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An updated Quantitative Water Air Sediment Interaction (QWASI) model for evaluating chemical fate and input parameter sensitivities in aquatic systems: Application to D5 (decamethylcyclopentasiloxane) and PCB-180 in two lakes

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HIGHLIGHTS

- The QWASI model is a fugacity-based mass balance chemical fate model.
- The model was reformatted to encourage “Good Modeling Practices”.
- The model was revised in spreadsheet format to accommodate sensitivity analysis.
- The model is illustratively applied to two chemicals in two lakes.
- Two strategies for assessing uncertainty are discussed.

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ABSTRACT

The QWASI fugacity mass balance model has been widely used since 1983 for both scientific and regulatory purposes to estimate the concentrations of organic chemicals in water and sediment, given an assumed rate of chemical emission, advective inflow in water or deposition from the atmosphere. It has become apparent that an updated version is required, especially to incorporate improved methods of obtaining input parameters such as partition coefficients. Accordingly, the model has been revised and it is now available in spreadsheet format. Changes to the model are described and the new version is applied to two chemicals, D5 (decamethylcyclopentasiloxane) and PCB-180, in two lakes, Lake Pepin (MN, USA) and Lake Ontario, showing the model's capability of illustrating both the chemical to chemical differences and lake to lake differences. Since there are now increased regulatory demands for rigorous sensitivity and uncertainty analyses, these aspects are discussed and two approaches are illustrated. It is concluded that the new QWASI water quality model can be of value for both evaluative and simulation purposes, thus providing a tool for obtaining an improved understanding of chemical mass balances in lakes, as a contribution to the assessment of fate and exposure and as a step towards the assessment of risk.

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1. Introduction

In 1983 a set of two papers was published describing simple models of chemical fate in lakes and rivers (Mackay et al., 1983a, 1983b), namely the QWASI (Quantitative Water Air Sediment Interaction) models. The models were compiled using the fugacity concept and have been made freely available as software from the website of the Canadian Centre for Environmental Modelling and

Chemistry (CEMC) and have been widely applied to specific environmental systems (e.g. Mackay, 1989). The QWASI lake model addressed here treats a well-mixed water body and includes the processes depicted in the mass balance figures presented later. Chemical inputs are by direct emissions, advective inflows of water and suspended particles, and deposition from an atmospheric compartment with a defined concentration by wet and dry aerosol and gaseous deposition. Steady state mass balance equations are compiled separately for the water and sediment compartments with chemical masses and concentrations expressed as fugacities in water and in sediment. Although the model simulates a steady

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state condition it can be adapted to treat dynamic conditions as a pair of first order differential equations that can be solved analytically or numerically. The equations are readily interpretable because all process rates are expressed as products of the fugacity in the source phase and a D value which can represent reactions, diffusion or advective transport. Full details of the equations and assumptions are given in the original papers and in the text by Mackay (2001). The simplicity of the equations has facilitated the extension of both the lake and river models to treat more complex multi-compartment systems such as segmented lakes (Ling et al., 1993; Diamond et al., 1994, 1996; Mackay and Hickie, 2000) or they can be set up as a number of well mixed connected river reaches (Warren et al., 2005, 2007; Ethier et al., 2008). Insights into the time response of the system can be obtained by examining the residence times of the chemical in each compartment.

In a recent paper (Hughes et al., 2012) we described an updated state of the science Equilibrium Criterion (EQC) model that is used to provide a screening level evaluation of the likely fate of a chemical that is introduced into a multi-media environment by various modes of entry. This evaluation can then be followed by a more detailed system-specific evaluation such as the QWASI model. This EQC screening level approach was described for the volatile permethyl siloxane D5 (decamethylcyclopentasiloxane, CAS No 541-02-6). In this paper we illustrate this type of evaluation by application of an updated state of the science QWASI model to D5 and PCB-180 (2,2',3,4,4',5,5'-heptachlorobiphenyl, CAS No 35065-29-3) for two aquatic receiving environments, namely Lake Pepin (MN, USA) and Lake Ontario. Whelan (2013) has recently reported a similar evaluation using a custom-modified QWASI model and illustrated the importance of determining parameter sensitivities. In the present study we expand on these simulations using the new QWASI Lake model. Finally, we outline and illustrate strategies for determining and displaying a comprehensive analysis of the sensitivities of the results to each input parameter and uncertainties in the key output parameters.

