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DRY DEPOSITION OF ATMOSPHERIC POLYCYCLIC AROMATIC HYDROCARBONS IN THREE *PLANTAGO* SPECIES

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Abstract—The concentrations of polycyclic aromatic hydrocarbons (PAHs) in the leaf wax of three *Plantago* species were determined weekly for 3 weeks. The almost glabrous, free-standing leaves of *Plantago major* and the sparsely hairy *Plantago lanceolata* leaves were more heavily contaminated with low molecular weight (MW) PAHs (MW $<$ 228) than the densely hairy, partly overlapping *Plantago media* leaves. This may be caused by the lower canopy roughness (higher aerodynamic resistance), the higher amount of leaf hairs (higher boundary resistance), and/or the higher leaf overlap (smaller accessible leaf area) of *P. media*. On the other hand, PAHs with MW $\geq$ 252 tended to show higher concentrations in *P. media* than in the other two species. This is likely caused by the dense layer of hairs on *P. media* leaves, which can efficiently intercept the largely particle-bound high MW PAHs. When the PAH concentrations were normalized to projected leaf surface area, the differences between *P. media* and the other two species became significant ($p < 0.05$) for the high MW PAHs, while the differences for the low MW PAHs decreased. Although the differences in PAH concentrations between species are relatively small (factor 2–5), this study clearly shows that plant architecture and leaf hairs influence the dry deposition of PAHs.

Keywords—Polycyclic aromatic hydrocarbons Plants Foliar uptake Dry deposition Plant characteristics

INTRODUCTION

The aerial parts of all terrestrial plants are covered with a layer of hydrophobic cuticular wax. This wax accumulates semivolatile organic compounds (SOCs) from the ambient air, such as polychlorobiphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and polychlorodibenzo-p-dioxins and -furans (PCDD/Fs) (e.g., [1–3]). Vegetation thus constitutes a sink for atmospheric SOCs and is a basis for food chain transfer to animals and humans [4].

Besides the physicochemical properties of the compound and environmental conditions, the deposition of SOCs in vegetation is also determined by the characteristics of the plant [5]. To date, the influence of a limited number of plant characteristics on the deposition of SOCs has been studied. The variation in PAH concentrations of plant samples from a field experiment considerably decreased after normalization to the amount of plant lipids [6]. In a laboratory study in which an equilibrium between plant and air had been reached, differences in bioconcentration factors of different plants were attributed to plant lipid composition [7]. In nonequilibrium situations, uptake rates were shown to be dependent on exposed surface area of leaves and fruits [8] and total specific surface area of needle wax (determined with surface adsorption of pentachlorophenol) [9]. Although these studies have started to unravel the complex role plant characteristics play in foliar uptake of SOCs, a number of important properties remain to be investigated, such as the composition and structure of cuticular waxes, the architecture of the plant, and the presence of leaf hairs.

The uptake of compounds from the atmosphere in plant leaves involves three steps (Fig. 1). First, the contaminant is transported from the (turbulent) atmosphere to the laminar air boundary layer surrounding the leaf. In the second step, the contaminant has to be carried across the boundary layer. In this layer, the airflow is parallel to the leaf surface and the wind speed is highly reduced but increases with distance from the surface [10]. While in the bulk atmosphere, contaminants are transported convectively by turbulent wind eddies; gaseous compounds and particles $<$0.1 μm [11] can only be carried through the laminar boundary layer by Brownian motion, which is three to seven orders of magnitude slower than transport under turbulent conditions [12]. The final step in the uptake process is the interaction of the compound with the leaf surface. For gases, this means adsorption to the surface or permeation into the cuticle [9]. Deposited particles may simply adhere to the leaf surface, react with it chemically, or bounce off if the surface is smooth [11]. The dominant resistance to the uptake of SOCs is in the atmosphere or in the plant, depending on environmental conditions, plant characteristics, and the properties of the compound. The main compound property determining the dominating resistance is the K_{oa} (the partition coefficient between octanol and air), which is a descriptor for the partitioning between leaf wax and air. For compounds with a low K_{oa}, the cuticle is relatively impermeable and the plant resistance is the main resistance. On the other hand, compounds with a high K_{oa} (e.g., PAHs, PCDD/Fs) are highly soluble in the cuticle and therefore atmospheric resistance limits uptake [13].

