearson test was employed to detect those with significance (at least $p < 0.05$).

3. Results and discussion

As indicated in Table 1, the longer time data series are located in Norway (Svalbard Island), Sweden (Aspvreten), Finland (Pallas) and Czech Republic (Kosetice). Naturally, these are the series where the results supposedly have the highest statistical weight, although it is possible that they contain anomalous years or an intensified effect of statistical noise. In a similar way, there are also individual PAHs with more data available than others (with BaP on top of the list). Taking these facts into consideration, the results of the application of the methods described in section 2 to all the available datasets are presented below.

3.1. Concentration patterns of PAHs

In terms of the mean total PAHs concentrations, Fig. 2 presents the results for each of the 15 EMEP stations studied. The highest mean concentration was found in Kosetice (Czech Republic), with 0.994 ± 1.543 ng m^{-3}, followed by Preila (Lithuania), with 0.701 ng m^{-3}. In the latter case, only BaP was measured (see Table 2), implying that the levels could have been much higher if more PAHs were accounted for. In plain view, it is appreciable that the PAH levels tend to decrease towards the northernmost stations of the Scandinavian Peninsula. In these areas, the urban pressure and, consequently, the frequency of emitting sources is reduced compared to the United Kingdom or Central Europe, suggesting the lower influence of nearby sources for the background level measurements comparing to long-range transport phenomena. A similar trend was found by Jaward et al. (2004), in a passive air sampling campaign throughout Europe. In the same study, the authors reported analogous concentrations for several PAHs sampled with passive and active (Hi-Vol) devices in four UK sites. Pribylova et al. (2012) also observed high PAH concentration in the Central and Eastern Europe areas.

Considering the 15 PAHs individually and in the whole sampling grid, it can be seen in Fig. 3 that phenanthrene is overall the most abundant compound, with a mean total of 2.243 ± 1.772 ng m^{-3}, followed by Fluo, Flt and Pyr. The predominance of these “lighter” PAHs is frequently reported in literature, especially emphasising the dominant percentage of Phen in the PAH load (Jaward et al., 2004; Tsapakis and Stephanou, 2005; Van Drooge et al., 2010; Halse et al., 2011). Dimashki et al. (2001) found a similar pattern in the UK, but with mean concentrations about one order of magnitude higher, reflecting an urban-based sampling strategy. The levels of the heavier particulate-bound PAHs are lower and

Table 1
Characteristics of the EMEP stations measuring PAHs in air + aerosol.

EMEP code	Name	Country	Latitude	Longitude	Elevation (m)	Data range
NO0042G	Svalbard	Norway	78°54′ N	11°53′ E	474	1994–2010
FI0036R	Pallas	Finland	68°00′ N	24°14′ E	340	1996–2010
EE0009R	Lahemaa	Estonia	59°30′ N	25°54′ E	32	2007–2009
SE0012R	Aspvreten	Sweden	58°48′ N	17°23′ E	20	1995–2010
NO0002R	Birkenes II	Norway	58°23′ N	8°15′ E	219	2009–2011
SE0002R	Rörvik	Sweden	57°25′ N	11°56′ E	10	1995–2010
SE0014R	Råö	Sweden	57°23′ N	11°54′ E	5	2002–2008
LT0015R	Preila	Latvia	55°21′ N	21°04′ E	5	2000–2002
DE0001R	Westerland	Germany	54°55′ N	8°18′ E	12	2007–2010
DE0009R	Zingst	Germany	54°26′ N	12°44′ E	1	2007–2009
GB0014R	High Muffles	UK	54°20′ N	0°48′ W	267	2004–2008
BE0013R	Houtem	Belgium	51°15′ N	3°21′ E	10	2009–2010
DE0008R	Schmücke	Germany	50°39′ N	10°46′ E	937	2007–2010
CZ0003R	Kosetice	Czech Rep.	49°35′ N	15°05′ E	534	1999–2010
DE0003R	Schauinsland	Germany	47°54′ N	7°54′ E	1205	2007–2010

NOTE: Pallas and Preila stations in 2009 and Svalbard and Aspvreten stations in 2008 did not collect data.

Table 2
Number of valid data for each PAH in EMEP stations measuring air + aerosol.

EMEP code	Acy	Ace	Fluo	Phen	Ant	Flt	Pyr	BaA	Chry	BbF	BkF	BaP	IcdP	DahA	BghiP
NO0042G				169	168	169	169	168	12	24	24	167	169		169
FI0036R												36			
EE0009R				48	48	48	48	48				48	48	48	48
SE0012R												165	165		167
NO0002R	29	30	30	30	30	30	30	30				30	30		30
SE0002R				167	167	167	167	164		167	167	36			
SE0014R	203	179	203	203	201	191	191	203				188	203		198
LT0015R				92	92	92	92	92		82	82	92	92		
DE0001R				48	48	48	48	48				48	48	48	48
DE0009R				36	36	36	36	36				36	36	36	36
GB0014R				96	96	96	96	96	12	96	96	96	96		96
BE0013R				48	48	48	48	48				48	48	48	48
DE0008R				48	60	48	48	60	60		12	60	57	12	60
CZ0003R	12	72	144	144	132	144	144	132		60	60	144	132	12	36
DE0003R						24	24	24				24			24

similar among them. Still, BaP presents the higher mean concentration of this lot, with 0.144 ± 0.174 ng m^{-3}, reinforcing the need for a closer incidence on the study of this harmful contaminant, which will be presented later.

