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# Modeling the Potential Influence of Particle Deposition on the Accumulation of Organic Contaminants by Submerged Aquatic Vegetation

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Submerged aquatic vegetation can act as both a mitigating factor (e.g., reducing downstream impacts of pesticides following runoff/spray drift) and mobilizing factor (e.g., remobilization of contaminants from sediments) influencing the fate and distribution of organic contaminants in the environment. Consequently, there has been wide scientific and regulatory interest in assessing the role of these plants in different contamination scenarios. Mechanistic models describing the environmental fate of contaminants in submerged aquatic vegetation are useful tools for interpreting laboratory and field measurements in addition to providing valuable information to risk assessors. In this study, we developed a fugacity-based model to investigate the influence of particle deposition to plant surfaces on the fate and distribution of two substances in small ponds. The main motivation for conducting this study was to address the fact that the potential contribution of this process is not typically considered by many types of models describing contaminant dynamics in submerged aquatic vegetation. For the hydrophobic substance included in this evaluation (lambda-cyhalothrin), model performance was greatly improved by including this process. The model was also applied in a generic context to compare the importance of particle deposition versus direct water uptake as a function of chemical properties (log  $K_{OW}$ ) and concentration of suspended solids in the water column. The generic application demonstrated that contaminant mass transfer is dominated by particle deposition for chemicals with log  $K_{OW}$  greater than approximately 5.5–6 across a wide range of suspended solid concentrations and can be important even for low log  $K_{OW}$  substances in some circumstances. Further empirical and modeling studies are recommended to explore this process more comprehensively.

## Introduction

The role of aquatic vegetation in influencing the fate and distribution of organic contaminants in the environment has received considerable attention, particularly in the context of the development of natural buffer zones (e.g., ditches, wetlands) designed to mitigate potential downstream impacts of pesticides in receiving waters following runoff and spray-drift from agricultural areas. The importance of aquatic macrophytes and the associated phytoremediation processes that these plant communities support have been established in numerous laboratory and field studies and further research is ongoing (1–8). Conversely, there is also an interest in the potential for rooted aquatic vegetation to remobilize hydrophobic pollutants directly and indirectly from contaminated sediments (9–14). Mechanistic models simulating the distribution of organic contaminants in natural systems are useful tools to investigate these situations and explore the potential of vegetation to influence the fate of chemicals in both the abiotic and biotic environment.

Gobas et al. (15) investigated the bioconcentration of hydrophobic contaminants in submerged aquatic macrophytes and used the results of a laboratory study to develop a kinetic model that estimated uptake and elimination rate constants as a function of the octanol–water partition coefficient ( $K_{OW}$ ). No soil or sediment was present in the test system and the experiments were conducted using carbon-filtered water since the purpose of the experiments was to investigate uptake of contaminants exclusively from the dissolved phase of the water column. Other researchers have used field measurements to develop empirical relationships between chemical concentrations in sediments, roots, and shoots for a limited number of contaminants such as PCBs (16) or conducted laboratory studies to determine sorption isotherms (17). Because of their broader applicability however, generic rate constant expressions are highly desirable, particularly for the development of models aiming to screen large numbers of chemicals. For example, the rate constant expressions proposed by Gobas et al. (15) were incorporated into the U.S. EPA model TRIM.FaTE (18, 19).

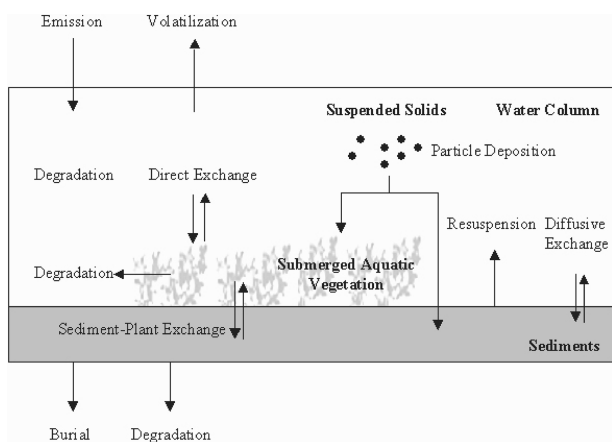
Submerged aquatic vegetation has a well-established impact on water flow and sedimentation dynamics in waters where significant biomass of plants is present. For example, flow velocities within plant stands are reduced significantly which can lead to increased deposition and retention of fine particulate matter in sediments associated with the stands (e.g., refs 20, 21). Another phenomenon relevant to the fate and distribution of organic contaminants is the deposition to and retention of fine particulate matter by plant surfaces themselves (22–24). Retention of particles by submerged aquatic plants in natural systems is greatly facilitated by the presence of a “sticky” periphyton layer (biofilm) typically coating the surface (24). Since hydrophobic contaminants have a strong affinity for particulate matter in the water column, deposition of particulate matter to submerged vegetation may represent a significant transport process not typically considered in many fate and exposure models. Therefore, the purpose of this study was to develop and evaluate a mechanistically based bioaccumulation model for submerged aquatic vegetation with the main objective of investigating the hypothesis that particle deposition to submerged aquatic vegetation is an important process for certain organic contaminants. A secondary objective of this investigation was to identify the conditions and physical–chemical properties that favor the mass transfer of con-

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**FIGURE 1.** Conceptual representation of the sediment–water–submerged aquatic vegetation system and environmental fate processes included in the model calculations.

taminants via particle deposition over uptake directly from the water column.