The atmospheric resistance (consisting of the aerodynamic and the boundary resistance, corresponding to, respectively, steps 1 and 2 in Fig. 1) is influenced by the plant architecture as well as the shape of the leaf surface. The surface roughness

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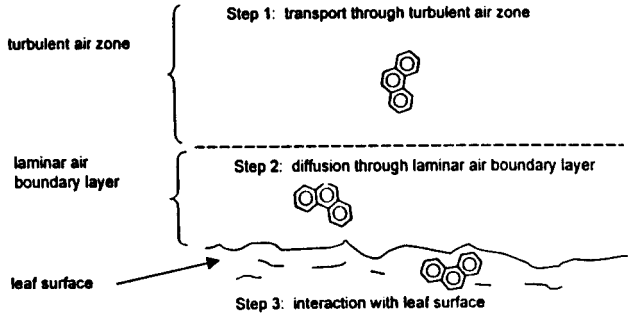

Fig. 1. Schematic representation of the three steps in the uptake process of hydrophobic organic compounds from air in leaf wax.

of the canopy influences aerodynamic transport (high roughness increases transport), while the roughness of the leaf surface is one of the factors that determines the thickness of the laminar boundary layer (δ_{bl}). The δ_{bl} is also influenced by wind speed and irradiation.

The PAHs are present in the atmosphere both in the gaseous phase and bound to particles. While PAHs with molecular weight (MW) <252 are predominantly present in the gaseous phase, large particle-bound fractions are found for PAHs with higher MW due to their low vapor pressures and high K_{oa} values [14,15]. Nakajima [16], and Kaupp [15] concluded from field experiments that for high MW PAHs the dry deposition of particles is a significant pathway for deposition on vegetation.

The deposition of particles is dependent on particle size, plant characteristics, and environmental conditions [17]. Several plant characteristics have been related to particle deposition. As for gases, a high aerodynamic surface roughness leads to efficient turbulent transport of particles [18]. Particle deposition on different types of vegetation was found to increase with increasing leaf area index [19,20]. Wind tunnel experiments with radio-labelled particles with sizes ranging from 0.03 to 44 μm have shown that hairy leaves are better particle collectors than glabrous leaves (e.g., [21–24]).

Plant architecture and leaf hairs may thus affect the deposition (rate) of gases and of compounds bound to particles. In this study, we compared the dry deposition of gaseous and particle-bound PAHs in three species of *Plantago* that differ in the amount of leaf hairs and in the architecture of the plant but have similar wax characteristics.

MATERIALS AND METHODS

Chemicals

Phenantrene (PHE, MW 178), anthracene (ANT, MW 178), benz[*a*]anthracene (B*a*A, MW 228), chrysene (CRY, MW 228), and benzo[*a*]pyrene (B*a*P, MW 252) were obtained from Sigma (St. Louis, MO, USA). Benzo[*k*]fluoranthene (B*k*F, MW 252) was obtained from Chem Service (West Chester, PA, USA) and benzo[*g,h,i*]perylene (B*ghi*P, MW 276) from Fluka (Buchs, Switzerland). Fluoranthene (FLUO, MW 202) and 5α-cholestane were purchased from Aldrich (Steinheim, Germany).

Chloroform (p.a.) and diethylether (p.a.) were obtained from Merck (Darmstadt, Germany), methanol (high performance liquid chromatography [HPLC] gradient grade) and acetonitrile (ACN, HPLC gradient grade) were obtained from Baker (Deventer, The Netherlands). Octadecylsilica (C_{18}) was purchased from Baker and prewashed with methanol and ACN before use.