For each station, the same behaviour is observed (Fig. 4), with the light molecular weight PAHs (3 and 4 rings) dominating whenever they were analysed. Particularly impressive is the case of High Muffles (UK), where the 3-ring PAHs have a presence of over 85%, again almost entirely due to the presence of Phen. In Preila (Lithuania), only BaP was measured, and the levels are considerably higher that those obtained for 5-ring PAHs in the other stations. This may be a reason for concern and a thorough analysis of the sources accounting for these levels should be conducted in future studies.

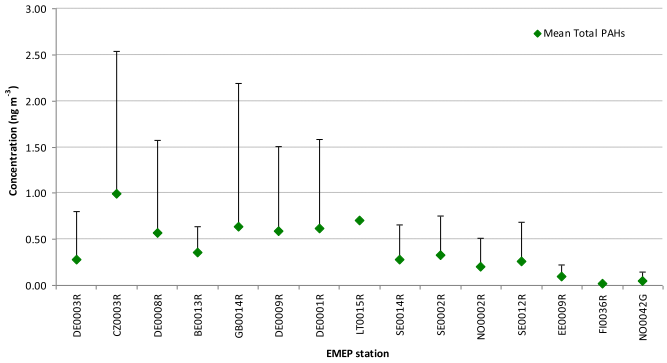

Fig. 2. Mean total concentration of PAHs in the 15 EMEP stations considered (listed in increasing latitude from left to right). Lines indicate the standard deviation (only plus side is presented).

3.1.1. Seasonal variations

The mean seasonal patterns of PAHs incidence show a remarkable consistency in terms of number of aromatic rings. As depicted in Fig. 5, the highest mean concentrations are always found in winter, followed by autumn, spring and finally summer. This is directly related to the number of emitting sources, which in turn are influenced by some meteorological conditions (Tham et al., 2008). Being a product of the combustion of fossil fuels, the reduction of the air temperature leads to an increased use of building heating systems and traffic, which are arguably two of the strongest contributors to the PAH burden in the environment (Hwang and Wade, 2008). Other authors found similar tendencies worldwide, not only in air but also in other media such as vegetation (Prevedouros et al., 2004; Ratola et al., 2010; Bernalte et al., 2012).

Considering each of the 15 sampling stations in study (Fig. 6), there is generally a similar trend, but in this case not as constant as for the whole dataset combined. For instance, in High Muffles (GB0014R), the distribution of the PAH concentrations is relatively identical among the four seasons, especially for the lighter compounds (3 and 4 rings). This may be a reflection of a not so pronounced difference in temperature and intra-annual rainfall, which motivates strong constancy of the emitting sources of these

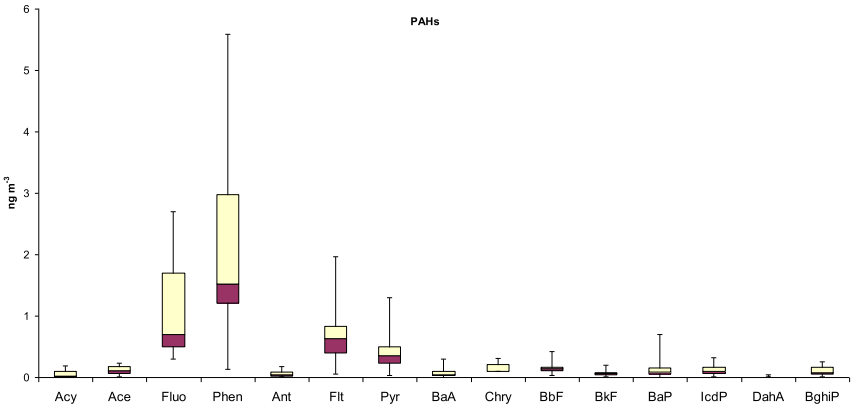

Fig. 3. Box-and-whisker plot for the concentration of each PAH for the whole dataset available.