## Materials and Methods

The potential influence of particle deposition on the accumulation of organic contaminants by submerged aquatic vegetation was explored by incorporating a representation of this transport process into a multimedia environmental fate model and then comparing model output (with and without this process) to observations. The details of the model development and application are presented in the following sections.

**Model Development.** The model was based on the fugacity approach (25) and is representative of small water bodies (e.g., ponds, ditches) containing submerged aquatic vegetation. It is a dynamic nonsteady state model that is appropriate for intermittent exposures, the typical scenario in applications when simulating the input of contaminants to receiving waters following pesticide runoff or spray drift. According to the fugacity approach,  $D$ -values ( $\text{mol Pa}^{-1} \text{h}^{-1}$ ) can be used to estimate diffusive and nondiffusive exchange of chemicals between environmental media (e.g., water column, sediments). The flux of chemicals associated with these transport processes ( $\text{mol h}^{-1}$ ) are derived from the  $D$ -values multiplied by the fugacity (Pa) of the chemical in each compartment. The fugacity ( $f$ ) of the chemical in each compartment is estimated as follows:

$$f = \frac{C}{Z} \quad (1)$$

where  $C$  is the concentration of the contaminant ( $\text{mol m}^{-3}$ ) and  $Z$  is the fugacity capacity of the compartment ( $\text{mol m}^{-3} \text{Pa}^{-1}$ ). In this case, the physical environment of the model included the water column, sediments, and vegetation. The water compartment comprised three phases (water, suspended solids, dissolved organic carbon) while the sediment comprised two phases (solids, pore water) and the vegetation was represented as a single phase. The definitions of the  $Z$ -values for each compartment/ phase are shown in Table S1 (Supporting Information).

A conceptual representation of the system and fate processes included in the model is presented in Figure 1 and a complete description of the model formulation and definitions of the  $D$ -values are provided in the Supporting Information. All processes shown in Figure 1 and described in the Supporting Information were included in the model simulations. However, since the focus of this investigation was the accumulation of organic contaminants by submerged

aquatic vegetation, only these process descriptions are presented here.

**Submerged Aquatic Vegetation–Water Column Exchange.** Submerged aquatic vegetation–water column exchange can occur through diffusive and nondiffusive processes. Diffusive exchange rates were based on Gobas et al. (15) who reported the following uptake ( $k_1$ ,  $\text{d}^{-1}$ ) and elimination ( $k_2$ ,  $\text{d}^{-1}$ ) rate constant expressions as a function of the octanol–water partition coefficient ( $K_{OW}$ ):

$$\frac{1}{k_1} = 0.002 + \frac{500}{K_{OW}} \quad (2)$$

$$\frac{1}{k_2} = 1.58 + 0.000015 K_{OW} \quad (3)$$

The  $D$ -value for exchange directly between the water column and vegetation was therefore defined as follows:

$$D_{WM} = k_U V_M Z_W \quad (4)$$

where  $k_U$  is the uptake rate constant per hour (i.e.,  $k_1/24$ ),  $V_M$  is the volume of submerged aquatic vegetation ( $\text{m}^3$ ), and  $Z_W$  is the fugacity capacity of water. The  $D$ -value for exchange from the macrophyte back to the water column was defined as

$$D_{MW} = k_R V_M Z_M \quad (5)$$

where  $k_R$  is the vegetation elimination rate constant per hour (i.e.,  $k_2/24$ ).

The representation of particle deposition to submerged aquatic vegetation was based on the analogous process of dry particle deposition to terrestrial plant surfaces (26). The transport  $D$ -value characterizing this nondiffusive process was therefore defined as follows:

$$D_{PM} = U_{DP} A_M Z_P \quad (6)$$

where  $U_{DP}$  is the mass transfer coefficient for particle deposition ( $\text{m h}^{-1}$ ),  $A_M$  is the effective surface area ( $\text{m}^2$ ) of the submerged vegetation compartment, and  $Z_P$  is the fugacity capacity of the particulate matter.  $U_{DP}$  was estimated as follows:

$$U_{DP} = \gamma_P \frac{C_{SS}}{\rho_P} \quad (7)$$

where  $\gamma_P$  is the mean particle settling velocity ( $\text{m h}^{-1}$ ),  $C_{SS}$  is the concentration of suspended solids (SS) in the water column ( $\text{g SS m}^{-3} \text{water}$ ), and  $\rho_P$  is the density of these particles ( $\text{g m}^{-3}$ ). As a first approximation (in the absence of site-specific observations),  $\gamma_P$  was assumed to be in the range of  $0.02$ – $0.08 \text{ m h}^{-1}$  ( $0.5$ – $2 \text{ m d}^{-1}$ ), the values suggested by Mackay (24) and Schwarzenbach et al. (27). A more detailed discussion of particle settling velocity is provided in the Supporting Information. Since Plunkte and Kersowoski (23) reported similar rates of particle deposition ( $\text{g m}^{-2} \text{d}^{-1}$ ) to plant surfaces and sediments in their experimental system, the same  $\gamma_P$  was applied to the  $D$ -value for particle deposition to sediments.