Plants

Plant species. Plantago species are herbs with a short stem. The leaves usually arise from the base of the stem and are spirally arranged. The leaves of *Plantago major* and *Plantago lanceolata* are relatively freestanding in the air, whereas the leaves of *Plantago media* are closely spreading on the ground and partly cover each other. *Plantago major* leaves (length 10–15 cm) are broad and almost glabrous, while *P. lanceolata* has sparsely hairy lanceolate leaves (length 10–15 cm) and *P. media* has ovate leaves (length 5–10 cm) with a dense layer of silky hairs.

Growth conditions. Plantago major (seeds from wild origin), *P. lanceolata* (seeds from wild origin), and *P. media* (seeds from Botanical Garden, Meise, Belgium) were grown in a greenhouse (temperature 28°C). After 4 weeks, they were put in separate pots, and 5 weeks later they were transferred to a colder greenhouse (temperature 15°C).

Fifteen weeks after sowing, the plants were fully grown and placed outdoors in an open greenhouse (missing the lower half of the walls) in the Botanical Gardens of Utrecht University, Utrecht, The Netherlands. This site is considered an urban area. The distance to the nearest highway is approximately 800 m, and the distance to downtown Utrecht is approximately 2.5 km. The potted plants were placed close to each other to minimize environmental variation. The plants were sprayed with collected rainwater twice a day. Temperature and relative humidity were measured continuously.

Samples. Leaf samples (8 g, quadruplicates) were taken on the last day in the greenhouse (day 0) and on days 6, 13, and 20. Each sample consisted of one leaf (*P. major*) or three to four leaves (*P. lanceolata* and *P. media*) originating from the same amount of individual plants. Leaf fresh weight was determined, after which leaf wax was extracted and analyzed for PAHs. Finally, leaf dry weight was determined. Using plants not subjected to PAH analysis, the wax content, the specific leaf area (SLA, cm^2 leaf area/g dry wt) and the ratios of the projected surface areas (A_{proj}) to total surface areas (TSA) of the three species were determined ($f_{proj} = A_{proj}/TSA$). Wax content was determined on days 13 and 20 ($n = 1$), while SLA was measured on each sampling day ($n = 4$). The f_{proj} was determined for a different batch of plants ($n = 3$).

Leaf wax and leaf area. To determine the amount and composition of the leaf wax, leaves were immersed in 2×25 ml chloroform for, respectively, 30 s and 10 s to extract leaf wax. Previous experiments have shown that, in this time period, the leaf wax is completely extracted (data not shown). Extracted leaves were dried in an oven (80°C, 24 h) to determine foliar dry weight. Extracts were filtered with a glass microfiber filter (pore diameter 2.7 μm; Whatman, Clifton, NJ, USA) to remove particles and, after evaporation to dryness, wax weight was determined. The composition of the leaf wax (redissolved in diethylether) was determined by gas chromatography (GC) (HP 5890A, Hewlett-Packard, Amstelveen, The Netherlands) equipped with a flame ionization detector before and after methylation with diazomethane on a 25-m CP-SIL 5CB-column (i.d., 0.32 mm; film thickness, 0.12 mm; carrier gas, N_2 at 4.10^4 Pa inlet pressure). Injector and detector temperature was 280°C. Oven temperature was 250°C for 45 min, then was quickly raised to 300°C and kept at that temperature for 45 min. Peaks were identified by relative retention time compared to the internal standard 5α-cholestane.

Leaf areas were measured with a leaf area meter (LiCor

Lincoln L1-3100, LiCor, Lincoln, NE, USA). To determine f_{proj}, the projected surface area (A_{proj}) was divided by the total surface area of the leaves (=2 × measured leaf area). A_{proj} was determined by taking photographs of the plants from a vertical view, clipping out the projected plant, and measuring the area of the clippings (correcting for the scale of the photos).