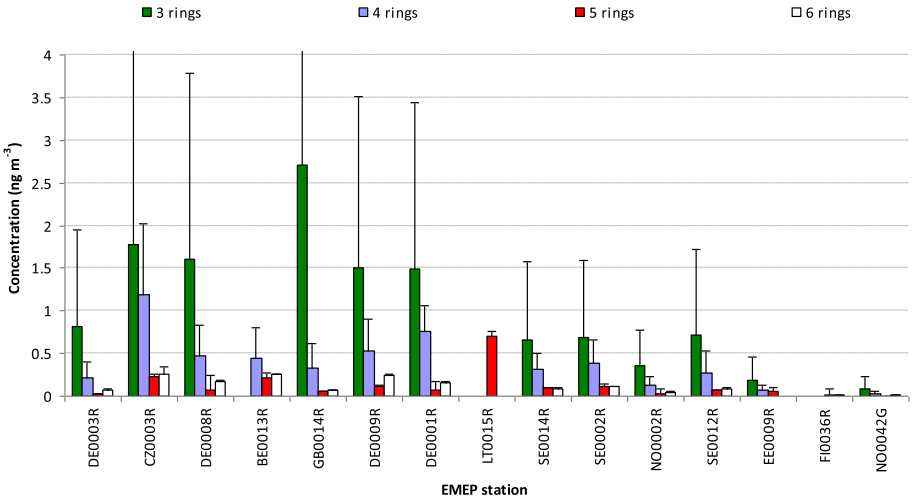

Fig. 4. Mean concentration of PAHs according to their aromatic rings for each sampling station and for the whole dataset available. Lines indicate the standard deviation (only plus side is presented).

pollutants. Overall, the least volatile PAHs show greater variations, with a strong predominance in the winter months comparing to the other seasons. Apart from the aforementioned increase of the sources in winter, gas-particle partition can also be affected by atmospheric transport phenomena (Baek et al., 1991), hence changing the seasonal dynamics of PAHs. Another phenomenon to take into account is the higher atmospheric presence of OH-radicals in the summer, enhancing the possibility of reactions with the PAHs

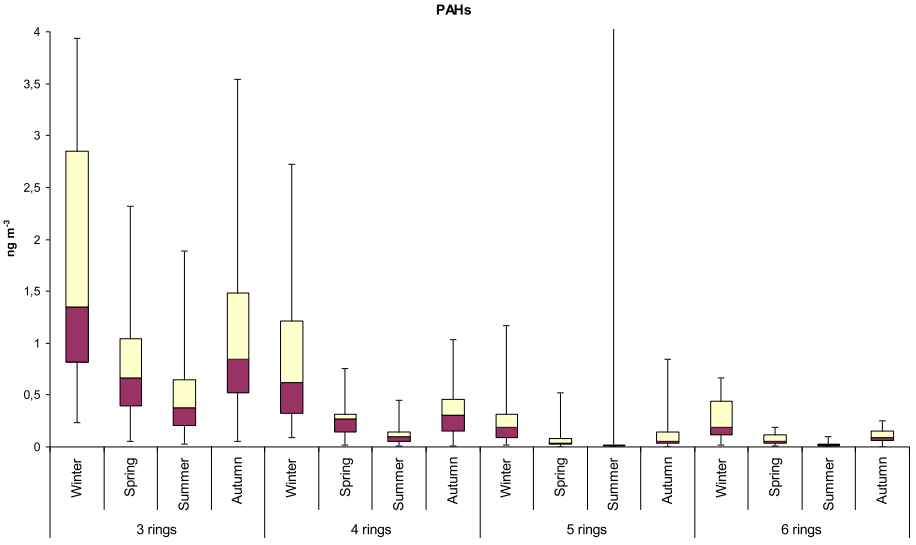

Fig. 5. Box-and-whisker plot for the mean seasonal variation of PAHs according to the number of aromatic rings for the whole dataset available.

(hence reducing their levels) to produce nitro-PAHs (Reisen and Arey, 2005). The only exception to these evidences was found in the Estonian station of Lahemaa, where the 5-ring PAHs have an almost 90% prevalence in the summer. There is no apparent reason for this behaviour, but it may have been caused by a local episode of severe contamination or a problem with the measurements at that particular time.

3.2. Trend analysis

This section will discuss the results of applying the Mann–Kendall test on the normalized dataset at each station for each compound tested in search of significant temporal trends. In the cases where this is verified (with a confidence level of 95%, i.e., $p < 0.05$), also the correlations with the meteorological variables were studied, to assess possible influences on the temporal patterns of each PAH. Finally, relationships between the presence of PAH and latitude were studied.

3.2.1. Temporal trends

In 35 (27.5%) of the 127 temporal series of individual PAHs studied, it was possible to establish statistically significant correlations, as can be seen in Table 3. Here, the slope represents the linear correlation between air concentrations of the PAHs and time (ng m^{-3} $month^{-1}$) and the cells in bold indicate the significant correlations found above 95% ($p > 0.05$).

