

Quantitative Structure—Property Relationships for Enhancing Predictions of Synthetic Organic Chemical Removal from Drinking Water by Granular Activated Carbon

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Granular activated carbon is a frequently explored technology for removing synthetic organic contaminants from drinking water sources. The success of this technology relies on a number of factors based not only on the adsorptive properties of the contaminant but also on properties of the water itself, notably the presence of substances in the water which compete for adsorption sites. Because it is impractical to perform field-scale evaluations for all possible contaminants, the pore surface diffusion model (PSDM) has been developed and used to predict activated carbon column performance using single-solute isotherm data as inputs. Many assumptions are built into this model to account for kinetics of adsorption and competition for adsorption sites. This work further evaluates and expands this model, through the use of quantitative structure—property relationships (QSPRs) to predict the effect of natural organic matter fouling on activated carbon adsorption of specific contaminants. The QSPRs developed are based on a combination of calculated topographical indices and quantum chemical parameters. The QSPRs were evaluated in terms of their statistical predictive ability, the physical significance of the descriptors, and by comparison with field data. The QSPR-enhanced PSDM was judged to give results better than what could previously be obtained.

Introduction

As awareness of the effects of trace level contamination on human health grows, many synthetic organic chemicals (SOCs) are now recognized as drinking water contaminants. The removal of such compounds from water has been of much interest for some time. The 1986 amendments to the United States Safe Drinking Water Act declared granular activated carbon (GAC) to be the best available technology for removing SOC. This is to say, if a treatability assessment revealed that a contaminant could be economically removed by GAC treatment, no further research would be required to justify a water utility's use of this technology to remove the contaminant.

Field-scale treatment data are considered the highest quality data on which to base such a treatability assessment. Unfortunately, these data do not exist for many contaminants, and they are often cost- and time-prohibitive to obtain. Therefore, single-solute isotherms in organic-free-water (OFW) are used when initially determining the practicality of activated carbon treatment for removing a contaminant from drinking water. OFW isotherms represent the ultimate capacity of a GAC for a contaminant of interest. However, for GAC columns that treat natural waters, there are many issues that can lower the carbon's capacity, and hence reduce the expected time to contaminant breakthrough. These include competition for adsorption sites from other contaminants, competition from background natural organic matter (NOM), preloading of NOM onto the activated carbon, biofilm development, temperature, pH, variable influent contaminant concentrations leading to desorption, and adsorption kinetics which are affected by carbon size and hydraulic loading rate. Among these issues, the effects of NOM on treatment have been found to be quite important. NOM is a subject of considerable interest for many aspects of drinking water treatment (1–5), and a number of researchers have evaluated the effects of background natural organic matter preloading on the adsorption of synthetic organic compounds (6–14).

Because GAC treatment occurs in flow-through contactors, these capacity-reducing issues cannot be directly evaluated from adsorption data developed using batch isotherm experiments. Therefore, there is much interest in developing mathematical models to predict the performance of carbon adsorbers. One such mathematical model is known as the Pore Surface Diffusion Model (PSDM) (6, 15). Bhuvendralingam (16) expanded upon this model, using the work of Zimmer (17) and Sontheimer et al. (6) to develop a method to account for changes in adsorption capacity with time, based on contaminant-specific capacity-reduction equations. Thereby, the PSDM allows water treatment professionals to estimate carbon use rates for GAC removal of the contaminant. The PSDM approach yields predictions which compare favorably with other approaches for estimating carbon use rates for some contaminants but not for others (18, 19). The contaminants showing variation were pesticides included on the USEPA Contaminant Candidate List (CCL, 20), the primary source from which future regulated drinking water contaminants may be selected. Consequently, it is necessary to resolve these estimation issues in order to enhance understanding of the capability of GAC to remove these potentially important contaminants.