Sample cleanup and PAH analysis

Leaves were not washed before extraction to make sure that particle-bound PAHs were not washed away. Extracts (made as described in the previous section) were evaporated to 10 ml under a gentle stream of nitrogen. One and a half grams of C_{18} was added, and the samples were further evaporated to dryness. Cleanup of the samples was performed by transferring the C_{18} to a 20-ml plastic syringe filled with a plug of quartz-wool and 1 g C_{18}, followed by elution with 18 ml of ACN. Samples were evaporated to 0.5 ml under nitrogen and injected in an HPLC-system with a Merck-Hitachi L-6200 Intelligent Pump (Merck, Darmstadt, Germany), which was supplied with a Chrompack ChromSpher PAH column (Chrompack, Middelburg, The Netherlands) connected to a Merck-Hitachi F-1050 Fluorescence Spectrometer (Merck). The column was eluted with ACN:millipore water 55:45 with a linear gradient to 65:35 in 6 min. A ratio of 65:35 was used for 2 min, followed by a 6-min linear gradient to 100% ACN. Then, 100% ACN was flushed for 7 min, after which the ACN:millipore water ratio was reset instantaneously to 55:45. The PAHs were identified by comparing the retention times of the sample peaks to those of the standards. Signal collection and data processing was performed on a computer with Chromcard 1.17 (Fisons Instruments, Loughborough, UK).

Recoveries of the PAHs were >90% except for B*ghi*P, which was 77 ± 9%. Procedural blanks (n = 4) were determined by extraction and cleanup with 50 ml chloroform.

RESULTS AND DISCUSSION

Plant waxes

The main components of the extractable cuticular waxes of *P. major* are linear alkanes ($C_{27}H_{56}$-$C_{33}H_{68}$) and triterpene acids (oleanolic and ursolic acid) [25]. The other two plants have a similar wax composition. The wax contents of the plants approximately doubled from day 13 to day 20. Nevertheless, the wax content relative to *P. major* remained fairly constant and differed not much from one (for *P. lanceolata*, the data were 0.82 and 0.86; for *P. media*, the data were 1.12 and 1.31 for days 13 and 20, respectively). The SLA of the plants was also similar and did not show a trend with time (144 ± 5, 123 ± 5, and 165 ± 12 cm²/g dry wt for *P. major*, *P. lanceolata*, and *P. media*, respectively). However, f_{proj} of *P. media* was significantly ($p < 0.01$) smaller than that of the other two species (0.22 ± 0.07, in contrast with 0.52 ± 0.03 for *P. major* and 0.48 ± 0.08 for *P. lanceolata*).

Time course of PAH concentrations and PAH profiles

PAHs were already present in the wax of the leaves from the greenhouse (day 0). The PAH concentrations in the plants (Fig. 2) increased by a factor of two to seven after they were transferred to the open greenhouse (with the exception of BghiP). This increase was likely caused by a higher supply of PAHs or smaller boundary layers around the leaves due to more wind outside. Concentrations on days 6 and 13 were similar, but on day 20, concentrations of most compounds

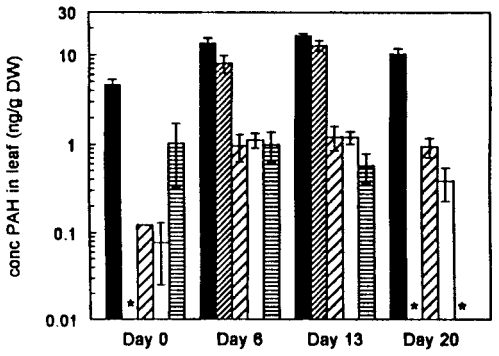

Fig. 2. The PAH concentrations (ng PAH/g dry weight of leaf) in leaf wax of *Plantago lanceolata* on the different sampling days. The PAHs are grouped according to their molecular weight (MW). Error bars = standard deviations of the four replicates; * = cases where the amount of PAHs in leaf wax was less than the amount in the procedural blanks plus two times the standard deviation. Solid bars represent MW 178; bars with closely spaced diagonal lines represent MW 202; bars with widely spaced diagonal lines represent MW 288; open bars represent MW 252; and bars with horizontal lines represent MW 276.

decreased considerably (Fig. 2, note the log scale). The reason for this decrease remains unclear. The only change from days 13 and 20 in the measured plant parameters was the twofold increase of the wax content, but this cannot explain a decrease in PAH concentration expressed per gram of leaf dry weight. Nevertheless, for the purposes of this study, namely the interpretation of differences between species, it is not of much relevance.