It is expected that the stations with longer datasets can more easily produce these correlations. This is the case of Aspvreten (Sweden) or Svalbard, where 5 out of 10 and 8 out of 11 PAHs showed significant temporal patterns. However, this was not the case of, for instance, Birkenes (south Norway) or Kosetice (Czech Republic), where long temporal series could only produce valid correlations for BghiP and Phen, respectively (Table 3). This can be due to the fact that the inputs of nearby sources in the latter stations were enough to dilute the general decreasing concentration trends verified for the majority of PAHs in this domain of study. In the case of Svalbard, the low presence of emitting sources suggests that the PAH levels found are predominantly due to long range atmospheric transport from lower latitudes, reflecting the aforementioned decreasing trend of the PAH concentrations. In fact, all

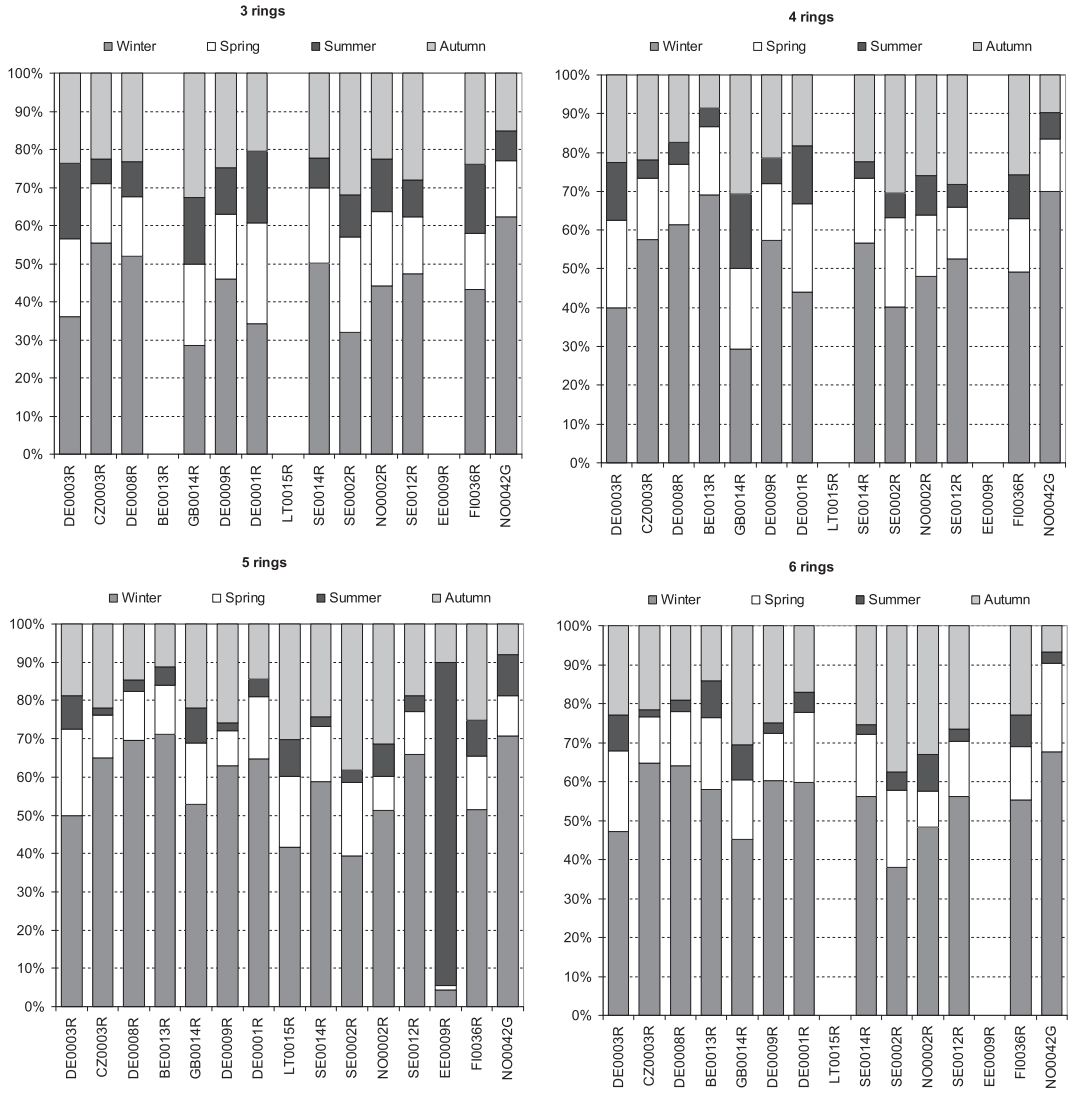

Fig. 6. Mean seasonal patterns of PAHs for each sampling station according to the number of aromatic rings for the whole dataset available.

Table 3
Results of the Mann–Kendall test for the temporal trends of the target PAH concentrations.

