Environmental Pollution 208 (2016) 233–240

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Gaseous polycyclic aromatic hydrocarbon concentrations are higher in urban forests than adjacent open areas during summer but not in winter – Exploratory study

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ARTICLE INFO

Article history:
Received 1 June 2015
Received in revised form
23 August 2015
Accepted 3 September 2015
Available online 26 September 2015

Keywords:
PAH
Ecosystem service
Passive sampler
Urban forest
Urban vegetation

ABSTRACT

While the potential of plants to uptake polycyclic aromatic hydrocarbons (PAHs) is widely acknowledged, empirical evidence of the effects of this process on local atmospheric PAH concentrations and human health is tenuous. We measured gaseous PAH concentrations using passive samplers in urban tree-covered areas and adjacent open, treeless areas in a near-road environment in Finland to gain information on the ability of urban vegetation to improve air quality. The ability of urban, mostly deciduous, vegetation to affect PAHs was season dependent: during summer, concentrations were significantly higher in tree-covered areas, while in the fall, concentrations in open areas exceeded those in tree-covered areas. During winter, concentrations in tree-covered areas were either lower or did not differ from those in open areas. Results of this study imply that the commonly believed notion that trees unequivocally improve air quality does not apply to PAHs studied here.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are formed through the incomplete combustion of carbon-containing fuels and pyrolysis of organic matter, a major source of which is traffic (Marr et al., 1999; Shen et al., 2011). The main concern with PAHs is their slow degradation combined with carcinogenic properties of some of the hydrocarbons – especially those with four or more benzene rings (IARC, 2010; Wang and Busby, 1993). Although about 500 different PAHs have been detected in urban air, only benzo[a]pyrene (BaP), a known carcinogen (IARC, 2010), is typically measured and used as representative of all PAHs. Generally less data are available on ambient air PAH levels than classical criteria air pollutants – such as particles and nitrogen oxides – due to the rather complex and expensive sampling and analytical procedures required for PAHs (Boström et al., 2002). Only recently PAHs were included in official air quality standards, when the European Union set a target value (1 ng m^{-3}) for BaP (EEA, 2012).

PAHs display various gas/particle partitioning characteristics, depending on the properties of the specific compound, environmental conditions and the sorbing surface. 2- and 3-ring PAHs are mostly in vapor form, 4-ring PAHs appear in both gas and particle phases, while larger PAHs, such as BaP, are mainly associated with particles (Howsam et al., 2000 and references therein). Gaseous or semi-volatile PAHs that comprise a major fraction of PAHs in urban air are often classified as class 3 carcinogens, implying inadequate data for their classification. While carcinogenicity of many of these substances is still questionable, for example fluoranthene has been considered as a possible important contributor to cancer risk (Boström et al., 2002; Busby et al., 1984; IARC, 2010). According to Ramírez et al. (2011) cancer risk can be underestimated if gaseous phase PAHs are not considered. However, there is no documented evidence suggesting that gaseous PAHs in the urban air relate to human health.

Plants cover over 80% of the earth's terrestrial surface area and, due to their large surface area and cuticle coating comprised of lipids and waxes, have a great potential to trap atmospheric pollutants, such as PAHs (Simonich and Hites, 1994; Wagrowski and Hites, 1997). The main pathway of this removal is from air to leaf,

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http://dx.doi.org/10.1016/j.envpol.2015.09.009
0269-7491/

when PAHs are deposited on vegetation surfaces and diffused into the cuticle and further into the leaf interior (Bakker et al., 2001; Jouraeva et al., 2002; Kuhn, 1998; Wang et al., 2008). Factors affecting the transport of PAHs from air to plant are many, including ambient concentrations of the compound and its atmospheric phase (gaseous/particle associated), compound properties, leaf surface characteristics and air temperature (Howsam et al., 2001 and references therein). Intercepted PAHs in the foliage are eventually transmitted to the soil via litterfall and rain, or are resuspended and revolatilized back into the atmosphere (Howsam et al., 2001).

The phase of a particular PAH determines its deposition mechanism on vegetation: higher molecular weights and thus particle-bound PAHs remain on the surface of a leaf, gaseous PAHs accumulate in the cuticle and the lightest compounds can penetrate further into leaf tissues (Bakker et al., 2001). Seasonality in PAH dynamics (ambient concentrations and gas/particle partitioning) has been observed, being attributed mainly to elevated emissions during winters, and lower temperatures that affect gas/particle partitioning (Harrison et al., 1996; Howsam et al., 2001). For example Lang et al. (2007) observed that gaseous 3- to 6-ring PAHs were more abundant during summer, while particle-bound PAHs were typical during winter, but Ma et al. (2010) reported higher concentrations for both phases during the winter season.