2. Application of the model

If an estimated or measured discharge rate of a chemical into a specific water body is available, for example from a Waste Water Treatment Plant (WWTP), the model can translate this rate into estimated steady state concentrations. These concentrations can be used for screening level assessments of fate and exposure, and can be compared with monitoring data to investigate whether the fate processes are being reliably simulated. The model also estimates the absolute and relative quantities of the chemical and its corresponding residence times in water, suspended particles, sediments and in the system as a whole. This can help guide monitoring efforts and provides insights into the likely response times of the compartments to changes in emission rates, for example, remediation following a reduction or cessation of discharges.

Rates of key transport processes are estimated, thus the relative importance of competing loss processes such as degrading reactions, volatilization, advective outflow and sedimentation can be ascertained. This can identify the key transport and transformation rate parameters and justify efforts to obtain more accurate values.

Finally, by using fugacity, the relative equilibrium status of compartments becomes obvious. An example is air–water exchange in which the net driving force for diffusion can be from water to air or air to water depending on local conditions. A simple direct estimate can be made of equilibrium concentrations in biota resident in water and sediment by assuming equi-fugacity to deduce equilibrium concentrations in organism lipids. These simple estimates can justify the application of more detailed

bioaccumulation and food web models such as that of Arnot and Gobas (2004).

It can be argued that the fate of a chemical in an aquatic system is not fully understood until a full mass balance accounting is obtained. The QWASI model can achieve this goal.

3. Specific changes to the model

Feedback from users of the model for the above purposes indicates a need for an updated version that better meets current scientific and regulatory needs. First, although the software version has proved to be reliable and easy to use, there has been an expressed need for a version that is based on a Microsoft Excel spreadsheet platform. The use of this spreadsheet makes the model more readily available to potential users and sensitivity and uncertainty analyses are more readily conducted. This latter aspect has proved to be particularly important from a regulatory perspective by enabling the user to identify the more sensitive parameters and explore the implications of variations in input parameter values. The use of this platform facilitates linking it to other spreadsheet models that may address the efficiency in WWTPs, run-off from soils and bioaccumulation in organisms or food webs. Accordingly, the new model is compiled as an Excel spreadsheet and, as with the previous version, it is freely available from the Trent University CEMC web site (www.trentu.ca/cemc).

Second, in the original model partition coefficients were calculated from solubility, vapor pressure and octanol water partition coefficient (K_{ow}), the last of which was used to estimate partitioning to organic carbon in sediments and particles as K_{oc} . Modern regulations often require empirical rather than estimated values derived from simple K_{ow} – K_{oc} quantitative structure–activity relationships (QSARs). More sophisticated methods are now available to calculate partition coefficients such as using Linear Free Energy relationships as reviewed by Endo et al. (2013) and quantum chemical programs such as SPARC (Hilal et al., 2003) and COSMOtherm (Klamt, 2005). In the new version all partition coefficients are input directly. Guidance is provided in a worksheet on the likely magnitude of certain values and their temperature dependence.

Third, all input data are summarized in three worksheets and space is provided to document the data sources and perceived accuracy of all input values. Notes and comments can be included. Such information is invaluable when the model output is examined by another party, for example during regulatory proceedings. This approach is in accord with the principles of Good Modeling Practice described by Buser et al. (2012).