The profiles of the PAHs of *P. major* and *P. lanceolata* were very similar, while the profiles of *P. media* showed a slightly different pattern. This is mainly due to a consistently lower contribution of PHE (40–55% instead of 50–60%) and a higher contribution of higher MW PAHs to the total amount of PAHs taken up (data not shown). For example, B*k*F accounts for ±4% in *P. media* and for ±1% in the other two plants. The FLUO also made up a major proportion of the total concentration (25–40%). The rest of the PAHs were only present in small amounts (0.5–10%).

The PAH concentrations in Plantago *species*

To compare the concentrations of PAHs in the three *Plantago* species, the results of day 13 are plotted in Figure 3. On the other sampling days, the species differences were similar and therefore the results of these days are not shown. Concentrations are expressed per gram dry weight since this parameter was measured for all samples, in contrast to the leaf area and the wax content, which were only determined for a number of control plants. However, the differences in the wax content and in the SLA between the species are relatively small and plots based on these units result in similar figures, showing the same significant differences as in Figure 3.

The concentrations of PAHs with MW 178 and 202, which are largely present in the atmosphere as gases [14,15], were two to five times higher in *P. major* and *P. lanceolata* than in *P. media* (Fig. 3).

For the higher MW PAHs, which are almost completely bound to particles [14–16], significant differences between the species could not be proved statistically (Fig. 3). The absence of statistical significance could be caused by the limited sample size that could be cleaned up efficiently. Hence, the amount of high MW PAHs that was extracted is relatively low (down to approximately 1 ng), resulting in low precision. Neverthe-

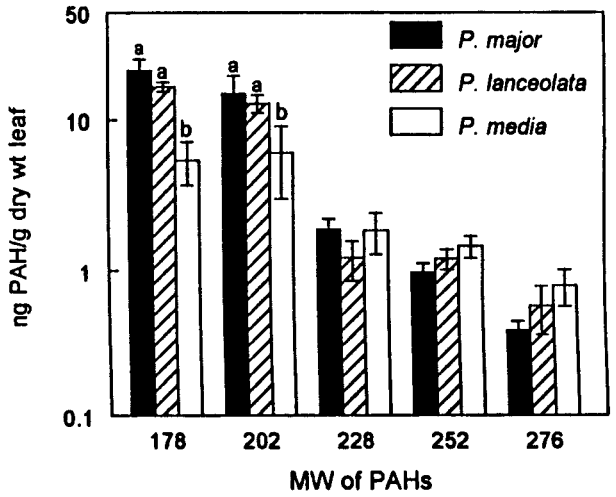

Fig. 3. The PAH concentrations (ng PAH/g dry weight of leaf) in leaf wax of the three *Plantago* species on day 13. The PAHs are grouped according to their molecular weight (MW). Differences between the average concentrations of the three species were tested with ANOVA (Prism 2.01, $p < 0.05$) and, when found significant, are so indicated in the figure with letters a and b.

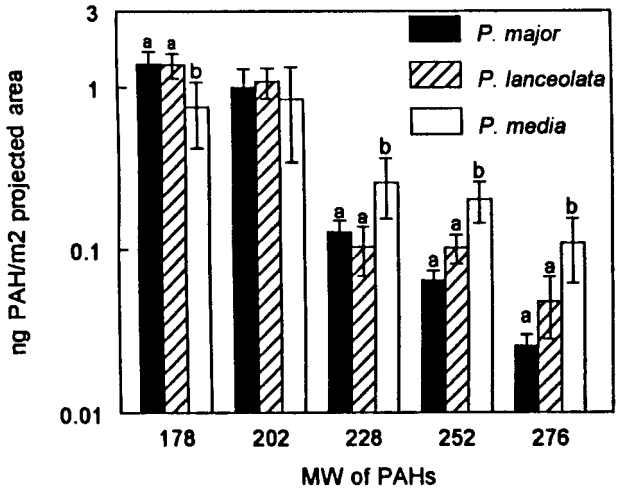

Fig. 4. The PAH concentrations per projected leaf surface area (ng PAH/m^2 A_{proj}) in the leaf wax of the three *Plantago* species on day 13. The PAHs are grouped according to their molecular weight (MW). Error bars represent standard deviations of the four replicates (taking into account the error in the determination of A_{proj}). Differences between the average concentrations of the three species were tested with ANOVA (Prism 2.01, $p < 0.05$) and, when found significant, are so indicated in the figure with letters a and b.

