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Long-term monitoring and seasonal analysis of polycyclic aromatic hydrocarbons (PAHs) measured over a decade in the ambient air of Porto, Portugal

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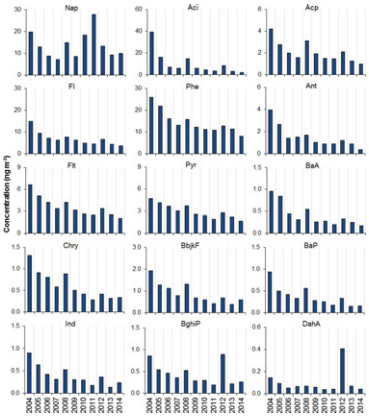
HIGHLIGHTS

- A trend of PAHs levels in ambient air over a decade (2004–2014) was studied.
- The PAHs concentration trend in ambient air showed a clear drop of the average values occurred during the period of study.
- Profiles of PAHs compounds, seasonal variations and possible sources of PAHs were assessed.
- TEF concept was applied and it was obtained the temporal evolution of carcinogenic potential.

GRAPHICAL ABSTRACT



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ABSTRACT

The present paper reports the analytical results of PAHs concentrations in ambient air obtained in the PM10 and gas-phase, from 2004 to 2014, in Porto, Portugal. As part of a monitoring programme conducted by IDAD — Institute of Environment and Development and supported by the regional municipal solid waste (MSW) management authorities, an extensive database of PAH concentrations in ambient air was collected in Porto's metropolitan area. During this period a total of 201 samples were collected in two sites classified as suburban. Analytical results showed a clear decreasing trend of total PAHs ($\sum$ PAH) and benzo[a]pyrene (BaP) levels during the period of study, especially in the first years of monitoring. The average annual concentrations of BaP were, throughout the monitoring period, lower than the target value for the annual average (1 ng m^{-3}) defined in the European legislation. PAHs levels showed a strong seasonality, with higher concentrations values during the colder months. The winter/summer ratio of $\sum$ PAH for the eleven years of study was 5, revealing the seasonal variation of PAHs in the studied area. The estimated toxic equivalency factors (TEFs) used to assess the contribution of the carcinogenic potential, confirmed a significant presence of the moderately active carcinogenic BaP and dibenz[ah]anthracene (DahA) in the samples collected in Porto. The ratio values of individual PAHs concentrations were used as diagnostic tool to identify the possible origin of PAH in the ambient air of Porto. Based on diagnostic ratios, it may be concluded that automobile traffic emissions, mainly related to diesel vehicles, were the

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major contribution of PAH levels in the ambient air, although some others contributions, such as coal and wood combustion, were identified.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a mixture of organic compounds released into the atmosphere as gases or particles during the incomplete combustion of organic material (EPA, 2001). PAHs are often attached to airborne particles.

PAHs may be originated from saturated hydrocarbons under oxygen-deficient conditions. Pyrolysis and pyrosynthesis are two main mechanisms that can explain the formation of these compounds (Ravindra et al., 2008). The major anthropogenic atmospheric emission sources of PAHs include biomass burning, coal and petroleum combustion, coke and metal production and road traffic (Zhang et al., 2005).

PAHs are ubiquitous in the environment (IARC, 2010; IPCS, 1998). Exposure to PAHs can occur through inhalation of ambient air and tobacco smoke and ingestion of water, soils, food and pharmaceutical products. They can bioaccumulate, especially in invertebrates (EEA, 2014; EEA (European Environment Agency), 2014) and have effects on human health, being carcinogenic, and on the environment, being toxic to aquatic life and birds.

The chemical structure of PAHs varies with the number of rings, and therefore, the molecular weights. Depending on their structure, PAH compounds are divided into lower molecular weight compounds, when they contain up to 4 aromatic rings, and high molecular weight compounds, with more than 4 aromatic rings.

Based on their genotoxic and carcinogenic properties, there are 16 species of PAHs classified by the United States Environmental Protection Agency (US-EPA) as priority pollutants in the list of hazardous air pollutants (US-EPA, 1990). One of the best characterized and most toxic PAH is benzo(a)pyrene, which is generally used as the indicator of PAHs.