The effective surface area of the vegetation was estimated as

$$A_M = (\text{LAI}) A_W \quad (8)$$

where LAI is the leaf area index ( $\text{m}^2 \text{leaf m}^{-2} \text{water/sediment}$ ). LAI was estimated from the mass of leaves ( $M_L$ , dry g) and specific leaf area (SLA,  $\text{m}^2 \text{g}^{-1} \text{dry leaf}$ ) as shown below.

$$\text{LAI} = M_L \text{SLA} \quad (9)$$

The main advantage of estimating surface area from plant biomass in this way is that consistency between vegetation surface area ( $A_M$ ) and volume ( $V_M$ ) is assured in addition to

the fact that LAI scales to the reported biomass (i.e., g plant m<sup>-2</sup> water) in the test system. The LAIs calculated through this approach (see Table S3, Supporting Information) were broadly in agreement with published estimates (28) for LAI for a variety of submerged aquatic vegetation (LAI = 2–12).

Lastly,  $Z_p$ , the fugacity capacity of the suspended solids, was defined as

$$Z_p = Z_{w/f_{OC-P}} \alpha_{OC} K_{OW} \rho_p \quad (10)$$

where  $f_{OC-P}$  is the organic carbon content of the suspended solids,  $\alpha_{OC}$  is the proportionality constant (L kg<sup>-1</sup>) relating the sorptive capacity of organic carbon to octanol, assumed to be 0.41 (29), and  $\rho_p$  is expressed in units of kg L<sup>-1</sup>. Based on this definition, the uptake of contaminants via particle deposition is thus a function of the sorptive capacity of the suspended solids ( $Z_p$ ,  $f_{OC-P}$ ), plant characteristics (e.g., SLA), water column characteristics ( $C_{ss}$ ,  $\gamma_p$ ), and the  $K_{OW}$  of the contaminant of interest. Note that while the simple relationship employed to derive  $K_{OC}$  from  $K_{OW}$  was recently shown to yield reasonable results for nonpolar compounds in general (30) as well as for both test compounds considered here (see Supporting Information), the use of polyparameter linear free energy relationships (pp-LFERs) to predict  $K_{OC}$ s (and  $K_{DOC}$ s) for polar compounds with strong specific molecular interactions (e.g., hydrogen bonding) is recommended whenever possible.

**Model Application.** The model was applied to simulate the fate of lambda-cyhalothrin in two scenarios (31) and azinphos-methyl in one scenario (32, 33). In both studies, the test systems were dosed with a single application of pesticide (see Supporting Information) and the concentration of the substance was measured over time in samples of the water column, sediments, and aquatic vegetation. These case studies were selected for several reasons. First, *Myriophyllum spicatum*, the same type of macrophyte used by Gobas et al. (15), was the dominant plant type in the study using lambda-cyhalothrin. Lambda-cyhalothrin is also extremely hydrophobic (34) and therefore partitions extensively to organic matter in the water column. This tendency makes it an ideal substance to explore the potential importance of contaminant mass transfer to submerged aquatic vegetation via particle deposition. In addition, the water column in the test enclosures had a high concentration of suspended solids (~16 mg L<sup>-1</sup>) with a measured organic matter content greater than 70% (31). In addition, measurements from experiments conducted in enclosures with high and low density of biomass (145 g dry plant m<sup>-2</sup> vs 43 g dry plant m<sup>-2</sup>) were available. The case study with azinphos-methyl was selected as a contrasting example. This substance is more water-soluble (35) and has a much lower tendency to partition to organic matter (log  $K_{OW}$  = 2.75). The water in the test system contained a lower concentration of suspended solids (2.7 mg L<sup>-1</sup>) and also a lower plant biomass density (32, 33). Although the vegetation in this study was dominated by the macroalgae species *Chara globularis* and *Chara vulgaris*, this difference is also advantageous as it presents the opportunity to assess the performance of the model for a different type of submerged aquatic vegetation.