4. Application of the model to D5 and PCB-180

The updated QWASI model is applied here for illustrative purposes to two chemicals, D5 and PCB-180 in two lakes, Lake Pepin, which is a natural lake on the Mississippi River located approximately 80 km downstream of Minneapolis and Saint Paul, and Lake Ontario. These lakes differ greatly in size, depth, trophic status, and hydraulic retention time, Lake Pepin being some 12 d and Lake Ontario some 7 years. The properties of the chemicals and lakes are given in Table 1. The assumed temperature was 9 °C in Lake Ontario and 14 °C in Lake Pepin as suggested by Whelan (2013). Illustrative emission rates of 100 kg year⁻¹ of the chemicals are assumed for both lakes. It is emphasized that these rates are entirely hypothetical, the aim being to evaluate and compare both the relative fates of the two chemicals, behaviors in small and large lakes and display the model's capabilities.

Although it is subject to thermal stratification, Lake Pepin has a relatively low volume and short retention time and can probably

Table 1

Chemical and environmental properties and dispersion factors (k) used in the simulations of D5 and PCB-180 in Lake Ontario and Lake Pepin. All chemical properties are at 25 °C. References and justifications of chosen values are detailed in the [Supplemental Data](#).

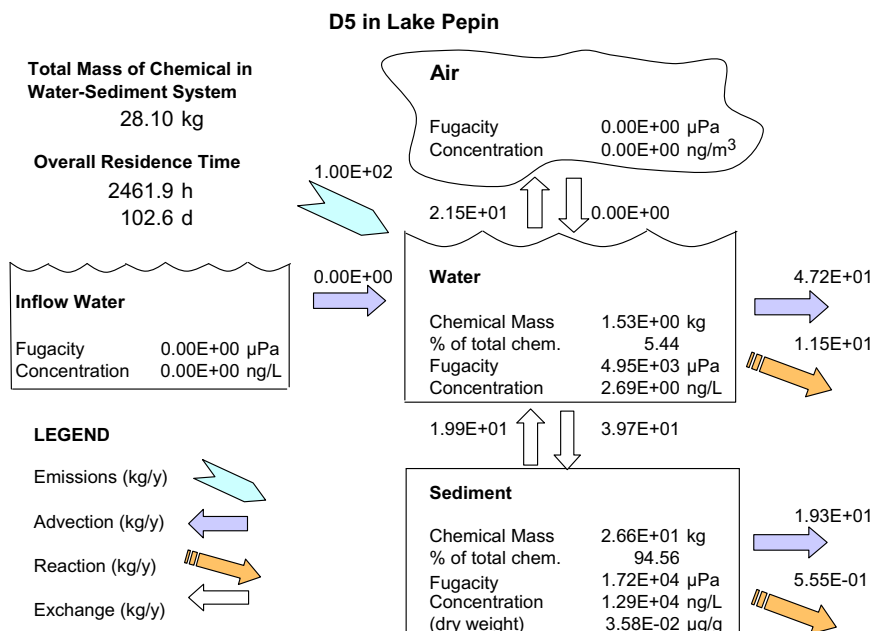
Property	Symbol	k	
		D5	PCB-180
K_{OC} (L kg ⁻¹)	KOC	2.00×10^5	3.89×10^6
K_{AW}	KAW	1.35×10^3	1.55×10^{-2}
Half-life in water (h)	HLW	207	2.4×10^5
Half-life in sediment (h)	HLS	7.44×10^4	3.3×10^5
		Lake Ontario	Lake Pepin
Surface area (m ²)		1.91×10^{10}	1.03×10^8
Lake volume (m ³)	VOL	1.64×10^{12}	5.67×10^8
Outflow (m ³ h ⁻¹)	OUT	2.6×10^7	2.0×10^6
Sediment depth (m)	SD	0.02	0.02
Conc. of susp. sed. (mg L ⁻¹)	CSS-W	0.64	10
Sed. dep. rate (g m ⁻² d ⁻¹)	SDR	1.4	33.4
Sed. resusp. rate (g m ⁻² d ⁻¹)	SRR	0.6	14.31
Sed. burial rate (g m ⁻² d ⁻¹)	SBR	0.6	14.31
f_{OC} , susp. sediment	Foc, ss	0.25	0.12
f_{OC} , resusp. sediment	Foc, re	0.035	0.035
f_{OC} , sediment	Foc, sed	0.04	0.04
Sed. solid fraction (vol vol ⁻¹)	CSS-S	0.15	0.15
Density of susp. sed. (kg m ⁻³)	DSS	2400	2400
Density of sed. solids (kg m ⁻³)	DS	2400	2400
Air-side volat. MTC (m h ⁻¹)	MTCA	1	1
Water-side volat. MTC (m h ⁻¹)	MTCW	0.01	0.01
Sed.-water diff. MTC (m h ⁻¹)	MTC _{SW}	0.0004	0.0004