less, when looking at the trends, concentrations of PAHs with MW 252 and 276 in *P. media* are highest and those in *P. major* lowest. This was also the case for the individual PAHs with MW 252, namely B*a*P and B*k*F. The same trend was found on the other sampling days as well.

The differences in PAH concentrations can be explained by the differences in plant characteristics of the three *Plantago* species. These can be divided in differences in surface roughness, leaf hairs, and leaf overlap.

Surface roughness

Because *P. media* is a low growing plant with its leaves spreading close to the ground, the aerodynamic surface roughness of the canopy will be lower than that of the other two plants. This will result in less aerodynamic turbulence and therefore to a lower supply of compounds from the bulk air. However, this will only lead to lower uptake if the aerodynamic component of the atmospheric resistance is the rate-limiting step of the process.

For particle-bound PAHs, it is expected that the lower surface roughness of *P. media* will have a similar effect on the deposition. Since the pattern of high MW PAHs in Figure 3 points in the opposite direction, surface roughness cannot explain this finding and is probably overcompensated for by another factor, such as leaf hairs and leaf overlap.

Leaf hairs

Surface irregularities on the leaf, such as veins and hairs, can induce turbulence and hence decrease δ_{bl}. On the other hand, a dense mat of hairs is likely to increase δ_{bl} by up to the thickness of the hair mat [12]. The effect of hairs on δ_{bl} was demonstrated by Woolley [26], who showed that the wind speed 0.5 mm above a soybean leaf increased by 40% after the hairs had been removed. In contrast with the other two plants, *P. media* leaves are densely hairy and therefore the δ_{bl} will be increased. This may cause a lower uptake rate for the gaseous PAHs if the boundary resistance is the main atmospheric resistance.

It is not clear from this experiment whether the turbulent or the laminar component of the resistance determined gas transport to the leaf surface. Therefore, no conclusions can be drawn about the actual cause of the lower uptake of the low MW PAHs in *P. media.* Both the lower surface roughness and the higher density of leaf hairs of *P. media* are possible explanations for this phenomenon.

The trend of *P. media* having the highest concentrations of high MW PAHs (Fig. 3) can be explained by its hairy leaves. The PAHs are largely bound to particles <2 μm [15,27–29]. It is known that in the size range 1 to 5 μm, the deposition by impaction (inertial motion) is inefficient and the presence of fine hairs may be of major importance in intercepting particles [30]. For particles smaller than 1 μm, diffusion becomes the dominant means of transport to the leaf surface, and the nature of the surface is not so important as for larger particles. However, once the particles have been deposited on the leaf surface, the hairy leaves act as an efficient particle trap. Because of the thick boundary layer, wind eddies cannot penetrate down to blow the particles off the leaf surface [10]. Another explanation for the increased particle collection efficiency of hairy leaves is that leaf hairs may cushion the impact and therefore reduce the bounce-off of particles [17].

Leaf overlap

The lower concentrations of gaseous PAHs in *P. media* may also be caused by the higher overlap of leaves of this plant. Because of the overlap, leaves may be less accessible for exchange of air, thus preventing the uptake of gaseous SOCs in the covered parts. This factor is not taken into account when expressing PAH concentrations per gram dry weight, per gram leaf wax, or per square meter of surface area. Therefore, we normalized the PAH concentrations (day 13) on the projected surface area (A_{proj}), which could be calculated for each sample from the measured total surface area and from f_{proj}, which was determined for the control plants. The PAH concentrations based on A_{proj} are plotted in Figure 4. Concentrations of PAHs with MW 178 were again lowest in *P. media*, while the concentrations of FLUO (MW 202) were similar in

the three plants. This suggests that A_{proj} may at least partially explain the observed differences for the gaseous compounds. After normalization to A_{proj}, the high MW PAHs showed significantly higher concentrations in *P. media* than in the other two plants (Fig. 4). This means that, per unit accessible surface area, the concentrations of particle-bound PAHs in *P. media* are significantly higher than those of the other two plants.