**Model Parameterization.** Input parameters describing the physical environment (e.g., water surface area, depth, sediment organic carbon content), submerged aquatic vegetation (e.g., biomass density, SLA, organic carbon content), and physical–chemical properties (e.g., partitioning, degradation half-lives) were required to conduct the model simulations. The model parametrization process was greatly facilitated by the fact that many of the required data were reported for both case studies (31–33). Physical–chemical properties for both substances are presented in Table S2 while parameters describing the physical environment are

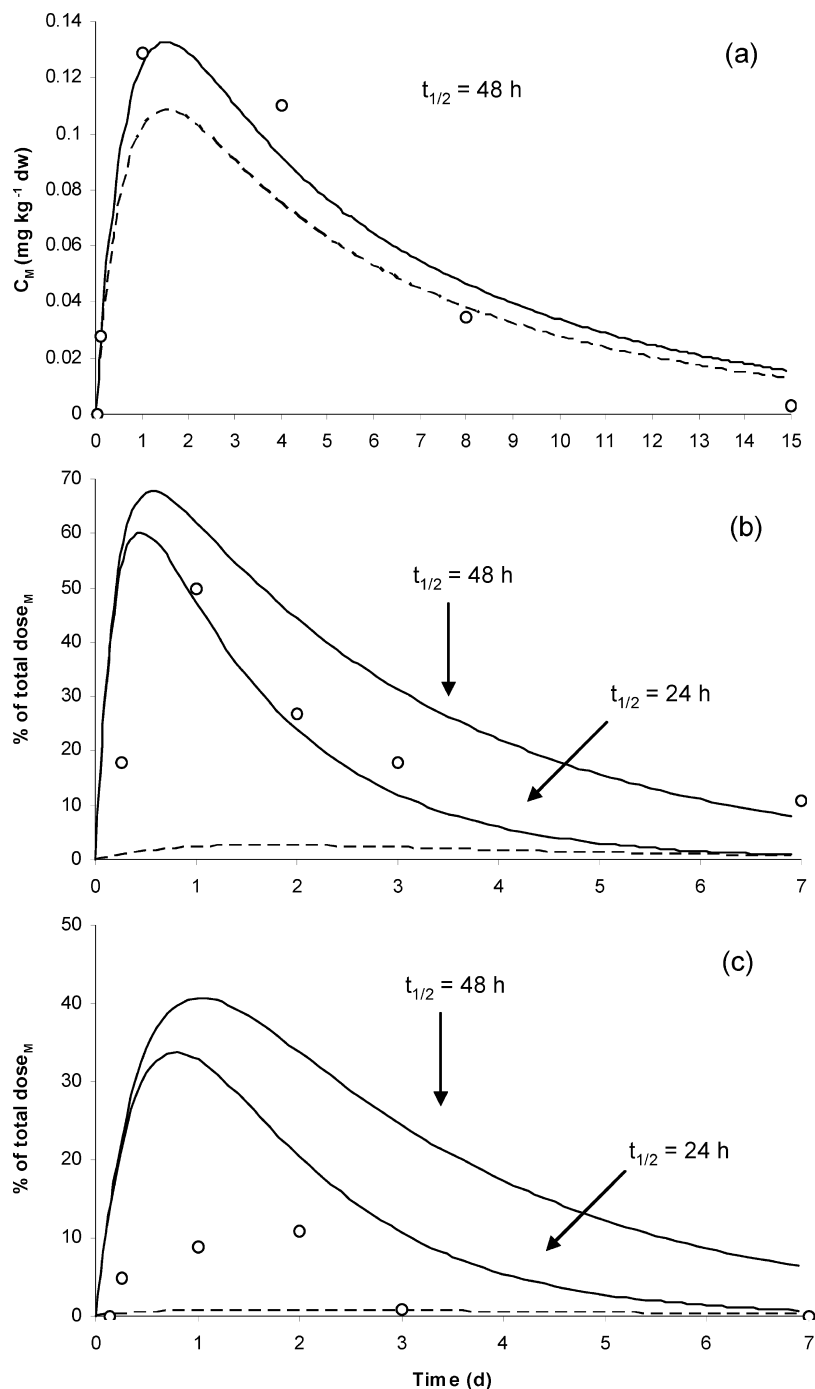
presented in Table S3 along with a brief description of the model parametrization process (see Supporting Information).

The most challenging aspect of the parametrization process was assigning degradation half-lives for these substances in the various compartments. Both substances are susceptible to base-mediated hydrolysis and therefore the degradation half-lives are strongly influenced by ambient pH and to a lesser extent, temperature. For example, the reported degradation half-lives due to aqueous hydrolysis for azinphos-methyl (Guthion) at 25 °C were approximately 28 days, 2.5 days, and 2 days at pH 8.6, 9.6, and 10.7 respectively (36). Lambda-cyhalothrin, resistant to degradation via aqueous hydrolysis at acidic and neutral pHs, reportedly degrades with a half-life of approximately 7–10 days at pH 9 (31, 37). The reported pH for the enclosure exposed to azinphos-methyl varied from 7.4 to 9.1 (32) while the reported pH at noon for the enclosures exposed to lambda-cyhalothrin simulated in this study was 7.6 ± 0.2 although measurement bias may have been a factor, as discussed in ref 31. In previous studies in the same mesocosms, measured pH typically ranged up to 9.5–10 during the day. This high pH is consistent with the expected influence of photosynthetic activity, which has been shown to elevate pH in natural waters above 9 (38, 39). Based on these considerations, degradation half-lives in the water column were assigned values of 3 and 7 days for azinphos-methyl and lambda-cyhalothrin, respectively, based on the assumption that pH exceeded 9 for substantial portions of the exposure period. Half-lives in sediment were assumed to be longer (10 days) due to the lower reported pH in this phase (e.g., ref 31). Hand et al. (2) studied the influence of submerged aquatic vegetation on the fate of radiolabeled lambda-cyhalothrin in laboratory and indoor microcosm studies. They reported that ester hydrolysis of lambda-cyhalothrin proceeded at a much faster rate in a plant–water system ( $t_{1/2}$  < 1 d) than would be expected from base-mediated hydrolysis alone. However, the precise contribution of plant-mediated and base-mediated hydrolysis was indeterminable. Based on these considerations, the degradation half-life for both lambda-cyhalothrin and azinphos-methyl for the vegetation compartment was assumed to be elevated in comparison to the water column. The default value for both chemicals was 2 days and model simulations were also conducted with lower values (0.5, 1 day).