To fulfill this objective, the present work presents an enhancement to the Bhuvendralingam (16) expansion of the PSDM model. To account for the capacity reduction arising from both the competitive adsorption and kinetics, Bhuvendralingam (16) assumed that all compounds belong to one of nine groups based on similarity of molecular structures. To apply Bhuvendralingam's expansion, the chemical of interest is placed into a structurally similar group, and the capacity-reduction equation of that group is then used in the calculation. Thus, capacity-reduction equations for all compounds are based on experimental data for a single representative compound from each group.

The work in the present paper is based on the observation that although it is a significant assumption in the PSDM model, there is no fundamental reason to assume that every synthetic organic chemical can be accurately described by the nine groups. It would be more accurate to measure the capacity-reduction equation for each compound indepen-

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dently, but this would be a cost- and time- prohibitive prospect. Therefore, it seemed reasonable to investigate alternate means to obtain these capacity-reduction equations through predictive methods. One technique for predicting properties of chemicals for environmental applications that has gained much attention recently is the use of quantitative structure–property relationships (QSPRs) (21–26). The use of QSPRs has increased in popularity due to the relatively recent development of high-performance desktop computer systems. QSPRs have been used for several aspects of carbon adsorption, such as adsorption of organic chemicals from air (27, 28), or from water onto activated carbon (29, 30) or activated carbon fiber (31–33).

In this paper, QSPRs were investigated as a means to obtain a more accurate assessment of preloading effects, specifically to refine the capacity equations that are a critical part of the current functioning of the PSDM. To develop the QSPRs, molecular and quantum chemical descriptors were correlated with existing experimental data to develop the preloading relationships for specific organic contaminants. The results are compared with the carbon adsorber characteristics calculated from preloading relationships using the original groups.

Theory and Approach

Pore Surface Diffusion Model. Carbon use rates were calculated via the PSDM model through the use of AdDesignS software (Michigan Technological University, Houghton, MI).

Although details of the PSDM model are found elsewhere, it is beneficial for purposes of later discussion to summarize some aspects of this model used for calculating carbon use rates. In the PSDM (6, 15), the film transfer rates were determined from the Gnielinski correlation (34), and the surface diffusion terms were determined by relating the surface diffusion flux to the pore diffusion flux (6). In the PSDM, to account for the presence of fouling, the flux ratio was set so that pore diffusion was the predominant intra-particle mass transfer mechanism. Pore diffusion kinetics were determined from the calculated liquid diffusivity (35) and tortuosity. The tortuosity, also known as the labyrinth factor, is discussed in more detail elsewhere (6), and the tortuosity quantitatively correlates with preloading time (17).

The PSDM model directly accounts for changes in GAC capacity over time due to NOM preloading. This relationship has been represented as the time dependent Freundlich K (K_t) divided by the Freundlich K in organic-free water (K_o). For trichloroethene (TCE), Zimmer (17) and Sontheimer et al. (6) found the relationship to be

$$(K_t/K_o)_{\text{TCE}} = 0.01 * [A1 - A2*t + A3*\exp(-A4*t)] \quad (1)$$

where t is time of GAC adsorber operation in weeks and $A1$, $A2$, $A3$, and $A4$ are empirical kinetic constants specific to a given SOC, water matrix, and GAC type. In the present study, 65.3, 0.676, 34.7, and 1.008 were selected for $A1$, $A2$, $A3$, and $A4$, respectively. These were the values determined for Karlsruhe groundwater (average dissolved organic carbon 0.8 mg/L during the study period); the use of these values yields median results when compared to other waters studied to date (6, 17). Thus, the results using these $A1$, $A2$, $A3$, and $A4$ values may provide a good starting point for the use of GAC at a particular location, although properties of the specific location's source water may need to be examined to determine the applicability of GAC for contaminant removal at a particular site.

Development of relationships such as eq 1 for other contaminants requires a great deal of experimental work, so Bhuvendralingam developed a correlation that related the

capacity reduction for a given contaminant to that for trichloroethene (16).

$$(K_t/K_o)_{\text{contaminant}} = a(K_t/K_o)_{\text{TCE}} + b \quad (2)$$

where a and b are the correlating factors that were determined for a number of contaminants that were then used to represent entire classes of compounds (16). In keeping with the intent as discussed in the Introduction, this paper will investigate the prediction of the correlating factors, a and b , for individual contaminants through the use of QSPRs.