The projected surface area (A_{proj}) represents the surface area that is not covered by other leaves. Since in this approximation the vertical dimension is lost, the use of A_{proj} is only acceptable when using plants with highly similar morphology. Although the plants used in the present study belong to the same genus, they differ in height, which complicates the interpretation. The greater height of *P. major* and *P. lanceolata* may increase the interception of PAHs. However, in spite of the possible extra deposition caused by their height, high MW PAH concentrations in *P. major* and *P. lanceolata* expressed per projected area (A_{proj}) were lower than those of *P. media,* which emphasizes the effective particle collection of hairy leaves.

Since A_{proj} probably reflects a minimal accessible leaf area, only a rough idea of the influence of the plant architecture on the deposition of PAHs can be obtained in this way. Besides, since gases are able to diffuse much faster than particles due to their higher diffusion coefficients, the use of A_{proj} as a measure of accessiblity will likely be of more relevance for particle-bound compounds than for gaseous compounds.

Kinetics

The explanations given in the previous sections consider the atmospheric resistance. The atmospheric resistance can only determine the measured concentrations if an equilibrium between air and plant has not been reached. This is because, in an equilibrium situation, the concentrations in the plant are independent of the aerodynamic turbulence and boundary layer thickness. Because of the lack of data for the kinetic behavior of PAHs in *Plantago,* the time needed to reach equilibrium was estimated from data measured for other plants. Since the elimination rate decreases with increasing K_{oa} [31], only studies in which compounds were used with log K_{oa} values comparable to those of PHE, ANT, and FLUO (which have a log K_{oa} of, respectively, 7.4 [32], 7.8 [32], and 8.6, calculated from [33]) were chosen. From the elimination rate constants reported in these studies, the time needed to achieve 95% of the equilibrium concentration ($t_{0.95}$) was calculated with the first-order one-compartment kinetic bioconcentration model. In studies in which plants were exposed in chambers containing a ventilator, i.e., under turbulent conditions (thin boundary layers), $t_{0.95}$ values ranged from 3 d for anthracene in grass [32] to 107 d for PCB 18 in spruce needles [34]. Under non-turbulent lab conditions, a value of 31 d for mirex [35] was found, whereas in a field study, concentrations of chlorinated organic compounds (PCBs, DDT, etc.) in spruce needles were still increasing after 5 years [36]. These results indicate that it is not likely that equilibrium will be reached within 3 weeks of exposure in an open greenhouse.

CONCLUSION

The densely hairy, partially overlapping leaves of the low growing *P. media* contain less low MW PAHs and more high MW PAHs than the almost glabrous *P. major* and sparsely hairy *P. lanceolata* leaves, which are relatively free-standing in the air. The measured trends are consistent and indicate that leaf hairs and plant architecture can affect deposition rates of SOCs in different ways for gases and particles. These results may be important for predictive models, in particular with regard to the concentration of SOCs in food crops. Food crops are only exposed to contaminated air for a relatively short time period and an equilibrium between air and plant will probably not be achieved. Neglecting the effects of plant architecture may result in overestimation of concentrations of gaseous SOCs in the plant. On the other hand, deposition of particle-bound SOCs may be underestimated for leaves with a dense layer of hairs. However, differences in foliar concentrations of SOCs between plant species in these experiments were not large, usually less than a factor of five. In future studies, plants with a wider range of characteristics need to be studied to investigate maximum possible concentration differences in food crops. Only then will it be clear which plant properties should be included as parameters in predictive models.

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