be treated as fairly well mixed. In contrast, the larger Lake Ontario is expected to display considerable spatial variation in concentrations because of multiple chemical sources, intense thermal stratification, near-shore currents induced by wind and riverine inputs as well as differential sedimentation in near-shore areas and in deep basins. The calculated concentrations must therefore be regarded as only order-of-magnitude averages. An example of such an evaluation is the application of a dynamic version of the single water compartment QWASI model to Lake Ontario for total PCBs by Mackay (1989) that provided estimates of concentrations in water and sediment that were generally consistent with monitoring data. To simulate concentrations in large lakes more accurately requires

a multi-compartment and possibly a seasonally dependent model such as the LOTOX 2 Lake Ontario model of Kaur et al. (2012).

The results of the four simulations are given in Figs. 1–4 as mass balance diagrams. The model output includes a complete tabulation of all properties, concentrations, fugacities, masses and flow and reaction rates. A sample is provided in the [Supplementary Data](#).

Figs. 1–4 show that there are large differences in fate between the two chemicals and between the two lakes. The chemicals display differences in fate, mainly because of their differences in degradation half lives and their air–water partition coefficients (K_{AW}). Volatilization is much more significant for D5 which has a K_{AW} 10^5

**Fig. 1.** Mass balance diagram for D5 in Lake Pepin.

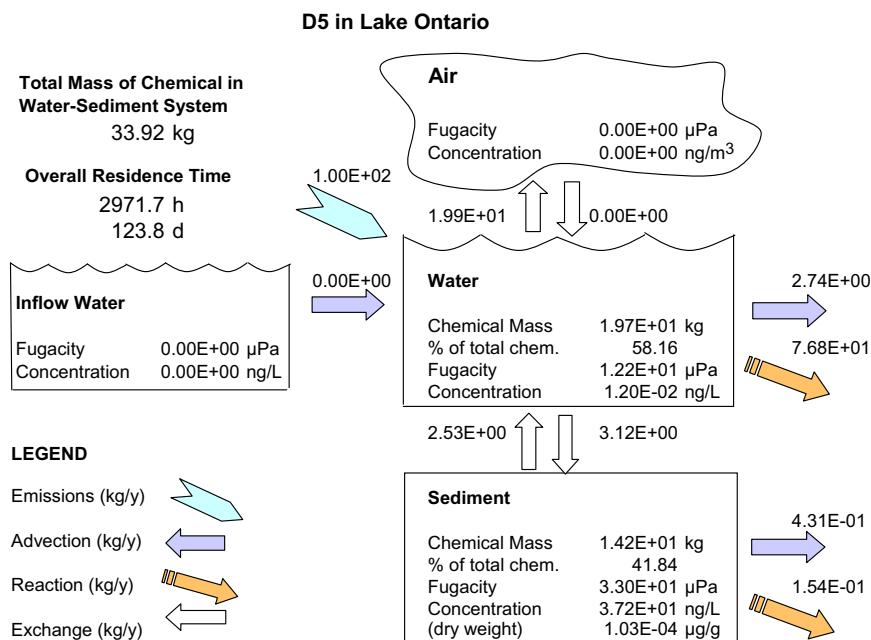


Fig. 2. Mass balance diagram for D5 in Lake Ontario.