Quantitative Structure–Property Relationships (QSPRs).

The use of QSPRs enables the conservation of resources by predicting the results of time-consuming and expensive experiments. Briefly, a QSPR is a mathematical relationship between the structure of a chemical and a property possessed by that chemical. For biologically active compounds, QSPRs are usually referred to as quantitative structure–activity relationships (QSARs) (25). To describe molecular structure attributes, various “descriptors”, which quantify different structural attributes of the molecule, are calculated (26) and correlated with a “training set” of experimentally determined values of the property of interest. In addition to the calculated descriptors, this correlation may also involve other, usually simple, properties of the chemical and employ simple linear regression, multilinear regression, stepwise regression, and/or neural networks. A statistical test is applied to the resulting equations to determine if the equations are predictive. Due to the nature of statistics, the equations that give the best fit do not necessarily have physical meaning. Often, however, a QSPR may be constructed in which the descriptors are physically meaningful. Descriptors that do not require any extensive computational ability (22, 26, 33, 36) have been popular, although quantum-chemical descriptors have become increasingly popular due to the relatively recent widespread availability of sufficient desk-top computational capability to permit the use of computationally intensive ab initio methods (21, 37).

The QSPR development effort involved using the commercial software CAChe (Fujitsu, Beaverton, OR). The software also provides an interface for calculating descriptors, developing QSPRs, and providing regression statistics. Quantum chemical descriptors were computed using MOPAC 2000 for semiempirical calculations and DGauss, a density functional program, for ab initio calculations. A large number of basis sets were investigated for both quantum chemical computation programs. The input geometry of each molecule was based on the geometry provided by the National Institutes of Standards and Technology via their on-line physical property database (38). The majority of the compounds tested were in this database. For compounds that were not, input data from an analogous compound were modified appropriately. The experimental data presented by Bhuvendralingam (16), along with that for nitrophenol (6, 17), were used as a training set for the QSPR model.

Results and Discussion

QSPR Development. As mentioned above, the data of Bhuvendralingam (16), along with that for nitrophenol (6, 17), were used as a training set for the QSPR model. The individual compounds used in the training set, along with their experimentally determined a and b values, are listed in Table 1. Table 1 does not list 3,4-dinitrophenol (16), which after initial calculations was excluded from the training set since its inclusion resulted in poor regression performance, particularly for quantum chemical descriptors. This may not be too surprising, given that a recent study of semiempirical molecular mechanical calculations of nitro compounds revealed that dinitro compounds may produce results that differ significantly from experimental values (39).

TABLE 1. Fitting Parameters Used in Capacity-Reduction Equation (Eq 2) for Specific Contaminants as Reported in the Literature and Determined through the Use of QSPRs (Eqs 3 and 4)

group (surrogate contaminant) ^a	literature ^a fitting parameters		QSPR fitting parameters		difference (QSPR–literature)	
	<i>a</i>	<i>b</i>	<i>a</i>	<i>b</i>	Δa	Δb
pesticides (atrazine)	0.00	0.05	0.00	0.14	0.00	0.09
trihalomethanes (chloroform)	1.00	0.00	1.10	−0.11	0.10	−0.11
chlorinated hydrocarbons (1,4-dichlorobenzene)	0.59	0.41	0.69	0.30	0.10	−0.11
phenols (2,4-dichlorophenol)	0.65	0.35	0.69	0.24	0.04	−0.11
polynuclear aromatics (methylene blue)	0.32	0.68	0.37	0.65	0.05	−0.03
not assigned (nitrophenol)	0.65	0.47	0.65	0.54	0.00	0.07
aromatics (toluene)	0.90	0.10	0.78	0.22	−0.12	0.12
halogenated alkane (1,1,1-trichloroethane)	1.20	−0.20	1.22	−0.12	0.02	0.08
halogenated alkene (trichloroethene)	1.00	0.00	0.81	0.00	−0.19	0.00
average ^b					0.0 ± 0.1	0.0 ± 0.1

^a Reported in Bhuvendralingham (16), except for nitrophenol which is reported in Zimmer (17). ^b Reported as $x \pm \sigma_{n-1}$.

