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LOCALIZATION OF DEPOSITED POLYCYCLIC AROMATIC HYDROCARBONS IN LEAVES OF *PLANTAGO*

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Abstract—After deposition to foliage, polycyclic aromatic hydrocarbons (PAHs) may remain on the leaf surface, accumulate in the cuticular wax, or diffuse into the remaining interior of the plant. In a field study, the location of deposited PAHs in the leaves of two *Plantago* species was determined. To this aim, leaves of *Plantago major* and *Plantago media* were divided into three fractions. First, the leaves were washed (wash-off fraction), then cuticular wax was extracted (wax fraction). Finally, the remaining leaf material was extracted (interior fraction). The presence of PAHs could be demonstrated in all three fractions. For both plants, the distribution of PAHs over the three fractions changed with molecular weight (mol wt) of the PAHs. The wash-off fraction increased with increasing molecular weight, likely because high molecular-weight PAHs occur predominantly bound to particles, which can be readily washed off from the leaves. In contrast, the amount of PAHs detected in the interior of the leaves decreased with increasing molecular weight. This can be explained by a slow desorption of the PAHs from the particles and a low diffusion rate of the larger molecules. This study shows that washing reduces the amount of high molecular-weight PAHs on plant surfaces. Therefore, washing of leafy vegetables is important to minimize human dietary intake of PAHs.

Keywords—Polycylic aromatic hydrocarbons Plants Deposition Foliar uptake

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are products of incomplete combustion, which are omnipresent in the atmosphere. These compounds can, just as semivolatile organic compounds in general, be deposited to plant surfaces (e.g., [1–3]). This accumulation process causes indirect human exposure to PAHs through the consumption of leafy vegetables [4]. Many of the PAHs are known carcinogens or mutagens, especially the ones with a relatively high molecular weight, such as benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and PAHs with a higher molecular weight [5].

During accumulation of semivolatile organic compounds in plants, they first adsorb to the surface and then diffuse into the cuticle of the plant [6]. The cuticle is an extracellular, nonliving, lipid layer, which forms the interface between the atmosphere and the plant, thus protecting the plant from desiccation [7]. It is usually characterized by the presence of two specific classes of lipids, i.e., soluble waxes and insoluble polyester cutins [8].

In several studies in which needles and leaves were separated in a cuticular wax and an inner compartment by means of fractionated extractions, semivolatile organic compounds were present in both compartments [9–13]. Hence, compounds may accumulate in the cuticular waxes but may also diffuse through the waxes to accumulate in the cutin or possibly the interior of the needle/leaf.

Polycyclic aromatic hydrocarbons are present in the atmosphere both in the gaseous phase and bound to particles. The particle-bound fraction increases with increasing K_{OA} (octanol/air partition coefficient) of the compound [14]. For example, air measurements in Manchester in 1991 showed that 99% of the PAH phenanthrene (mol wt 178, log K_{OA} 7.6 [15]) occurred in the gaseous phase [16]. For the larger PAH pyrene (mol wt 202, log K_{OA} 8.8 [15]), this percentage amounted to 87%, whereas the high molecular-weight PAH benzo[*a*]pyrene (mol wt 252, log K_{OA} 9.6–11.6, calculated from K_{OW} [17] and reported values for Henry's Law constant [18]) was predominantly (>99%) present bound to particles [16].

The question arises whether the fate of deposited particle-bound PAHs is analogous to that of gaseous PAHs. Larsson and Sahlberg [19] demonstrated that washing of lettuce with water removed a considerable amount of the high molecular-weight PAHs but little of the small PAH phenantrene. Therefore, they suggested that only PAHs with a low molecular weight could be sorbed into the cuticle. High molecular-weight PAHs, mainly associated with particles, would remain on the leaf surface [19]. In contrast with these findings, Kaupp [12] rinsed corn leaves with water and ethylenediaminetetraacetic acid (EDTA) solutions and found that only a small fraction of the high molecular-weight PAHs and polychlorinated dibenzo-*p*-dioxins and dibenzofurans was removed in this manner. This suggests that compounds can be desorbed from particles and diffuse into the cuticle or that the particles may be encapsulated in the cuticle [12].