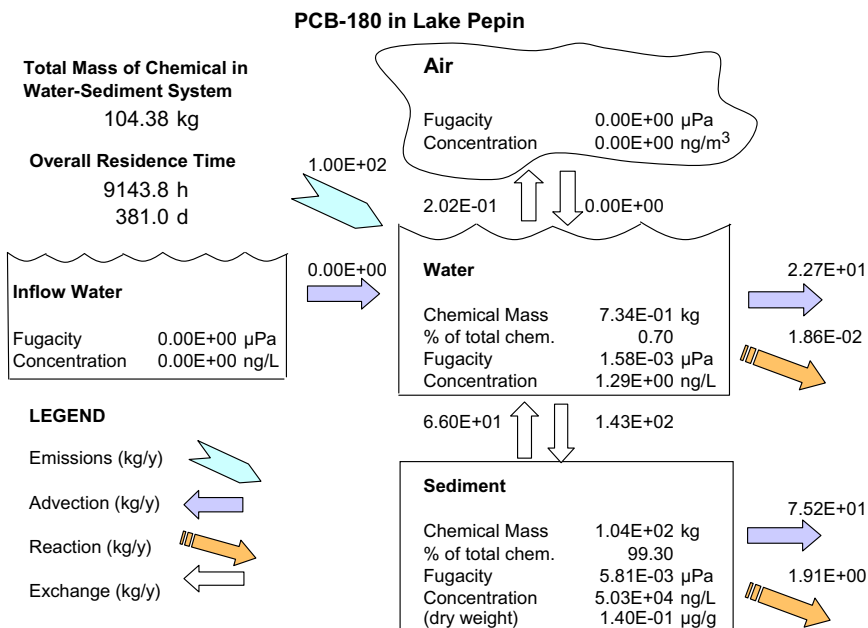


Fig. 3. Mass balance diagram for PCB-180 in Lake Pepin.

times that of PCB-180. Both substances have large K_{OCs} , thus they tend to associate with suspended matter in the water column and are subject to appreciable sedimentation. The mass distributions between water and sediment (which are independent of emission rates) of the two chemicals in Lake Pepin are similar. In contrast, in Lake Ontario 42% of the D5 partitions to sediment compared to 99% of the PCB. An interesting and at first counterintuitive observation is that the residence times of the less persistent D5 is similar in Lake Pepin (103 d) to that in Lake Ontario (124 d). The reason for this is that in the shallower Lake Pepin there is faster sediment deposition of D5 of 40 kg year^{-1} compared to 3 kg year^{-1} in Lake Ontario, resulting in a higher fraction of the chemical mass in the sediment. The overall residence time is a function of the half lives in water and sediment and the relative mass distributions

as influenced by the sediment water partition coefficients, the relative volumes or depths and by the rates of other loss processes such as volatilization. The slow degradation rate of PCB-180 results in residence times of about a year in Lake Pepin and about 20 years in Lake Ontario.

The key output quantities are regarded as the relative steady state masses in water and sediment and their respective residence times or persistence. These residence times not only express the potential of the chemicals to establish high concentrations, but they also indicate the time that would be required for substantial elimination of the substances from the aquatic ecosystems. For a more rigorous evaluation of remediation response times a Level IV model is required. The model also explains the cause of these differences by quantifying the relative magnitudes of the various