		Schauinsland	Kosetice	Schmücke	Houtem	High Muffles	Zingst	Westerland	Preila	Råö	Rörvik	Birkenes II	Aspvreten	Lahemaa	Pallas	Svalbard
Acy	Slope											0				**-2.3×10^{-5}**
	$p < 0.05$											No				**Yes**
Ace	Slope		2.8×10^{-4}									5.2×10^{-4}				-3.0×10^{-6}
	$p < 0.05$		No									No				No
Fluo	Slope		-8.4×10^{-4}									6.0×10^{-3}				**-4.8×10^{-3}**
	$p < 0.05$		No									No				**Yes**
Phen	Slope	-9.3×10^{-5}	**-8.0×10^{-3}**	**-2.2×10^{-2}**		-3.2×10^{-2}	-2.2×10^{-2}	-7.0×10^{-3}		1.0×10^{-3}	-5.2×10^{-4}	2.0×10^{-3}	**-3.0×10^{-3}**		2.3×10^{-4}	**-4.6×10^{-4}**
	$p < 0.05$	No	**Yes**	**Yes**		No	No	No		No	No	No	**Yes**		No	**Yes**
Ant	Slope	-8.3×10^{-5}	-1.8×10^{-4}	-7.0×10^{-6}		-8.2×10^{-4}	0	0		4.9×10^{-6}	-6.9×10^{-5}	0	**-5.7×10^{-5}**			**-2.5×10^{-5}**
	$p < 0.05$	No	No	No		No	No	No		No	No	No	**Yes**			**Yes**
Flt	Slope	1.0×10^{-3}	-1.0×10^{-3}	3.0×10^{-3}	**-1.2×10^{-2}**	0	0	0		-1.9×10^{-6}	-2.0×10^{-3}	1.0×10^{-3}	**-1.0×10^{-3}**		-1.5×10^{-4}	**-1.2×10^{-4}**
	$p < 0.05$	No	No	No	**Yes**	No	No	No		No	No	No	**Yes**		No	**Yes**
Pyr	Slope	**1.0×10^{-3}**	-3.6×10^{-4}	2.0×10^{-3}	-4.0×10^{-3}		0	0		-4.1×10^{-5}	-4.2×10^{-4}	8.1×10^{-4}	**-5.1×10^{-4}**		-5.8×10^{-5}	**-6.2×10^{-5}**
	$p < 0.05$	**Yes**	No	No	No		No	No		No	No	No	**Yes**		No	**Yes**
BaA	Slope	-1.3×10^{-4}	-1.0×10^{-4}	-5.6×10^{-5}	**-4.0×10^{-3}**	**8.8×10^{-4}**	0	2.5×10^{-4}		-8.3×10^{-5}	2.6×10^{-4}	-2.1×10^{-4}	-4.5×10^{-4}		-5.0×10^{-5}	-2.0×10^{-6}
	$p < 0.05$	No	No	No	**Yes**	**Yes**	No	No		No	No	No	No		No	No
Chry	Slope					**2.0×10^{-3}**										
	$p < 0.05$					**Yes**										
BbF	Slope		6.5×10^{-4}							6.2×10^{-5}	**-6.9×10^{-4}**		-4.6×10^{-4}		-9.1×10^{-5}	
	$p < 0.05$		No							No	**Yes**		No		No	
BkF	Slope		-1.7×10^{-4}							-2.5×10^{-5}	**-2.8×10^{-4}**		-1.6×10^{-4}		**-2.5×10^{-5}**	
	$p < 0.05$		No							No	**Yes**		No		**Yes**	
BaP	Slope	-9.7×10^{-6}	-1.3×10^{-4}	-1.5×10^{-5}	-2.0×10^{-3}	**1.0×10^{-3}**	0	0	-4.0×10^{-3}	-1.1×10^{-4}	8.7×10^{-5}	-7.7×10^{-5}	**-9.7×10^{-5}**	-7.2×10^{-4}	-1.3×10^{-5}	**-1.5×10^{-5}**
	$p < 0.05$	No	No	No	No	**Yes**	No	No	No	No	No	No	**Yes**	No	No	**Yes**
IcdP	Slope	-4.6×10^{-4}	-4.6×10^{-4}	-7.6×10^{-4}		**2.0×10^{-3}**	0	0		-3.4×10^{-6}	-2.0×10^{-4}	1.9×10^{-4}	-7.4×10^{-5}		**-3.9×10^{-5}**	**-7.6×10^{-6}**
	$p < 0.05$	No	No	No		**Yes**	No	No		No	No	No	No		**Yes**	**Yes**
DahA	Slope	-4.2×10^{-4}		**-1.7×10^{-4}**			0	0								
	$p < 0.05$	No		**Yes**			No	No								
BghiP	Slope	**-4.7×10^{-4}**	8.444	**-1.0×10^{-3}**	**-6.0×10^{-3}**	**1.0×10^{-3}**	0	0		**-2.4×10^{-4}**		**-6.2×10^{-4}**	-1.8×10^{-4}		-2.2×10^{-5}	-2.8×10^{-6}
	$p < 0.05$	**Yes**	No	**Yes**	**Yes**	**Yes**	No	No		**Yes**		**Yes**	No		No	No

Slope – resulting from the linear correlation between air concentrations normalised by the "type year" and time (ng m^{-3} $month^{-1}$); $p > 0.05$ – level of significance of 95% or above; no – no significant correlation found; Yes and boldface cells – a significant correlation (>95%) was found.