Because of the conflicting results of the studies mentioned above, the location of deposited PAHs in or on plant leaves is unclear. The aim of the present study was to localize PAHs in leaves of *Plantago major* and *Plantago media*. To this end, we determined the distribution of PAHs over three fractions

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of plant leaves, i.e., that attached to the leaf surface, that sorbed in the extractable cuticular wax, and that in the inner parts of the leaf. *Plantago* species are herbal plants, which were previously used in accumulation experiments with PAHs [20].

EXPERIMENTAL

Chemicals

Phenantrene (PHE), anthracene, benzo[*a*]anthracene, pyrene, chrysene, and benzo[*a*]pyrene (B*a*P) were purchased from Sigma (St. Louis, MO, USA). Benzo[*k*]fluoranthene (B*k*F) was obtained from Chem Service (West Chester, PA, USA), benzo[*g,h,i*]perylene from Fluka (Buchs, Switzerland), and fluoranthene from Aldrich (Steinheim, Germany). We chose these eight PAHs since, although they do not represent the whole range of molecular weight, they cover the whole range in gas-particle distribution in the atmosphere. In addition, the selected PAHs include the most abundant compounds (PHE and fluoranthene) and the most carcinogenic one (B*a*P).

Benzo[*e*]acephenantrylene (used as internal standard) and EDTA were purchased from Aldrich (Steinheim, Germany). Serdolit (PAD I) was purchased from Brunschwig (Amsterdam, The Netherlands) and was prewashed with acetonitrile, methanol, and ethanol before use. Methanol (high-performance liquid chromatography [HPLC] gradient grade) and acetonitrile (HPLC gradient grade) were obtained from Baker (Deventer, The Netherlands). Dichloromethane (DCM, HPLC grade) was purchased from Merck (MSD, Haarlem, The Netherlands). All other solvents were of analytical quality and were obtained from Baker (Deventer, The Netherlands).

Plant and site description

Plantago species are herbs with a short stem. The leaves usually arise from the base of the stem and are spirally arranged. Leaves of *P. major* (L.) are broad, almost glabrous, and are relatively free-standing in the air. *Plantago media* (L.) leaves have a dense layer of silky hairs. They are closely spreading on the ground and partly cover each other. *Plantago major* (seeds from wild origin) and *P. media* (seeds from Meise, Belgium) were grown in a greenhouse (temperature, 28°C). In July 1999, at the age of five months, the plants were planted in a plot of the Botanical Gardens of Utrecht University (Utrecht, The Netherlands). This site is considered an urban area. The distance to the nearest highway is about 400 m and to downtown Utrecht approximately 4 km. To prevent contact of the leaves with the soil, the ground was covered with a plastic cloth. Small pieces were cut from the cloth, after which the plants were placed in the created holes. The soil under the holes was covered with small pebbles. During the experiment, temperatures varied from 13 to 23°C (average 18.6°C). The experiment lasted 72 d, with rain events on 19 d (on 6 d, >5 mm rain). From day 57 until day 72, the plants were watered twice a week (without wetting the leaves) since in this period there was only 1 mm of rainfall (data from weather station De Bilt, which is 1.4 km from the site). After 72 d in the field, the plants were harvested. From *P. media* plants, young leaves (3–5 weeks old) were sampled from the top and old leaves (8–10 weeks) from the lower parts of the plant. From *P. major*, only young leaves (five–seven weeks) were sampled.