The role of vegetation in absorbing PAHs has raised considerable interest during recent decades. The deposition of PAHs has been observed to be substantially higher into forest soils compared to an adjacent clearing (Horstmann et al., 1997; Matzner, 1984) due to vegetation uptake and subsequent transfer to the soil. Additionally, higher vegetation exerts a greater drag on the air, promoting downward fluxes of atmospheric compounds (Monteith and Unsworth, 2013). Deposition velocity of gaseous PAHs into the forest canopy has been observed to be relatively high, exceeding the deposition velocity of particle-bound PAHs (Horstmann and Mclachlan, 1998; Su et al., 2007). Furthermore, the dry deposition of both gaseous and particle-bound PAHs seems to be higher in a deciduous than coniferous forest (Horstmann and Mclachlan, 1998). Choi et al. (2008) measured PAH levels along a vertical gradient from below to above the canopy, and found that only the uptake of gaseous PAHs was fast enough to be detected as lowered concentrations at the upper part of the canopy. For particle-bound PAHs, the levels were more uniform at all heights, implying less efficient uptake of these PAHs (Choi et al., 2008).

Although vegetation has been suggested to remove various pollutants and thus improve urban air quality at the city- and local scale (e.g. Alberti, 2008; Nowak et al., 2006), contrasting views have been reported (e.g. Pataki et al., 2011; Vos et al., 2013). While vegetation on a large scale acts as an important vector of PAHs from the air into terrestrial systems (e.g. Böhme et al., 1999; Wania and McLachlan, 2001), empirical evidence of the significance of this removal capacity for ambient concentrations on a local scale is, to our knowledge, non-existent. A crucial, practical question is whether air quality and thus human well-being would be improved in the proximity of foliage or other adsorbing structures, or would it deteriorate due to, e.g. reduced turbulence and thus reduced pollution dispersion as modeled by Vos et al. (2013). Furthermore, seasonality in the effect of vegetation on PAH concentrations should be apparent, most obviously due to the lack of deciduous leaves in winter resulting in zero uptake by leaves (e.g. Söderström et al., 2005). Despite the substantial amount of literature in which various aspects of vegetation-PAH interactions have been assessed, to our knowledge there are no studies conducted in urban settings in which aerial concentrations of PAHs have been measured and compared between tree-covered and open locations during different seasons. Consequently, knowledge on the influence of urban vegetation on human health in terms of PAH removal is virtually non-existent.

The objective of this study was to test whether the normative approach according to which vegetation can efficiently improve local urban air quality applies to gaseous PAHs. We hypothesize that (i) ambient air is cleaner, in terms of gaseous PAHs, in tree-covered areas than in adjacent open areas in urban near-road environments and, (ii) that the potential of urban parks/forests to remove PAHs is season dependent, resulting in lower relative concentrations under canopies during leafy period. This study is part of a larger study in which the impact of vegetation on several air pollutants is explored (see Setälä et al., 2013).

2. Materials and methods

2.1. Study area

We measured PAH levels using passive sampling devices (PSDs) in urban remnant forest and park areas and in adjacent open (treeless) areas in the cities of Helsinki (60°10′15″N, 24°56′15″E) and Lahti (60°59′00″N, 25°39′20″E) in southern Finland. Ten sites in each city were established close to roads with moderate to heavy traffic flows. Each site included a pair of sampling points: one in an open area and another in a tree-covered area, typically less than 100 m apart (altogether 40 sampling points in the two cities). The open areas were typically small or medium-sized city squares or other treeless areas close to a road where soil surfaces were either impervious (e.g. asphalt) or pervious with mown lawns. The tree-covered areas were either urban parks of various sizes with scattered, mainly deciduous trees and lawns, or urban forest remnants of mixed forest type (deciduous and coniferous trees) and rich understorey vegetation. Within each site, the samplers were placed at equal distances from the road where possible, but not always on the same side of the road. Distances from the measuring points to the road varied between 8 and 67 m, and average distances for open and tree-covered measuring points were 32.5 m and 36.5 m, respectively. A set of environmental variables were measured from all sites (Table 1). The sampling sites and setup are described in detail in Setälä et al. (2013).

Measurements of PAHs were conducted during two seasons: from 9 August to 10 September 2011 (late summer, referred to as summer) and from 7 March to 11 April 2012 (late winter, i.e. winter). The PSDs were deployed for 30 days during both periods.

2.2. Time series measuring campaign

A time series measuring campaign was conducted two years after the main study in 2013, to confirm previous results and to acquire more detailed information on the PAH concentrations during the growing season. Two new study sites were selected, both with an open (meadow, set-aside field) and tree-covered (remnant forest) location at similar distances from the adjacent road. The one site pair was at the outskirts of the city of Lahti, situated 39 m from a road with 20,000 cars per day, and the other pair 45 m from a highway with 20,000–40,000 cars per day (City of Lahti, 2012). Gaseous PAHs were sampled during five sampling periods between May and November, each sampling period lasting approximately 30 days. The periods were 28 May–28 June, 28 June–5 August, 8 August–12 September, 13 September–14 October and 16 October–11 November.

2.3. Sampling method

The passive sampler device (PSD) used in this study was similar to semipermeable membrane device (SPMD) (Huckins et al., 1990),