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Lichens as an integrating tool for monitoring PAH atmospheric deposition: A comparison with soil, air and pine needles

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Lichen PAH concentrations can identify geographic areas that may be out of compliance with regulatory standards.

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ABSTRACT

The aim of this study was to validate lichens as biomonitors of PAH atmospheric deposition; for that, an inter-comparison between the PAH profile and concentrations intercepted in lichens with those of air, soil and pine needles was performed. The study was conducted in a petro-industrial area and the results showed that PAH profiles in lichens were similar to those of the air and pine needles, but completely different from those of soils. Lichens accumulated higher PAH concentrations when compared to the other environmental compartments and its concentrations were significantly and linearly correlated with concentrations of PAHs in soil; we showed that a translation of the lichen PAHs concentrations into regulatory standards is possible, fulfilling one of the most important requirements of using lichens as biomonitors. With lichens we were then able to characterize the air PAHs profile of urban, petro-industrial and background areas.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are semi-volatile organic compounds, distributed both in the vapor- and particle-phases of the air. PAHs occur naturally in the environment, and are generated by forest fires and volcanic eruptions; however, the largest amount of PAHs is released into the environment by human activities (Edwards, 1983). Anthropogenic PAHs result mainly from pyrolytic processes, especially the incomplete combustion of organic materials during industrial activities, home heating, power generation, incineration and vehicle emissions (ATSDR, 1995; Garban et al., 2002; Dyke et al., 2003; Mastral et al., 2003), and as well as from petroleum cracking and refining in petrochemical industries, and during chemical manufacturing (Kaldor et al., 1984; Mehlman, 1992; Lin et al., 2001; Yang et al., 2002).

In recent years, PAHs have received increased attention in air pollution studies because some of them are highly carcinogenic and mutagenic (IARC, 1983). The US Environmental Protection Agency (EPA) has promulgated 16 unsubstituted PAHs (EPA-PAH) as priority pollutants to be monitored in the environment.

Several methods have been used to assess environmental levels of PAHs, such as soil and sediment, vegetation, food, water and air analyses (Srogi, 2007). The use of biomonitors (living organisms) to evaluate environmental contamination has advantages, as they are easier to sample, allow a long-term monitoring with a large number of sampling sites, and also the simultaneous determination of several pollutants within the same matrix (Wolterbeek, 2002). For air pollution assessment, lichens (symbiotic associations of fungi and algae/cyanobacteria), mosses and pine needles are the most commonly used organisms (Holoubek et al., 2000; Conti and Cecchetti, 2001; Onianwa, 2001; Migaszewski et al., 2002; Landers et al., 2008). Lichens have been used to monitor metals, sulfur, nitrogen, fluoride, radionuclides and a variety of organic compounds, such as dioxins and furans, polychlorinated biphenyls, and substances from organochloride pesticides (Villeneuve et al., 1988; Calamari et al., 1991; Garty, 2000; Augusto et al., 2004, 2007).

Regarding PAHs, there are only a few studies using lichens as biomonitors of these compounds (Herzig, 1989; Owczarek et al., 2001; Migaszewski et al., 2002; Guidotti et al., 2003; Domeño et al., 2006; Blasco et al., 2006, 2007, 2008; Naeth and Wilkinson, 2008; Shukla and Upreti, 2009). The majority of these studies was conducted in natural and forested ecosystems or in urban environments. To date, no comparison of PAH levels and profiles between

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lichens, soil, pine needles and air has been published for a highly industrialized (mainly petrochemical) and populated area. As PAHs are largely associated with industrial and urban activities, such a comparison will increase our ability to interpret lichen PAH profiles in past and future studies, especially for environmental authorities and decision makers.

The main aims of this study are: to compare PAH concentrations and profiles in lichens with those of soil, air and pine needles in a petro-industrial region; to characterize PAH profiles in urban, industrial and background areas; and to evaluate the possibility of cross-walking PAH concentrations in lichens into concentration units used in regulatory standards.

2. Materials and methods

2.1. Sampling for comparison between lichens, soil and pine needles

In January 2008, 34 samples of the species lichen *Parmotrema hypoleucinum* (Steiner) Hale, were collected at a number of sites within the highly industrialized region of Sines, located on the SW coast of continental Portugal, facing the Atlantic Ocean (Fig. 1). This region encompasses several important industrial facilities established since the late 1970s: a coal-fired power station, an oil refinery, a chemical plant and, more recently, an industrial landfill as well as many other smaller industries. Moreover, urban development has recently increased.