## Results and Discussion

The predicted and observed fate of lambda-cyhalothrin and azinphos-methyl in submerged aquatic vegetation and sediments and water are shown in Figures 2 and 3, respectively. As in the original publications, predictions and observations for lambda-cyhalothrin are expressed as a % of total dose and for azinphos-methyl are expressed as concentrations. The predicted values in submerged aquatic vegetation with particle deposition to vegetation excluded from the model calculations are also shown in Figure 2.

**Submerged Aquatic Vegetation.** Overall, the model (including particle deposition) generated predicted values that are in reasonable agreement with the observations for submerged aquatic vegetation, particularly for the azinphos-methyl and high-density lambda-cyhalothrin scenarios (Figure 2). The overestimation of the observed data for the low-density lambda-cyhalothrin scenario may partly be related to the fact that submerged aquatic vegetation was pruned in this enclosure. If leafy material were preferentially removed, the assumptions used to calculate leaf area index (LAI) from total biomass may have overestimated this value and hence the transport  $D$ -value for particle deposition to vegetation. Model performance in the vegetation compartment in both lambda-cyhalothrin scenarios is improved when the degradation half-life in this compartment is 24 h as opposed to



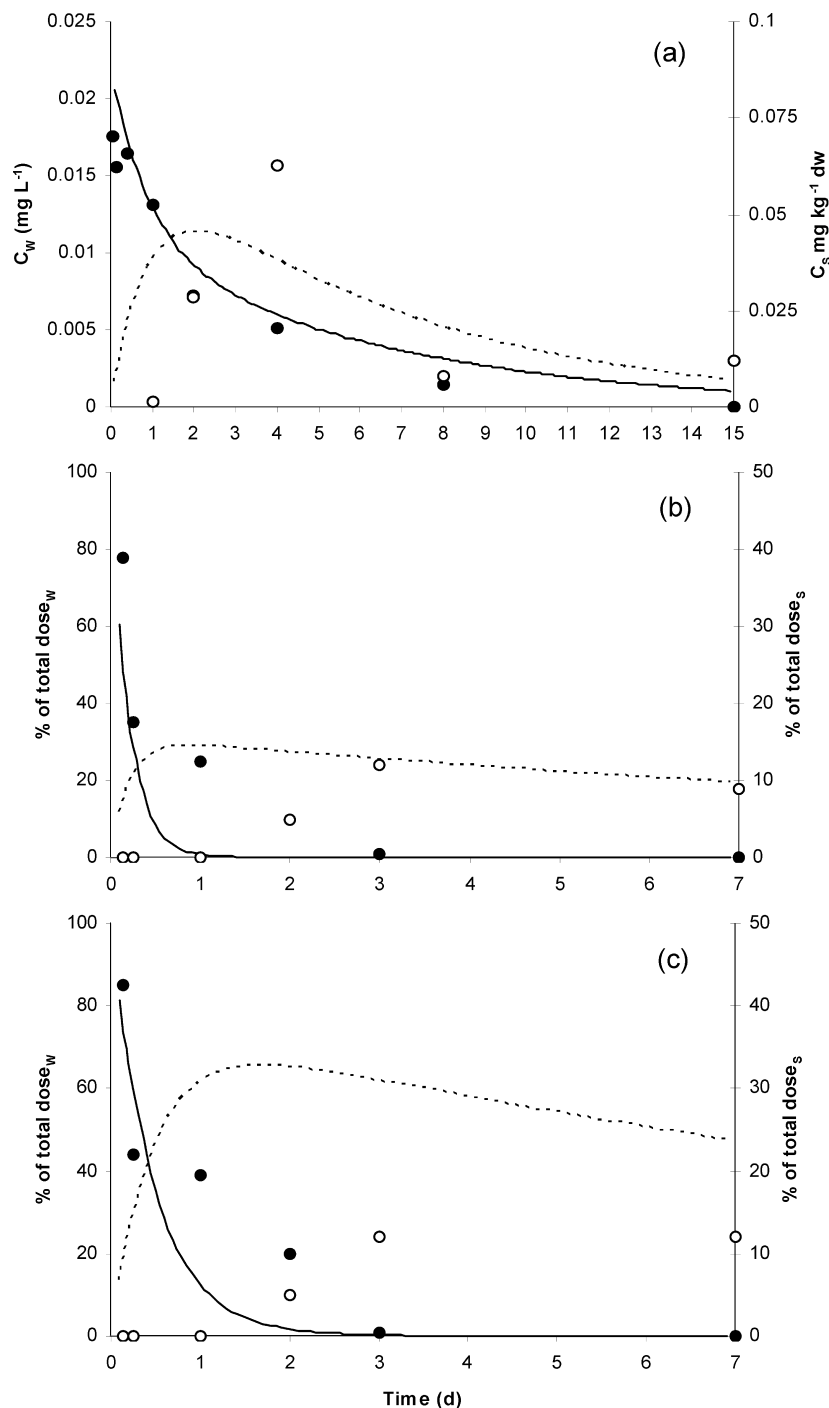
**FIGURE 2.** Predicted (lines) vs observed (circles) values of (a) azinphos-methyl, (b) lambda-cyhalothrin (high density), and (c) lambda-cyhalothrin (low density) in submerged aquatic vegetation including (solid lines) and excluding (dashed line) particle deposition. See refs 31 and 32 for observed values.