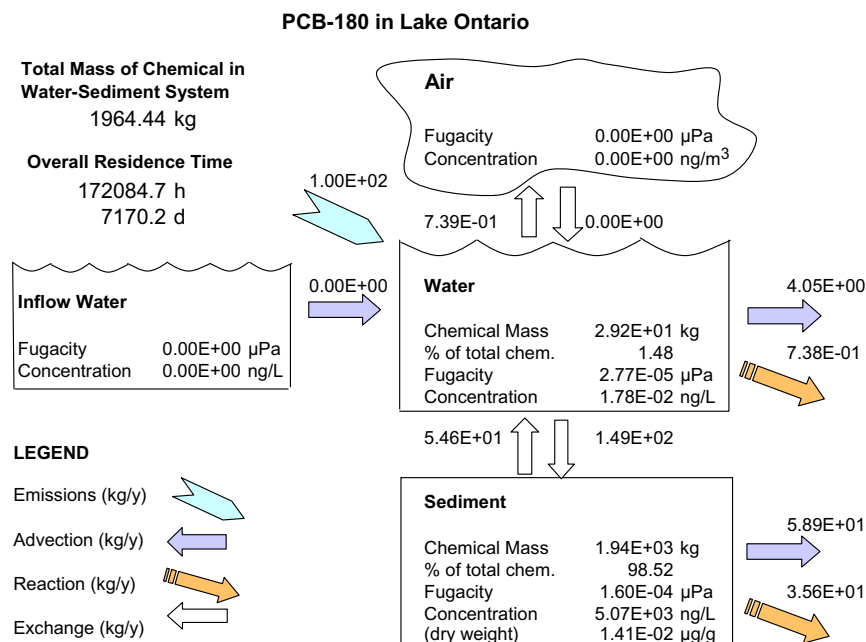


Fig. 4. Mass balance diagram for PCB-180 in Lake Ontario.

contributory processes by advection from water (outflow), from sediments (burial), degrading reactions in water and sediment and volatilization. A significant mass transfer rate as revealed by the model may indicate a need to obtain a more accurate estimate of the corresponding rate coefficient. Also of interest are the relative fugacities in water and sediment since these can provide insights into bioaccumulation in pelagic and benthic organisms and may justify the application of more detailed food web models.

In conclusion, the environmental fate of these two substances in the two lakes is a complex function of physical chemical properties and the relative dimensions and hydrology of the two lakes. Insights into the key differences in fate are best obtained by comparing the relative chemical masses in the water and sediment as well as their relative equilibrium status as indicated by the ratios of sediment and water fugacities. A mass balance model is essential for elucidating the effects of these parameters, for identifying the most significant fate processes and for providing a visual depiction of the key fate processes.

5. Sensitivity and uncertainty analyses

Mass balance models such as QWASI are necessarily simplifications of a complex reality and as such it is inevitable that the output results are subject to error. If the model is used as a simple evaluative tool the aim is to obtain output data that correctly reflect all the inherent equations, values of input parameters and simplifying assumptions and provide a depiction of the principal fate and transport processes. Rates of chemical input can be illustrative rather than real and no validation is sought. On the contrary, if the model is used as a predictive or simulation tool, real emission data are required and there is a possibility of validating the results by comparison with monitoring data. It then becomes important to define, explain and justify the magnitude of the likely errors in model output quantities, especially in relation to available data. There is a considerable literature on this topic including analyses of error propagation and uncertainty in the compilation by Saltelli et al. (2006) and specific applications to fugacity models by MacLeod et al. (2002), Kühne et al. (1997) and Luo and Yang (2007).

5.1. Strategies for assessing uncertainty

Two general strategies can be adopted. Commercial software that performs random sampling such as Crystal Ball can be used to generate a distribution of all output quantities from assumed distributions of the input quantities by employing Monte Carlo or the more efficient Latin Hypercube methods. The results can include details of the contributions of the various input variances to the output variances, thus identifying the most important sensitivities. Defining the distribution of input parameters requires the mean or median value and selection of an appropriate distribution, log-normal being used here because it samples only positive values. Uncertainty can be expressed by the related variances, standard errors, confidence intervals, and dispersion factors. A total number of samples is selected sufficient to cover the range of input values. This was done for the four cases described earlier.

The second strategy is to do the analyses by “One At A Time” (OAT) direct inspection. This is also illustrated here and is shown to give similar results. This can be labor intensive if done by hand, or it can be coded in a macro. It can provide additional insights into the nature and causes of the sensitivities and uncertainties. Any analyses should include documentation of the initial steps.