gas-phase PAHs (3 and 4 rings) except Ace observe this tendency. The European Environment Agency (EEA) reports that the PAH emissions from 1990 to 2010 decreased in the majority of countries, but in some others like Iceland, Denmark, Italy or the Baltic countries, the opposite was found (EEA, 2014). Even considering that EEA reports values for countries and not specific sites, and mainly of total PAHs instead of individual compounds, a good correspondence of air concentration and emission trends could be found, for equivalent time frames. For instance, for BaP, emissions for Sweden decreased 20% and air levels 56%. Similar trends were found for Germany and Westerland (−48% and −68%, respectively) and Czech Republic and Kosetice (44% and 7%, respectively). So it is not surprising that in some cases trends of increasing concentrations can be found. Interestingly, in the current study this was found for all the significant correlations observed in High Muffles (UK). The EEA indicates that the UK had one of the highest decreases in total PAH concentrations from 1990 to 2010, but nevertheless, from 2004 to 2008 (the available data time frame) the opposite was seen for BaA, Chry, BaP, IcdP and BghiP. In fact, for BaP, the emissions in the UK decreased by 25% between 2004 and 2008, but the air concentrations in High Muffles increased by 106%. These compounds, all predominantly associated to particulate matter in the atmosphere, are less prone to long range transport, which suggests that nearby sources may be responsible for these exceptions, associated to an increased urban or industrial pressure in the surrounding areas.

3.2.2. Correlations with meteorological parameters

Table 4 presents the correlations of the meteorological parameters available for each sampling site. Again, the longer time series produce the most significant correlations, but there are not too many clear trends within the domain of study. For instance, for temperature, most gas-phase PAHs with valid relationships present negative correlations, in line with the typical decrease of emissions in warmer seasons. But the same behaviour is not seen for particulate-bound PAHs such as BghiP in Schauinsland (Germany) or BaP and IcdP in Svalbard. Other authors tend to report a general inverse relationship with both gas-phase and particulate PAHs (Motelay-Massey et al., 2003; Barrado et al., 2011), but also indicate the possibility of some variability in these tendencies due to local specificities of emitting sources and climate, including the influence of the boundary-layer height (Prevedouros et al., 2004; Ravindra et al., 2008). On the other hand, it is accepted that precipitation tends to deposit especially the particulate matter from the atmosphere (Amodio et al., 2014). In fact, even future climate change scenarios suggest the rise in concentrations of the particulate material associated to the projected decrease in rainfall (Jiménez-Guerrero et al., 2012). This could be seen in some cases (DahA in Schmücke and BbF in Rörvik), but not for others (Svalbard or Birkenes II). In the latter cases, snowfall is abundant, so precipitation in form of rain may not be as abundant as in areas further south. Or, since rainfall is usually associated with the colder

Table 4
Significant correlations (r, $p < 0.05$) of PAH concentrations and meteorological parameters for the series with temporal correlations (in bold the cases where PAH concentrations also have a significant temporal trend).

		Acy	Ace	Fluo	Phen	Ant	Flt	Pyr	BaA	Chry	BbF	BkF	BaP	IcdP	DahA	BghiP
Svalbard	Avg temperature	**−0.142**	0	**−0.221**	**x**	**x**	**x**	**x**	0				**0.216**	**0.262**		0
	Cloud cover	**x**	0	**0.428**	**0.595**	**x**	**0.752**	**x**	0				**0.798**	**0.331**		0
	Precipitation	**x**	0	**x**	**x**	**0.213**	**0.228**	**x**	0				**0.796**	**0.287**		0
	Rel humidity	**x**	0	**x**	**0.264**	**0.209**	**0.989**	**0.263**	0				**0.913**	**0.334**		0
	Max temperature	**x**	0	**x**	**0.425**	**x**	**−0.714**	**0.425**	0				**0.673**	**0.288**		0
	SR	**x**	0	**x**	**x**	**x**	**x**	**x**	0				**x**	**x**		0
Pallas	AT, MT, PP				0		0	0	0		0	**x**	0	**x**		0
Lahemaa	AT, MT, PP												0			
Aspvreten	Avg temperature				**−0.162**	**x**	**−0.189**	**−0.141**	0		0	0	**x**	0		0
	Max temperature				**−0.200**	**x**	**−0.235**	**−0.183**	0		0	0	**−0.211**	0		0
	Cloud cover				**0.204**	**x**	**0.204**	**x**	0		0	0	**x**	0		0
	PP				**x**	**x**	**x**	**x**	0		0	0	**x**	0		0
Birkenes II	Precipitation	0	0	0	0	0	0	0	0				0	0		**0.431**
	Solar radiation	0	0	0	0	0	0	0	0				0	0		**0.401**
	AT, MT, CC	0	0	0	0	0	0	0	0				0	0		**x**
Rörvik	Avg temperature				0	0	0	0	0		**−0.256**	**−0.229**	0	0		
	Precipitation				0	0	0	0	0		**−0.287**	**x**	0	0		
	MT, SR				0	0	0	0	0		**x**	**x**	0	0		
Råö	Precipitation				0	0	0	0	0		0	0	0	0		**−0.252**
	Cloud cover				0	0	0	0	0		0	0	0	0		**0.348**
	AT, MT				0	0	0	0	0		0	0	0	0		**x**
Preila	AT, MT, PP, RH												0			
Westerland	AT, MT, PP				0	0	0	0	0				0	0	0	0
Zingst	AT, MT, PP, SR, CC, RH				0	0	0	0	0				0	0	0	0
High Muffles	AT, MT, PP				0	0	0		**x**	**x**			**x**	**x**		**x**
Houtem	AT, MT, PP, SR, CC, RH						**x**	0	**x**				0			**x**
Schmücke	Avg temperature				**−0.586**	0	0	0	0				0	0	**−0.309**	**x**
	Max temperature				**−0.572**	0	0	0	0				0	0	**x**	**x**
	Rel humidity				**0.269**	0	0	0	0				0	0	**x**	**x**
	Precipitation				**x**	0	0	0	0				0	0	**−0.369**	**x**
	SR, CC				**x**	0	0	0	0				0	0	**x**	**x**
Kosetice	Avg temperature		0	0	**−0.281**	0	0	0	0		0	0	0	0		0
	Max temperature		0	0	**−0.238**	0	0	0	0		0	0	0	0		0
Schauinsland	Avg temperature				0	0	0	**x**	0				0	0	0	**0.936**
	Max temperature				0	0	0	**x**	0				0	0	0	**0.582**
	Precipitation				0	0	0	**x**	0				0	0	0	**0.804**
	Solar radiation				0	0	0	**x**	0				0	0	0	**0.794**
	Cloud cover				0	0	0	**x**	0				0	0	0	**0.957**
	Rel humidity				0	0	0	**0.697**	0				0	0	0	**0.936**