48 h. Further improvements for all scenarios could potentially be achieved by calibrating estimated parameter values such as the particle settling velocity ( $\gamma_p$ ), LAI, and growth rate constant ( $k_G$ ) in addition to degradation half-life but such an effort was not undertaken due to the limited number of measurements and lack of information on the uncertainty of these observations. Instead, model predictions for submerged aquatic vegetation based on the high-density lambda-cyhalothrin scenario were generated across a range of values for particle settling velocity and degradation half-life. These two parameters were selected because of the difficulty in assigning values based on reported conditions in the test systems (as opposed to others such as the concentration of suspended solids,  $C_{SS}$ ). These additional results are presented

in the Supporting Information (Figure S2) and demonstrate that the predicted results in submerged aquatic vegetation are relatively constrained across the range of the two input parameters considered. The results shown in Figure S2 also indicate that model performance is superior for all settling velocities assumed when the degradation half-life of lambda-cyhalothrin in the vegetation compartment is assumed to be 24 h.

An additional elimination pathway to represent particle sliding (loss of particles from leaf surface) could also be introduced to the model since this process was briefly discussed, but not quantified, by Plunkte and Kozerski (23). However, the “sticky” periphyton layer coating the surface of the plants may greatly limit this process. Preliminary model



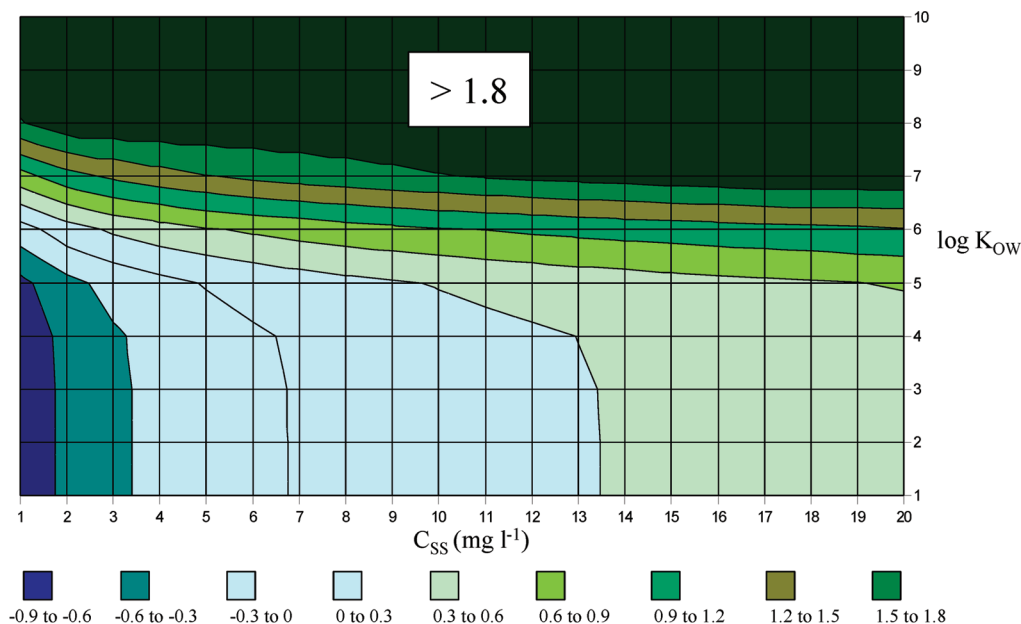


**FIGURE 3.** Predicted (lines) vs observed (circles) values of (a) azinphos-methyl, (b) lambda-cyhalothrin (high density), and (c) lambda-cyhalothrin (low density) in the water column (solid lines, closed circles) and sediments (dashed line, open circles) including particle deposition to submerged aquatic vegetation. See refs 31 and 32 for observed values.

simulations including a  $D$ -value representing this process (similar to resuspension from sediments) also indicated that degradation dominates the total elimination rate in the vegetation and particle sliding has little influence on predictions. Thus, for these substances, further information about degradation half-life in plants from laboratory or field studies would be highly valuable to better constrain this parameter value. However, transfer of contaminants back into the water column via particle sliding may be an important consideration for recalcitrant chemicals for which degradation is negligible.