For the compounds in Table 1, a number of quantitative physical properties and descriptors were selected as inputs into the QSPR-development software package. This selection was based on the literature and also on which properties were thought to be related to carbon adsorption phenomena. These included conformational energy, connectivity indices, dielectric energy, dipole moment (and components thereof), electron affinity, group count (nitro, primary amine, and secondary amine), heat of formation, ionization potential, molar refractivity, molecular orbital energies, molecular weight, octanol–water partition coefficient (experimental and calculated), polarizability, shape indices, solvent accessible area, steric energy, and valence connectivity indices. After the software calculated the descriptors, multilinear regressions were performed to correlate the parameters with the training set, and based on maximizing r^2 and rCV^2 values, the final QSPRs developed to describe the capacity reduction (eq 2) were

$$a = -0.0624 \log K_{ow} - 1.15 \log \kappa_2 - 0.166N_{am} + 1.37, \\ r^2 = 0.932, \quad rCV^2 = 0.791 \quad (3)$$

$$b = 0.0441\alpha + 0.406D_{z(hybrid)} - 0.250, \\ r^2 = 0.892, \quad rCV^2 = 0.481 \quad (4)$$

where K_{ow} is the experimental octanol–water partition coefficient, κ_2 is the second-order shape index, N_{am} is the sum of the number of primary and secondary amine groups, α is the molecular polarizability calculated by Dgauss (DZVP basis set), and $D_{z(hybrid)}$ is the largest hybrid component of the dipole moment perpendicular to the long molecular axis. $D_{z(hybrid)}$ was calculated by MOPAC 2000 (PM3 basis set) and was manually extracted from the output file of the MOPAC 2000 program.

One way of looking at QSPRs is to evaluate the correlation between experimental and calculated properties. For this purpose, several types of statistical calculations were performed. First, the rCV^2 values, which are indicative of the predictive power of the QSPR, were calculated, and they are greater than 0.2 for both a and b , which adds confidence to the predictive power of the QSPR (36, 40, 41). A rCV^2 value of less than 0.2 indicates low predictive power, and a rCV^2 value of less than 0.0 indicates no predictive power. Second, the differences were calculated between the experimental parameters and those estimated using the QSPRs. As seen in Table 1, these differences are generally minor and may be attributed to both experimental error and uncertainty in the calculated values. In fact, the average difference for both the a and b values is 0.0 ± 0.1 , and the individual differences between the experimental and calculated values are the same within the 95% confidence limit of the average. Third, an

ANOVA analysis was performed to investigate whether the correlation between the experimental and QSPR-estimated a and b values represented a statistically significant trend. The resulting p values of the slope were 0.00002 and 0.0001, respectively, indicating that the trends are significant at the 95% confidence level, i.e., p was less than 0.05. The significant trend reflected in the p values arises statistically in part from the number of compounds in the training set compared to the relatively small number of descriptors, i.e., the degrees of freedom in the regression. In summary, the results of all these statistical analyses lend confidence to the QSPRs in eqs 3 and 4. As noted previously, it is possible for a QSPR to be statistically valid, but to have little physical meaning, so it is useful to examine each term for physical meaning relative to the carbon adsorption process.