Blank spaces – no data available; 0 – no significant temporal correlation with PAH concentration; x – correlation not significant; AT – average temperature; MT – maximum temperature; PP – precipitation; SR – solar radiation; CC – cloud cover; RH – relative humidity.

months, the influence of the increase in emitting sources derived from the temperature decrease dilutes the influence of wet scavenging from the atmosphere. In any case, the correlations found were not conclusive as to the impact of climatic factors in the temporal trends of PAH levels, also due the lack of information to build a complete database for all PAHs and sampling stations.

3.2.3. Correlations with latitude

The mean total concentration of PAHs present a negative correlation with the latitude of the sampling stations, with a clear statistical significance ($p < 0.01$). However, individually, although the same inverse trend applies in almost all cases (IcdP and DahA are the exceptions), it was only possible to obtain significance for four PAHs (Phen, Flt, Pyr and BghiP, with $r = -0.642$, -0.623, -0.582 and -0.617, respectively). In some cases (Ace, Acy, Fluo, Chry or DahA) the low number of concentration series available may have contributed to this lack of relationships. Still, the general decrease of the PAHs incidence with an increase in latitude is in line with the aforementioned distribution of the sources (both in number and intensities) in the southernmost areas of the domain in study.

4. Human health guidelines

Legislation or guidelines for the presence of PAHs in the atmosphere are very scarce. Due to its proven carcinogenic properties, BaP has been taken as a reference to set some legal or reference limits, as the hazardous effects of this chemical can affect human health in a widespread range of ways (Gallo et al., 2008; Kim et al., 2013). According to the EEA, BaP emissions in the EU increased by 11% between 2002 and 2011, linked with a 24% rise in emissions from fuel consumption associated to commercial, institutional and household sectors (European Environment Agency, 2013). The EU established a target value of 1 ng/m^3 for the annual mean presence of BaP in the atmosphere not to be exceeded in the PM10 fraction (European Commission, 2005), and between 22% and 31% of the urban EU population was exposed to concentrations above that limit in the period 2009 to 2011, with a particular incidence in central and eastern Europe (European Environment Agency, 2013). If the World Health Organisation (WHO) reference level is considered (0.12 ng/m^3), this exposure climbs up to 94% of the EU urban population in 2011.

In this sense, we compiled the annual mean concentrations of BaP from the EMEP network and evaluated the levels exceeding the three reference values mentioned above: EU legal limit (1 ng/m^3), EU lower assessment threshold (0.4 ng/m^3) and WHO reference level (0.12 ng/m^3). In this section, data measured in air + aerosol (the core dataset of this work) was complemented to the levels in PM10 and only aerosols (Table 5). Results indicate that the highest limit is never surpassed, but the EU threshold and especially the WHO reference are exceeded in several occasions. In the first case, the BaP levels were above 0.4 ng/m^3 in Preila (Latvia) and Kosetice (Czech Republic) in the early 2000s and more recently in Diabla Gora (Poland), Illmitz (Austria) and Rucava (Latvia). All these stations are relatively close to each other (considering a continental scale) and these findings are in line with the EEA report stating higher BaP concentrations in the central/eastern part of Europe (European Environment Agency, 2013). Several other areas fall above the WHO reference, with an increasing frequency in the recent years and again mainly comprising central and eastern Europe. Nevertheless, also in High Muffles (UK) in 2008 and in Niembro (Spain) in 2010 this limit was exceeded in the PM10 fraction. On the other hand, Rörvik (Sweden) was slightly above the WHO threshold in 1995 but has been decreasing the presence of BaP ever since, as is the

Table 5
Values of BaP mean annual concentrations obtained from the EMEP sampling network surpassing guidance limits issued by the European Union (EU) and the World Health Organisation (WHO).