The most important consideration in the context of this study is the comparison between model results including and excluding particle deposition as a contaminant transport

process. In the azinphos-methyl simulations, the predicted fate in the submerged aquatic vegetation was similar with and without particle deposition in stark contrast to both lambda-cyhalothrin simulations where the observed macrophyte concentrations could not be accounted for if only direct uptake ( $D_{WM}$ ) from the water column was considered. The model results can be explained by comparing the magnitude of the  $D$ -values for each process. In the azinphos-methyl scenario,  $D_{WM}$  is approximately three times greater than  $D_{PM}$ , the  $D$ -value for particle deposition, and therefore contaminant mass transfer to the vegetation compartment is dominated by diffusive exchange. Furthermore, the direct uptake process ( $D_{WM}$ ) can kinetically "compete" for contaminants in the water column against  $D_{PD}$  (deposition to



**FIGURE 4.** Contour plot of the ratio of  $D$ -values for contaminant transfer from water column to submerged aquatic vegetation via particle deposition and direct water uptake ( $D_{PM}:D_{WM}$ ) on  $\log_{10}$  basis as a function of  $\log K_{OW}$  and the concentration of suspended solids ( $C_{SS}$ ,  $\text{mg L}^{-1}$ ). Note that  $D_{PM} = D_{WM}$  when the ratio on a  $\log_{10}$  scale is zero.

the sediments, equivalent to  $D_{PM}/LAI$ ). In contrast,  $D_{PM}$  is nearly 2 orders of magnitude greater than  $D_{WM}$  in the lambda-cyhalothrin scenarios due to the greater fugacity capacity of the suspended solids ( $Z_p$ ) and also to some extent, the higher concentration of suspended solids ( $C_{SS}$ ) in the water column. Thus in the absence of particle deposition ( $D_{PM}$ ), contaminant mass transfer directly from the water column to vegetation ( $D_{WM}$ ) is not competitive with  $D_{PD}$  and the contaminant accumulates in the sediments instead (results not shown). Potential bias in the uptake rate constant expressions proposed by Gobas et al. (15) is also possible. However, to obtain predictions in good agreement with observations,  $k_U$  (see eqs 2 and 4) must be increased by nearly 2 orders of magnitude (i.e.,  $D_{WM}$  made equivalent to  $D_{PM}$ ) in these simulations. Given that Gobas et al. (15) derived the rate constant expressions including substances with  $\log K_{OW}$  values approaching that of lambda-cyhalothrin, the magnitude of the bias implied by these simulations seems unlikely.

**Sediments and Water Column.** The model results in the sediments and water column (Figure 3) show a reasonable agreement between predictions and observations for this type of modeling exercise (i.e., within a factor of 5 or less). One important discrepancy is the poor representation of the uptake of contaminants into the sediments over the first 1–2 days of the experiment. In all three experiments, the contaminant mass transfer into sediments appears to be retarded during the initial exposure period, resulting in a more sigmoid-shaped curve. One possible explanation for the observations is that the contaminant only gradually became well-mixed throughout the water column. In the model, the water column is assumed to be instantaneously well-mixed and it is also assumed that all phases within a compartment are at equilibrium. As such, the fastest rate of deposition into the sediments occurs immediately after dosing (with suspended solids at their highest fugacity). The discrepancy is fundamentally one of model formulation and the assumptions inherent to this mathematical representation of the real system. A more complex representation of the physical environment and/or sediment–water exchange may yield improved model performance but is beyond the scope of this study.

The agreement between predictions and observations in the water column for azinphos-methyl is exceptional in comparison to both lambda-cyhalothrin scenarios, in which the contaminant dissipates from the water column more rapidly than observations would suggest (Figure 3). In the azinphos-methyl scenario, the  $D$ -values indicate that dissipation of contaminant from the water column is strongly dominated by degradation rather than deposition to sediments and vegetation (due to low  $C_{SS}$  and low plant biomass). The excellent agreement between observations and predictions suggests that the assumed degradation half-life in the water column was reasonable. The magnitude of the  $D$ -values in the lambda-cyhalothrin scenarios indicates that particle deposition rather than degradation strongly dominates the dissipation of this contaminant from the water column. This result holds even if the degradation half-life of lambda-cyhalothrin in the water column is changed from 168 to 48 h. Therefore, the discrepancy between predictions and observations in the water column for lambda-cyhalothrin may also be related to the assumption of an instantaneously well-mixed water compartment. This is because the assumption of a well-mixed water compartment will lead to an overestimation of the mass of contaminant transferred to sediments and vegetation via particle deposition until complete mixing is approached. It is also important to note that the contribution of measurement uncertainty to the discrepancy between predictions and observations in the water column, sediments, and vegetation cannot be assessed.