Samples

Leaf samples (15 g, quadruplicates) consisted of 3 to 4 leaves (*P. major*) or 5 to 10 leaves (*P. media*), originating from two individual plants. Leaf fresh weight was determined. The samples were separated into three fractions. First, they were washed twice by shaking with 100 ml (for 90 and 30 s, respectively) of an EDTA solution (3×10^{-2} M, pH 5) in a beaker. The EDTA solution was extracted by refluxing with 100 ml cyclohexane for 10 min. Following the washing procedure with EDTA, leaves were immersed in 2×45 ml DCM for 30 s and 10 s, respectively, to extract cuticular wax. The DCM was concentrated until ±1 ml under a nitrogen flow. To this extract, 45 ml of methanolic potassium hydroxide (KOH) was added. Subsequently, the solution was refluxed for 30 min. Cyclohexane (100 ml) was added, and this was refluxed for 10 min. After extraction of cuticular wax, the leaves were ground with liquid nitrogen, 45 ml of methanolic KOH were added, and the same procedure was followed as used for the cuticular wax. All cyclohexane fractions (washing water, cuticular wax, and leaf interior) were concentrated to 1 ml under a gentle stream of nitrogen. Five milliliters of MeOH was added and the solution was concentrated to 2 ml. Cleanup of the samples took place on columns (Ø 10 mm) filled with 2.2 g Serdolit. After addition of the sample, EtOH (5 ml), pentane (5 ml), and ethanol (5 ml) were eluted and discarded. The acetonitrile (45 ml) and ethanol were eluted. This was evaporated to 0.5 ml under nitrogen and analyzed by HPLC with fluorescence detection, as described in Bakker et al. [20].

Control samples

Procedural blanks were determined by extraction and cleanup of 90 ml DCM ($n = 2$) or 45 ml MeOH/KOH ($n = 3$). Recovery of the PAHs was determined by spiking a known amount of PAHs to leaf samples with known background concentrations and calculating the percentage recovered after cleanup (subtracting the background). Recoveries of the PAHs varied from 72 to 90% (±10–13%), except for B*k*F and B*a*P, which amounted to 64 ± 14 and 47 ± 12%, respectively.

It is not possible to determine the leaf dry weight, the wax content, and the specific leaf area (cm^2 leaf area/g dry wt) for the samples in which PAHs were determined without losing PAHs from the samples. Therefore, these values were determined in 15 g leaf material ($n = 1$) of plants that was not subjected to PAH analysis, according to the methods described in Bakker et al. [20]. These values were used to recalculate the PAH concentrations per gram fresh weight into concentrations per gram dry weight and per square centimeter of leaf.

The efficiency of the washing procedure with EDTA solution was checked and compared with that of water. This was done by washing leaves three times with 100 ml water or EDTA solution instead of two times with EDTA solution ($n = 3$).

Quantification of PAHs

Benzo[*g,h,i*]perylene (mol wt 276) could not be determined quantitatively due to the presence of an interfering peak of an unknown substance in the chromatogram. The presence of phenantrene in the fractions was sometimes difficult to prove due to high concentrations of PHE in the procedural blanks (72 ± 18 ng). Quantification of B*a*A and chrysene also suffered from relatively large blanks (7 ± 3 and 6 ± 1 ng, respectively). Only values that exceeded the average concentration in the blanks plus two times the standard deviation were taken into account.

The PAH concentrations were calculated by correcting for recovery percentages and subtracting the average amount measured in the blanks. Reproducibility of the measurements was

relatively low; the relative standard deviation varied from 5 to 80%, being <50% in 60% of the cases. In this article, PAH concentrations are expressed on a dry weight basis. The measured leaf parameters (fresh weight, leaf surface, and wax content) are similar for the different groups of leaves (data not shown). Therefore, plots based on these parameters will result in similar figures.

RESULTS AND DISCUSSION

Evaluation of the fractionation procedure

In the control experiment, no differences were found between the washing efficiency of water and the EDTA solution. Although EDTA was mentioned as effective for removing particles from leaves [12], in this study, water appears just as efficient.

Washing leaves for a third time with 100 ml (instead of twice) rendered solutions with amounts of PAHs with molecular weight of 178, 202, or 228 not exceeding those in the blanks. This demonstrates that these compounds were already washed off by the first 200 ml. However, significant amounts of B*k*F and B*a*P (mol wt 252) were present in the third washing solution. The behavior of B*k*F and B*a*P was similar. Whereas 69 ± 2% and 16 ± 3% were present in the first and second washing solutions, respectively, in the third wash, 13 ± 2% of the two compounds was found.