The lichen *P. hypoleucinum* was selected because it is ubiquitous and tolerates a variety of land-uses, such as urban, industrial, forestry and also background (unmanaged) areas. The collection was made mainly from branches and trunks of *Pinus pinea* L. (umbrella-pine) and *Quercus suber* L. (cork-oak). Samples were packed in brown glass bottles, protected from sunlight and immediately stored at 4 °C.

At 26 sites (24 coinciding with lichen sampling points), soil samples were also collected (Fig. 1). Samples were taken from the upper 5 cm of soil and placed in polyethylene bags. Once in the laboratory, soil samples were sieved through a 2 mm mesh screen, transferred to glass bottles in order to prevent adsorption by plastic, protected from sunlight and stored at 4 °C. At 10 sites (eight coinciding with lichen

sampling points), pine needle samples were also collected. Pine needles from *P. pinea* were selected because there are a considerable number of publications using this plant as biomonitor of toxic organic compounds, and because it is abundant in Europe. Samples were collected from the terminal part of branches, always at the same position on the tree, packed in brown glass bottles, protected from sunlight and immediately stored at 4 °C. All samples were extracted and analyzed for the 16 EPA-PAHs within two months.

2.2. Sampling for comparison between lichens and air

Particle-phase samples were collected at two sites (within the urban area of Sines and at the industrial area of Sines) using two high-volume air samplers, which operate with air flows of 66 m³/h and collect particles – PM10 (particles of 10 μm or less diameter) in the urban area and TSP (total suspended particles) in the industrial area – on 20.2×25.2 cm cellulose filters (Fig. 1). Over a two-month period (February and March 2008), 28 000 m³ of air was sampled at each site and 18 different samples were collected. Each sample corresponded to a 24 h period of continuous sampling. After sampling, the filters were dried, weighed and stored in the dark until analysis. Every 15-days two native lichen samples were collected close to each of the high-volume air samplers: *P. hypoleucinum* in the industrial area and Xanthoria parietina (L.) Th. Fr. in the urban area. Samples were packed in brown glass bottles, protected from sunlight and immediately stored at 4 °C. All samples were extracted and analyzed for the 16 EPA-PAHs; filters from each 15-days period (corresponding to the lichen samples exposure) were polled together.

2.3. Analytical procedure

All PAH analyses took place at the certified laboratory of the Portuguese Environmental Protection Agency (APA). For lichen, soil and pine needle analyses, approximately 2 g of sample was extracted in a Soxhlet with 200 mL of acetonitrile for 24 h. Each group of filters (corresponding to a 15-days sampling) was extracted as a whole. After extraction, all extracts were concentrated by rotary vacuum evaporation and cleaned-up in a florisil column with 30 mL of acetonitrile as eluting solvent. Subsequently, the extracts were again evaporated and concentrated with a gentle stream of purified $\rm N_2$ to 1 mL. The samples were analyzed by a high-performance liquid chromatograph (Hewlett Packard), using two columns (Agilent C18 and

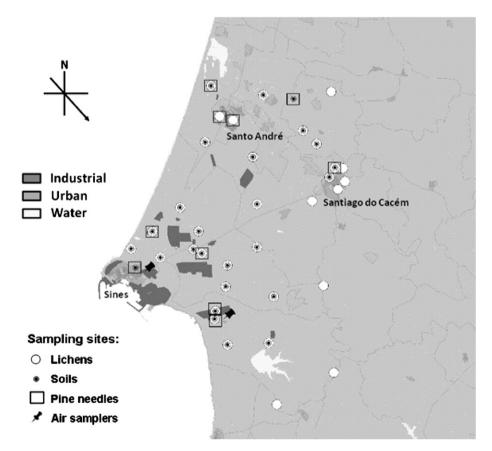


Fig. 1. Map of the study area (Sines), 20×30 km, showing the sampling sites for lichens (N = 34), soil (N = 26) and pine needles (N = 10), and the location of the two air samplers in the industrial and urban areas. Industrial and urban areas are represented by the darkest colors.

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