To reduce atmospheric PAHs concentrations, efforts have been made to lower PAH emissions by implementing laws and regulations, by improving combustion technologies, and by employing cleaner fuel sources, such as natural gas (Krystyna et al., 2007). Temporal trend data of concentrations in ambient air are an important key to measure the effectiveness of source reduction measures and regulatory controls. Trends of PAHs in ambient air have been studied in several countries in different types of sites, though there is still no air quality strict standard for these compounds. European Union directive 2004/107/CE proposed a target value of 1 ng m^{-3} benzo[a]pyrene for the total content in the PM10 fraction averaged over a calendar year (EUD (European Union Directive), 2004). The UK expert panel recommends a guide value (annual average) of 0.25 ng m^{-3} benzo[a]pirene, value that is exceeded in most urban areas and near industrial facilities, which emit PAHs (Ravindra et al., 2008).

According to the European Union's emission inventory report, between 1990 and 2012, total PAHs emissions dropped in the EU-28 by 60%, 51% for benzo[a]pyrene, 41% for benzo[b]fluoranthene, 53% for benzo[k]fluoranthene and 39% for the indeno[1,2,3-cd]pyrene (EEA, 2014; EEA (European Environment Agency), 2014). This decrease of PAHs emissions in Europe was attributed generally to less residential use of coal, improvements in abatement technologies for metal refining and smelting, as well as stricter regulations on emissions from the road transport sector (EEA, 2014; EEA (European Environment Agency), 2014).

In Portugal, the database of PAHs concentrations in ambient air is still limited and there are no published studies about the evolution trend of these compounds in the atmosphere. In 1998 an ambient air monitoring programme for a municipal solid waste (MSW) incinerator (commonly referred to as LIPOR II) in the metropolitan area of Porto was designed, in order to evaluate the effects of the operation of this facility on the surrounding area (Coutinho et al., 1998). This programme is currently in operation and has allowed to obtain data about the long-term monitoring for several ambient air pollutants (Coutinho et al., 1998, 2006, 2007, 2015). PAHs levels have been monitored in ambient air since October 2004, in two sampling points in the region of Porto through this monitoring programme. The objective of the current study is to (1) investigate and describe the atmospheric behaviour of PAHs using temporal data series collected throughout 11 years of monitoring in Porto's area and (2) to identify the possible sources of the atmospheric PAHs levels.

2. Materials and methods

2.1. Sampling and analytical method

Since the beginning of their monitoring until the end of 2014 a total of 201 ambient air samples of PAHs were collected in the vicinity of the MSW incinerator LIPOR II providing an extensive characterization of the atmospheric levels of these pollutants in the metropolitan area of Porto, as well as information about the temporal trend of the atmospheric concentration of these compounds.

The present study focuses on the treatment and analysis of air quality data obtained at two sampling points: Leça do Balio and Vila Nova da Telha, located approximately 3 km around the incineration plant LIPOR II (Fig. 1). Both sites are located in the outskirts of the city of Porto and are part of the Metropolitan Area of Porto. The Metropolitan Area of Porto corresponds to 2% (2042 Km^2) of the Portuguese territorial area with around 1.7 million citizens that correspond to 17% of the Portuguese population.

In addition to small and medium local industries, the most significant sources of air pollutants in the nearby area are the waste incineration plant of LIPOR II, an oil refinery, a steel industry, a port and an airport infrastructure. The presence of a large network of highways is also a major source of emissions to the atmosphere in the surrounding region. Vehicular transport emissions have been considered the main source of atmospheric pollutants in the area (Pereira et al., 2005; Slezakova et al., 2011).

From October 2004 to March 2011 air samples were collected with a monthly frequency in each site using an ambient air sampling apparatus for PAHs, carried out according to the German guideline VDI 3498 Part 2, 2002. Since April 2011 the monitoring was restricted to a single site, Leça do Balio. The sampling apparatus consisted of a filter system, a mast, a suction pump, a gas volumeter and a timer. Particulate matter in the air was collected on a glass fibre filter. Filterpassing matter was collected on a polyurethane foam (PUF) absorption filter. To monitor the effectiveness of sampling, a second polyurethane foam sampling unit was connected downstream. The sampling head also contains a PM10 pre-separation head. The reference volume is the volume of air that is drawn through the sampling device and measured with a gas volumeter during the sampling period (24 h).

The PAHs samples which were deposited on the glass fibre filter and absorbed in the PUF, were extracted and cleaned up from interfering components in a multistage separation process. Then they were quantified by gas chromatography/mass spectrometry (GC/LRMS), according to the VDI 3874 German guideline.

2.2. Data analysis

The present paper focuses on the description and interpretation of PAHs concentration data obtained between October 2004 and

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