For the terms in eq 3, the experimental octanol water coefficient, K_{ow} , is related to hydrophobicity and solubility which is clearly correlated to adsorption through the Polanyi, Dubinin, and Solvophobic models for carbon adsorption (42–46). In addition, K_{ow} plays a role in QSPRs developed for adsorption phenomena (23, 29, 30). N_{am} , the sum of the number of primary and secondary amine groups, is an indication of hydrophilic behavior. The amine functional group counts were implicated as an important descriptor in a QSPR study of water solubility (41), presumably by affecting the dipole–dipole, dipole-induced dipole, and hydrogen bonding interactions (36) that are also important in the GAC adsorption process. The second-order molecular shape index, κ_2 , is an indication of molecule shape. Several researchers (46–49) have demonstrated the effect of molecular shape, or size, on adsorption. Various molecular indices have also appeared in several QSPRs related to carbon adsorption (22, 23, 27, 31–33). Although there are a large number of number indices which may be calculated, there is no consensus on which is best. In developing the QSPR in eq 3, eight other molecular indices were investigated, but none of them produced a satisfactory result. The κ_2 shape index has been applied to predict the ability of molecules to fill cavities, such as in drug design applications (26). This situation bears an intriguing and gratifying analogy to the filling of a porous site on an activated carbon surface by a contaminant molecule.

For the descriptors in eq 4, the molecular polarizability (α) and dipole moment ($D_{z(hybrid)}$) may be related to adsorption through the interaction energy between the adsorbate, adsorbent, water, and presumably in this case, the background natural organic matter (43, 48, 50). Physically, the molecular polarizability and dipole moment can be used to describe nonionic molecular interactions that generally occur through dipole–dipole, dipole–induced dipole, and induced dipole–induced dipole mechanisms. The dipole interactions

TABLE 2. Fitting Parameters for the Chemical Groupings and Individual Contaminants as Determined by QSPRs

group (surrogate compound) ^a contaminant	group fitting parameters ^a		QSPR fitting parameters		difference (QSPR-group)		sum of squares difference	
	<i>a</i>	<i>b</i>	<i>a</i>	<i>b</i>	Δa	Δb	group	QSPR
halogenated alkanes (1,1,1-trichloroethane)	1.20	-0.20	0.84	0.11	-0.36	0.31	11.1	0.3
1,2-dichloropropane	0.00	0.05						
pesticides (atrazine)								
aldicarb sulfone			0.42	0.42	0.42	0.37	19.0	2.8
aldicarb sulfoxide			0.39	0.56	0.39	0.51	14.1	4.0

^a Reported in ref 16.

are related to the dipole moment, and induced dipole interactions are a function of the molecular polarizability. The overall dipole moment was employed in a QSPR developed for adsorption of gaseous chemicals to microporous carbon adsorbents (28). Breaking up the overall dipole moment into its geometrical components has been previously utilized (21), and the components are sometimes presented as the sum of the point charge and hybrid part. In the present QSPR, only the hybrid part of the dipole is used. The hybrid part arises from molecular orbitals that presumably interact with orbitals arising from the activated carbon surface. Thus, the hybrid part of the dipole moment would seem to be appropriate in terms of the electronic interaction between the adsorbing chemical and the activated carbon.

Comparison of Field-Scale Data to PSDM Predictions With and Without QSPRs

Taken as a whole, it appears that the QSPRs (eqs 3 and 4) generally contain terms that are reasonable when considering the physical phenomena that occur during adsorption. As a measure of the practical applicability of the QSPRs developed, field scale data were compared with PSDM model predictions, calculated with and without the use of the QSPRs. Details of field-scale conditions appear elsewhere (51, 52); carbon adsorption break-through data were available for 1,2-dichloropropane, aldicarb sulfoxide, and aldicarb sulfone in groundwater (total organic carbon of 2.0 mg/L). Using PSDM, carbon adsorption data were predicted using *a* and *b* parameters determined with the QSPR approach and without, i.e., by assigning each contaminant to the "representative group" of compounds defined in the literature (16). Table 2 shows the "representative group" parameters, *a* and *b*, previously reported (16) along with those calculated using the QSPR approach for the contaminants studied herein.

Table 2 indicates the calculated differences between the *a* and *b* values for the QSPR and "representative group" approaches. Interestingly, the differences are of similar magnitude for all these compounds, even though 1,2-dichloropropane appears to be structurally more similar to its "representative group" compound, 1,1,1-trichloroethane (in the halogenated alkane group), than aldicarb sulfoxide and aldicarb sulfone are to their "representative group" compound, atrazine (in the pesticide group). This suggests that it can be difficult to estimate structural similarity, particularly as it relates to carbon adsorption, by "representative group" descriptions, e.g., halogenated alkane or pesticide. In fact, the pesticide group description does not define the contaminant's structure in any way.