The effect of cross-contamination of the other fractions is likely smaller but cannot be completely ruled out. Although the wash-off fractions did not contain measurable amounts of cuticular wax, small quantities of wax may have eroded during the washing procedure, thereby contributing to the amount of PAHs in the wash-off fraction. Extraction of PAHs from the interior of the leaves during cuticular wax extraction is not very likely, as during the short extraction times (30 + 10 s) chlorophyll was not extracted [21]. On the other hand, no large contribution of wax-borne PAHs is expected in the interior fraction, as experiments have shown that additional cuticular wax was not extracted with DCM after 30 s (data not shown). However, it should be noted that the collected fractions are operationally and not physically defined.

PAH concentrations of whole leaves

The PAH concentrations of the whole leaves were calculated by adding the amounts of the three fractions and dividing the total amount by the dry weight of the sample. The PAH concentrations of the whole leaves (the measured PAHs grouped by molecular weight) varied from several tens of nanograms to more than 1 μg/g dry weight (Fig. 1). Throughout this article, molecular weight 178 represents PHE and anthracene; molecular weight 202 is fluoranthene and pyrene. The molecular weight 228 is B*a*A and chrysene, while molecular weight 252 represents B*k*F and B*a*P. The differences between the PAH concentrations of the two kinds of *P. media* leaves (Fig. 1) are caused by the differences in exposure period, i.e., old *P. media* leaves (8–10 weeks) have higher concentrations than young ones (3–5 weeks). While young *P. media* leaves have the shortest exposure period, the PAH concentrations of molecular weight 202, 228, and 252 in these leaves are higher than those of *P. major* (5–7 weeks). The only exception is formed by PAHs with molecular weight 178, the presence of which could not be shown in young leaves of *P. media.* As high molecular-weight PAHs are predominantly bound to particles in the atmosphere [12,16], the higher concentrations of these compounds in young *P. media* leaves are probably due to their hairy surface [20].

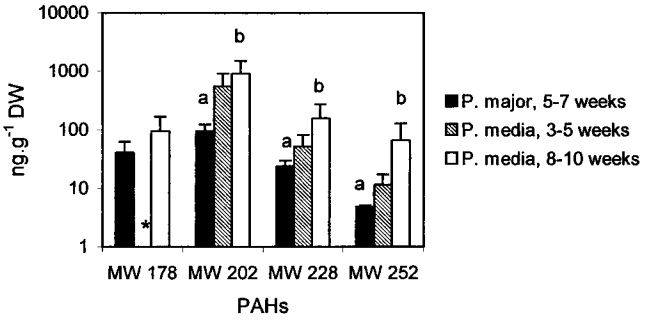

Fig. 1. Polycyclic aromatic hydrocarbon (PAH) concentrations (ng/g dry wt) in the leaves of *P. major* (5–7 weeks old, black bars) and in young leaves (3–5 weeks, hatched bars) and old leaves (8–10 weeks, white bars) of *P. media.* The PAHs are categorized into groups according to their molecular weight (mol wt). Error bars represent standard deviations of the four replicates. Statistically significant differences between the average concentrations of the groups (analysis of variance [ANOVA], Prism 2.01, $p < 0.05$) are indicated in the figure with letters a and b. *, The amount of PAHs in the samples was less than the amount in the procedural blanks plus two times the standard deviation (for mol wt 178, i.e., <108 ng).

Since the hairy leaves of *P. media* can more effectively retain particle-bound PAHs than the glabrous *P. major* leaves [20], it was expected that the wash-off fraction of high molecular-weight PAHs was higher for *P. media* than for *P. major.* Although the average wash-off fraction of the old *P. media* leaves was indeed consistently higher than that of *P. major,* the differences were not statistically significant. This is due to the relative high variation in the data.