	Location	Country	Latitude	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
AIR+ AEROSOL	Svalbard	Norway	78°54' N	0,014	0,011	0,008	0,013		0,008	0,013	0,003	0,003	0,003	0,003	0,002	0,004	0,004	0,002	0,004	0,004
	Pallas	Finland	68°00' N			0,015	0,020	0,017	0,014	0,006	0,020	0,025			0,014	0,016		0,005		0,022
	Lahemaa	Estonia	59°30' N														0,097	0,171	0,144	
	Aspvreten	Sweden	58°48' N				0,058			0,048	0,039	0,073	0,053			0,073		0,059		0,055
	Birkenes II	Norway	58°23' N																0,033	0,029
	Rörvik	Sweden	57°25' N		0,126		0,100	0,064			0,078	0,070								
	Råö	Sweden	57°23' N									0,064	0,130	0,073	0,085	0,070	0,047	0,042		0,154
	Preila	Latvia	55°21' N							0,858	0,529	0,717								
	Westerland	Germany	54°55' N														0,093	0,113	0,086	0,157
	Zingst	Germany	54°26' N														0,158	0,195	0,192	
	High Muffles	UK	54°20' N											0,023	0,025	0,035	0,047			
	Houtem	Belgium	51°15' N																0,243	0,187
	Schmücke	Germany	50°39' N														0,114	0,117	0,108	0,148
	Kosetice	Czech Rep.	49°35' N						0,266	0,175	0,226	0,589	0,359	0,274	0,420	0,245	0,333	0,263	0,300	0,247
	Schauinsland	Germany	47°54' N														0,047	0,050	0,040	0,057
PM10	Rucava	Latvia	56°09' N																	0,381
	High Muffles	UK	54°20' N															0,149		0,072
	Diabla Gora	Poland	54°09' N															0,573		0,748
	Kollumerwaard	Holland	53°20' N																0,059	0,074
	De Zilk	Holland	52°18' N																0,094	0,091
	Illmitz	Austria	47°46' N																0,638	
	Iskrba	Slovenia	45°34' N															0,227	0,220	0,238
	Niembro	Spain	43°42' N													0,037	0,021			0,128
AEROSOL	Zoseni	Latvia	57°08' N											0,180	0,048	0,099	0,083	0,095		
	Rucava	Latvia	56°09' N											0,163	0,335	0,483	0,206			

Yellow highlight: WHO reference level = 0.12 ng/m^3; Salmon highlight: EU lower assess threshold = 0.4 ng/m^3

common rule for the Scandinavian countries. An exception can be found in Räo (Sweden), with 0.154 ng/m^3 in 2010.

Although all this dataset shows compliance with the EU legal limit, given that in several areas of Europe BaP levels are reported as below the EU lower assessment threshold of 0.4 ng/m^3 (European Environment Agency, 2013), these results highlight that special care must be taken to ensure the adequate measures for an effective protection the environment.

5. Conclusions

After this study, it can be said that the regions with the highest individual concentrations of PAHs within the considered domain are central Europe and the Baltic Sea areas. This reflects a significant presence of local concentration sources. The Finnish and Norwegian stations (located in higher latitudes) are recurrently those with lower levels, particularly Svalbard, where almost all the presence of PAHs is due to the long distance atmospheric transport. Seasonal trends are relatively constant, with higher percentages of PAHs occurring in winter and lower in summer, a reflection of increased emission sources as traffic or building heating in the colder months of the year. Spring and summer showed a similar behaviour, with an input of 15% and 25% to the annual total. The exception is High Muffles, in the UK, which shows greater homogeneity between seasons. This may be a reflection of the not so pronounced difference in temperature and intra-annual rainfall, which can motivate some constancy of the emission of these pollutants. Across the board the data series are highly variable, suggesting the existence of certain factors affecting higher levels in some cases and lower in others. Perhaps the most important is the existence of long time series that incorporate a significant intra-annual variability.

In terms of temporal trends, a downward trend in the levels of PAHs can be perceived as we approach the North Pole, the areas that supposedly have lower number of emission sources. This suggests that central Europe is the core of the historic PAH sources, presenting higher concentration values. The positive trends identified in the UK may be due to its own source fingerprint or transported from the east, showing annual patterns are different from the rest of stations. Moreover, negative concentration gradients *versus* latitude were found for four PAHs: Phen, Flt, Pyr and BghiP.

Another important finding is the concentration of BaP in Preila (Latvia), which is considerably high compared to other stations and almost reaches the mean annual EU legal limit for BaP in the atmosphere (1 ng/m^3). In fact, taking into account the several guideline values established by the European Union and the WHO, Europe may face in the near future an important problem derived from the increasing emissions of BaP and the other PAHs in the recent years. The lack of databases with extensive geographical coverage and long temporal periodicity hamper most of the efforts to assess this issue efficiently. This is why modelling approaches can give a very positive contribute and it use should be increasingly encouraged in future studies.

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