Figure 1 compares experimental data to PSDM contaminant removal predictions employing the "representative group" approach and also the QSPR approach. The results for 1,2-dichloropropane, aldicarb sulfone, and aldicarb sulfoxide are shown. The visual improvement in the predictions via the QSPR approach is reassuring with regards to the ability of the QSPR to enhance the predictions. In fact, the

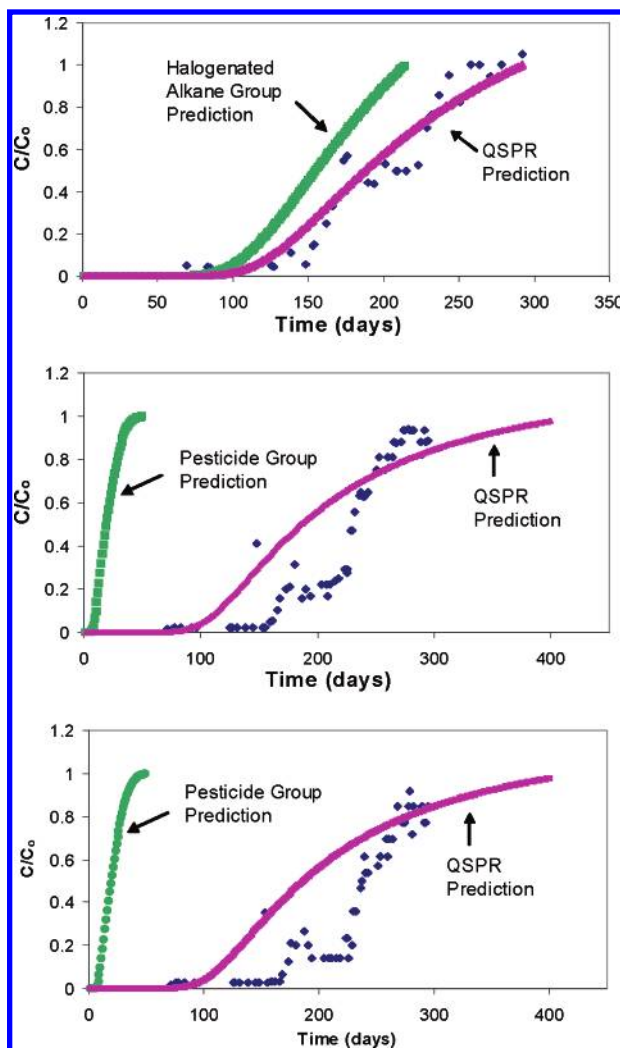


FIGURE 1. (Top) Plot of C/C_0 values for 1,2-dichloropropane in the effluent of the granular activated carbon contactor fed by a groundwater. Also shown is the C/C_0 values predicted through the use of the capacity reduction equation of the "halogenated alkane" group, along with the C/C_0 values predicted through the use of the capacity reduction equations based on QSPR. (Middle) Similar plot for aldicarb sulfone, comparing the results calculated using the "pesticide" group to those of the QSPR prediction. (Bottom) Similar plot for aldicarb sulfoxide.

QSPR approach results in a visually dramatic change for aldicarb sulfone and sulfoxide, because the "pesticide group" predictions do not appear to intersect the experimental data at all. Previous work (19) with other compounds has shown "pesticide group" predictions to have inherent discrepancies as compared to other groups. To quantitate the differences, the sum-of-squares difference between the experimental data and the predicted values are reported in Table 2. In all cases,

the sum of squares difference is lower for the QSPR prediction than for the "pesticide group" prediction.