**Generic Comparison of Particle Deposition and Direct Uptake.** The results presented in Figure 2 can be more clearly understood by comparing the magnitude of the  $D$ -values for direct uptake from the water column ( $D_{WM}$ ) and particle deposition ( $D_{PM}$ ) as a function of  $K_{OW}$  and the other properties that influence these processes. To facilitate such a comparison, the ratio of  $D_{PM}:D_{WM}$  (on a  $\log_{10}$  basis) as a function of  $\log K_{OW}$  (1–10) and  $C_{SS}$  (1–20  $\text{mg L}^{-1}$ ) using the high-density *Myriophyllum spicatum* parameter set is shown in Figure 4. A  $\log_{10} D_{PM}:D_{WM}$  ratio of zero represents the situation where  $D_{PM} = D_{WM}$  while positive values indicate that  $D_{PM} > D_{WM}$  and negative values indicate that  $D_{PM} < D_{WM}$ . This comparison indicates that direct uptake from the water column will only dominate in systems with low concentra-

tions of suspended solids (e.g.,  $C_{SS} < 2 \text{ mg L}^{-1}$ ) for chemicals with a  $\log K_{OW} < 5.5$ . As the concentration of suspended solids increases, the magnitude of  $D_{PM}$  increases in direct proportion and mass transfer of substances with lower  $K_{OW}$  become dominated by this transport process. It is important to realize that other parameters (e.g.,  $f_{OC-P}$ , SLA,  $\gamma_P$ ) have an influence on the exact location of the transition boundaries. For example, as particle settling velocity ( $\gamma_P$ ) decreases, the  $D_{PM}$  decreases proportionally and the boundary where  $D_{PM} = D_{WM}$  shifts to the right. The same is true for  $f_{OC-P}$  and SLA, which demonstrates that both plant morphology and particle quality (i.e., organic matter content) influence the fate of contaminants in submerged aquatic vegetation in addition to particle quantity ( $C_{SS}$ ) and physical-chemical properties. Note that since both  $D_{WM}$  (based on  $V_M$ ) and  $D_{PM}$  (based on  $A_M$ ) are calculated from plant biomass density, the ratio of  $D_{PM}:D_{WM}$  is independent of this parameter and the results of the analysis shown in Figure 4 can be considered universal, given the same values for other inputs. However, although  $D_{WM}$  is calculated using plant volume ( $V_M$ ), in reality, direct exchange of contaminants with the water column is ultimately a function of surface area as well and the uptake and elimination rate constants ( $k_1$  and  $k_2$ ) are related to the specific leaf morphology of the plant species used in the experiments. As such, the results presented in Figure 4 are more illustrative than definitive but still yield useful insights into conditions that result in a higher contaminant flux via particle deposition compared to direct exchange with the water column.

**Future Considerations.** Based on the modeling exercise conducted in this study, it is reasonable to conclude that the flux of contaminants to submerged aquatic vegetation via particle deposition is substantial for many organic chemicals. The generic assessment (Figure 4) indicates that under typical natural conditions (e.g.,  $C_{SS} = 5\text{--}10 \text{ mg L}^{-1}$ ), contaminant mass transfer via particle deposition will dominate for chemicals with a  $\log K_{OW}$  greater than approximately 5.5–6 and can contribute significantly in many other circumstances even for chemicals with relatively low  $\log K_{OW}$  (e.g., when  $C_{SS} > 15 \text{ mg L}^{-1}$ ). Therefore, this study provides justification for incorporating a representation of this process into models simulating the fate and distribution of pesticides in small water bodies. It would also be interesting to incorporate this additional uptake pathway for submerged aquatic vegetation into a food-web bioaccumulation model for lakes and rivers.

The results of the model application also suggest some useful experiments that could be performed in order to increase confidence in the model. For example, it would be interesting to attempt uptake studies using hydrophobic recalcitrant substances (to remove the confounding influence of degradation) in identical laboratory conditions as described by Gobas et al. (15) and then repeat the experiment with increasing concentrations of suspended solids in the water column. This type of experiment would provide strong evidence for or against the importance of particle deposition and could also suggest ways to refine and improve the mathematical description of the process. An alternative empirical approach for assessing the importance of particle deposition for hydrophobic contaminants would be to calculate apparent uptake rate constants based on observations from aquatic mesocosms as in Hand et al. (2) for systems with widely varying concentrations of suspended solids ( $C_{SS}$ ). Assuming that the sorptive capacity of the suspended solids was similar across the mesocosms (i.e., similar OC content), a positive relationship between  $C_{SS}$  and apparent uptake rate constants might be observed if particle deposition dominates the mass transfer of contaminants from the water column.

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## Supporting Information Available

A description of the model formulation and parametrization, three tables of parameter values, and two additional figures. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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