Distribution of PAHs over three fractions

The distribution of the PAHs over the three fractions, i.e., wash-off, cuticular wax, and remaining leaf interior, changes with the molecular weight of the PAHs for both plant species (Fig. 2). The fraction found in the washing solution increases with increasing molecular weight of the PAHs, while the fraction in the leaf interior decreases. This trend is evident for all three kinds of leaves and will be discussed below.

The PAH distribution over the three fractions is similar for the three groups of leaves. Although the wash-off fractions seem to be largest for old *P. media* leaves and smallest for leaves of *P. major,* the differences are not statistically significant due to the relatively large standard deviations of the PAH concentrations of the fractions.

Wash-off fraction. The increase of the wash-off fraction with increasing molecular weight of the PAHs is most likely due to the wash-off of particles. As high molecular-weight PAHs have larger particle-bound fractions in the atmosphere, this leads to a higher contribution of particle-bound deposition to plant leaves. This is in agreement with McLachlan's framework for the interpretation of measurements of semivolatile organic compounds in plants [22]. In this framework, the dominant deposition processes are distinguished as a function of the K_{OA} of the compound. For compounds with high K_{OA} values, such as high molecular-weight PAHs, particle-bound deposition is the controlling process.

Due to the inefficiency of the used method to wash off PAHs with molecular weight 252, as mentioned earlier, the wash-off fraction of this group of compounds is somewhat (10–15%) underestimated in Figure 2. Consequently, the frac-

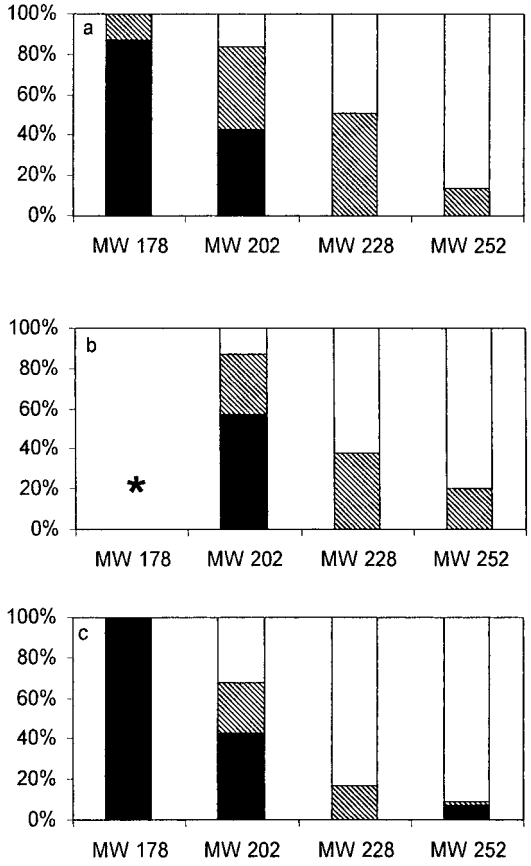

Fig. 2. Distribution of polycyclic aromatic hydrocarbons ([PAHs] according to their mol wt) over the three fractions wash-off (white bars), extractable cuticular wax (hatched bars), and leaf interior (black bars) for (**a**) leaves (5–7 weeks old) of *P. major*, (**b**) young leaves (3–5 weeks) of *P. media*, and (**c**) old leaves (8–10 weeks) of *P. media*. *, None of the fractions contained amounts exceeding those in the procedural blanks plus two times the standard deviation (for mol wt 178, i.e., <108 ng).

tion in the cuticular wax is overestimated due to the amount that was not washed off with EDTA but extracted together with the cuticular wax. Correction of the fractions for this artifact makes the observed trend (increasing wash-off fraction with increasing molecular weight of the compounds) even more evident.