General Applicability and Limitations of the QSPRs in the PSDM Model

The data above suggest that QSPRs may help to both increase the quality of the prediction and also simplify the application of the PSDM, by eliminating the need to place a particular contaminant in a group. The general applicability of QSPRs in the PSDM model is related to a number of issues, some related to the PSDM model itself, and some related to the QSPRs, which essentially provide enhanced input parameters for the PSDM. For example, natural waters can differ significantly in the quantity and composition of their dissolved natural organic matter (NOM), which is implicated in the reduction of adsorption capacity for SOCs. In applying the PSDM, the user should select kinetic constants (eq 1) appropriate for the water under study. Five sets of these constants are supplied with the PSDM software to represent a range of natural waters (16); each one affects to a different extent the time predicted for contaminant breakthrough. Figure 1 alludes to the reasonableness of this approach, in that the kinetic constants selected were for a representative groundwater, and the water in the field test was also a groundwater.

In the PSDM calculations, the relative capacity reduction equation for specific chemicals (eq 2) is considered to be independent of the choice of constants for eq 1. Thus, the QSPR enhanced estimation of the parameters in eq 2 is, in principle, independent of the NOM in the water. Indeed, examination of eqs 3 and 4 reveals no terms that are necessarily related to the composition of the NOM. Eq 3 contains terms related to properties of the adsorbing species, and eq 4 contains terms that seem to relate to the electronic interaction of the adsorbing species with the adsorbent, i.e., GAC. Capacity reduction by NOM functions partly by competition for adsorption sites, although NOM can alter transport properties of the GAC, e.g. via pore-blockage. However, the terms in eqs 3 and 4 do not seem to reflect the latter.

A sophisticated QSPR treatment could be devised to investigate any potential, direct interaction of NOM with GAC and adsorbing species that is not accounted for in eqs 3 and 4. There are a number of technical factors that limit the application of QSPRs with regard to this type of study. For example, even in the same water source, NOM may have a range of structures, some of which may be very large and difficult to experimentally determine. Even if the precise chemical structures could be experimentally determined, the variability, number, size, and complexity of these molecules may require substantial computing power. Accordingly, until technology exists to accomplish both the needed experiments and computations, it may be beneficial to consider the merits suggested by Figure 1 and Table 2 and to continue applying the assumption of the PSDM model that the capacity reduction equations are independent of NOM.

Another issue involving the applicability of QSPRs relates to the ability to calculate the terms in eqs 3 and 4 for a variety of compounds. These terms can often be calculated using programs that are available for desktop computer use, enabling their widespread application. For example, the terms in eq 3 are relatively straightforward to estimate from the molecular structure. K_{ow} , if it is not known experimentally, can also be estimated by a number of programs, including the software used in this study and EPA's EPI Suite software (53). The terms in eq 4 rely on quantum-chemical calculations, which leads to several intricacies. For example, the QSPRs may be limited in their application to dinitro-

compounds, perhaps due to limitations in the computational methods for these compounds, although the QSPRs might be improved through the use of a technique recently reported to correct for these limitations (39). Similar fundamental issues relate to quantum mechanical calculation of other large and complex molecules, as well as those containing transition and perhaps metalloid substituents. However, if, like typical dinitro compounds, such compounds absorb with sufficient strength (i.e., have sufficiently large Freundlich K values) then PSDM model predictions may not be affected much. Accordingly, improvement of the QSPRs for these compounds may not be warranted.

In summary, from a practical standpoint, the detailed resolution of the above issues may not be necessary to accomplish the purpose of the present work, which was to investigate QSPRs as a means to estimate the capacity reduction (eq 2) of numerous compounds. Accordingly, these QSPRs might be directly applied to a large number of other chemicals, such as CCL compounds (55). Experimental data can never be replaced with QSPR calculations; however, obtaining experimental data for a large number of compounds would take years to experimentally produce and would be prohibitively expensive. Therefore, the use of QSPRs can provide another valuable modeling tool for improving the initial evaluation of the effectiveness of GAC treatment for compounds of interest to human health and the environment.

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The views expressed herein have not been subjected to U.S. Environmental Protection Agency review and therefore do not necessarily reflect the views of the Agency, and no official endorsement should be inferred. Mention of trade names or commercial products in this article does not constitute endorsement or recommendation for use by the U.S. EPA.

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