The presence of the high molecular-weight PAHs in the wash-off fraction agrees well with the results of the study of Larsson and Sahlberg [19]. These authors also showed a large wash-off of high molecular-weight PAHs; washed lettuce leaves contained only a part of the PAHs of nonwashed leaves (30–60% for PAHs with mol wt 228, 13–17% for mol wt ≥252). In contrast, concentrations of the low molecular-weight compound PHE in washed leaves amounted to 90% of the concentrations of the nonwashed leaves. However, the results of Kaupp's study [12] are different. This author found 4% of high molecular-weight PAHs and polychlorinated dibenzo-*p*-dioxins and dibenzofurans in water that was used to rinse corn leaves and at most 20% in the EDTA solution in which the leaves were subsequently shaken. Together, only 24% of the high molecular-weight compounds were located at the leaf surface. It is not clear what caused the difference between the results of Kaupp [12] on the one hand and those of Larsson and Sahlberg [19] and the present results on the other hand. The differences may be due to differences in plant characteristics, the thoroughness of the washing procedure, and the environmental conditions. For example, in this study, the collection of particles was probably very effective since there was hardly any natural washing during the experiment (less than 1 mm of rain during the last three weeks). This may have resulted in relatively high amounts of particle-bound PAHs. On the other hand, the relatively high temperatures during the experiment may have increased the gaseous to particle-bound concentration ratio of PAHs in the atmosphere. In addition, differences in roughness of the leaf surface (particle collection efficiency) and/or in epicuticular wax characteristics (particle embedding efficiency) between the plants of the different studies may have been responsible.

Interior fraction. In contrast with the wash-off fraction, the fraction of PAHs that is present in the interior of the leaves decreases with increasing molecular weight of the compound (Fig. 2). This can be explained by the fact that low molecular-weight PAHs can reach the leaf interior in a relatively short time. High molecular-weight compounds will need more time to cross the cuticular waxes. As compounds can move through the cuticle exclusively by molecular diffusion, their mobility is inversely proportional to their molar volume [23,24]. Furthermore, as these compounds occur predominantly bound to particles, they will have to desorb from the particles before entering the cuticle. Because these compounds are very hydrophobic, they have a low tendency to desorb from the particle and therefore to enter the wax. Another entry route may be the encapsulation of the PAH particle complex by the cuticular wax. Since the complex will have a lower diffusion rate due to its larger volume, this process will take considerably more time than diffusion of a molecule of a low molecular-weight PAH.

Whereas PAHs with molecular weight 178 and 202 are present in the interior of all leaves (with one exception, see Fig. 2), only the interior of old *P. media* leaves contained PAHs with molecular weight 252 in this case B*k*F (Fig. 2). The other leaves probably did not live long enough to allow diffusion of B*k*F into their interiors. This demonstrates that a compound with molecular weight 252 can traverse the cuticular wax of these leaves within a period of 8 to 10 weeks. The presence of PAHs with molecular weight 228 in the leaf interiors could not be demonstrated due to the relatively high blanks of these compounds.

The occurrence of high molecular-weight compounds in the interior of needles and leaves is consistent with the results of other research [9–13]. In several of these investigations [9,10,12], just as in the present study, the amount of compound found in the inner leaf/needle was correlated to the molecular weight of the compound. Nevertheless, since pine needles can have exposure times up to several years, large fractions are sometimes found in the interior of these plants [9,13].

CONCLUSION

The location of airborne PAHs in leaves of *P. major* and *P. media* is related to the molecular weight of the compounds. Low molecular-weight PAHs, for which gaseous deposition is dominant, are predominantly present in the cuticular wax and the interior of the leaves, whereas high molecular-weight PAHs, deposited via particles, mainly remain at the leaf surface.

Washing of *Plantago* leaves, either with water or with an EDTA solution, efficiently reduces the amount of high molecular-weight PAHs, including the potent carcinogens ben-

zo[*k*]fluoranthene and benzo[*a*]pyrene. Consequently, although the plant species and the weather conditions in this study may not be representative for the growing of food crops in general, this shows that human intake of high molecular-weight PAHs can be reduced by the thorough washing of vegetables.

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