5.2. Definition of parameters

Table 1 lists the key input parameters and the magnitude of the expected variability expressed as a dispersion factor (k) on a log-normal basis to avoid the inadvertent definition of negative quantities. Here k is defined as the factor about the mean or most likely value which includes 95% of the expected values. For example, a mean of 50 and a dispersion factor of 2 imply that 95% of the expected values lie between $50/2$ and 50×2 . This factor is close in value to two standard deviations on a log normal basis. This assignment of dispersion factors is ultimately a matter of judgment based on observations of reported values, perceived accuracy and inherent variability attributable to environmental factors such as temperature. To our knowledge there is no universally accepted, rigorous non-judgmental process to guide this assignment. A

preferred approach may be to solicit estimates of dispersion factors from a group of experts who can enter into a dialog to suggest consensus values, thus avoiding excessive optimism or pessimism.

5.3. Results from random sampling

The analysis was performed using Crystal Ball version 7.3.1. The simulations were based on repeated Latin Hypercube random sampling ($n = 10000$) of the probability distributions defined for the principal sources of variation of each assumed parameter. The overall sensitivity was determined by the Spearman rank order correlation coefficient between the input and output data. In addition, the contributions of the inputs to the total variance of the outputs were calculated as the square of the correlation coefficient normalized to the sum of the squared correlation coefficients. The outputs chosen for examination are the concentration in water and in sediment and the residence time. A summary of the key findings are presented here.

For D5 in Lake Pepin, the median concentration in water is 21.8 ng L^{-1} with a dispersion factor of 1.39. For the sediment the median is $273 \text{ ng g}^{-1} \text{ DW}$ and the dispersion factor is 2.31. The residence time has the largest dispersion ($k = 4.33$). In simple terms, it is expected that 95% of the output quantities will lie within a factor in the range 1.4–4.3 of the value calculated by the model. The concentration in water is primarily controlled by the volatilization mass transfer coefficient (MTCW) with contributions from the degradation rate in water, sediment deposition rate, K_{OC} and outflow rate (Fig. 5). The concentration in sediment is controlled by K_{OC} , the sediment deposition and burial processes, the sediment half-life having little influence because it is so long. The overall residence time is controlled by the sediment depth, density of sediment solids, K_{OC} , sediment deposition and burial rates, and concentration of particulate in the water column.

For D5 in Lake Ontario dispersion factors of 2.31 in the concentration in water and 3.94 in the sediment were calculated. The residence time has a dispersion factor of 3.29. In simple terms, it is expected that 95% of the output quantities will be within a factor of approximately 2.3–3.9 of the median or mean value calculated by the model. The concentration in water is almost entirely controlled by the hydrolysis half-life in water (HLW) and the concentration in sediment by the HLW and K_{OC} , with the sediment half-life having little influence because it is so long. The variance in overall residence time is also controlled by the HLW. This is due to both the high dispersion ($k = 3.00$) of the HLW and the model sensitivity to this parameter.

For PCB-180 in Lake Pepin dispersion factors of 1.88 and 1.75 are estimated for the concentrations in water and sediment respectively. For Lake Ontario the corresponding factors are 2.21 and 2.04. The residence times have dispersion factors of 3.67 in Lake Pepin and 2.53 in Lake Ontario. In both lakes the concentration in water is most sensitive to the sediment deposition rate. Interestingly, the concentration in Lake Pepin is largely determined by the sediment burial rate while there are several factors of approximately equal influence in Lake Ontario.

Dispersion in output quantities are functions of uncertainty in input parameters and model sensitivity. The details of the RS analyses can be examined to assess the relative importance of each. The OAT analysis described in Section 5.4 also provides a simple and accessible method for evaluating this.

5.4. Uncertainty and sensitivity by OAT inspection

The following is based on the studies by MacLeod et al. (2002) and Slob (1994) on this topic. A simple sensitivity analysis is done first in which a test is made of the magnitude and sign of the sensitivity of each output parameter (y) to each input parameter (x).

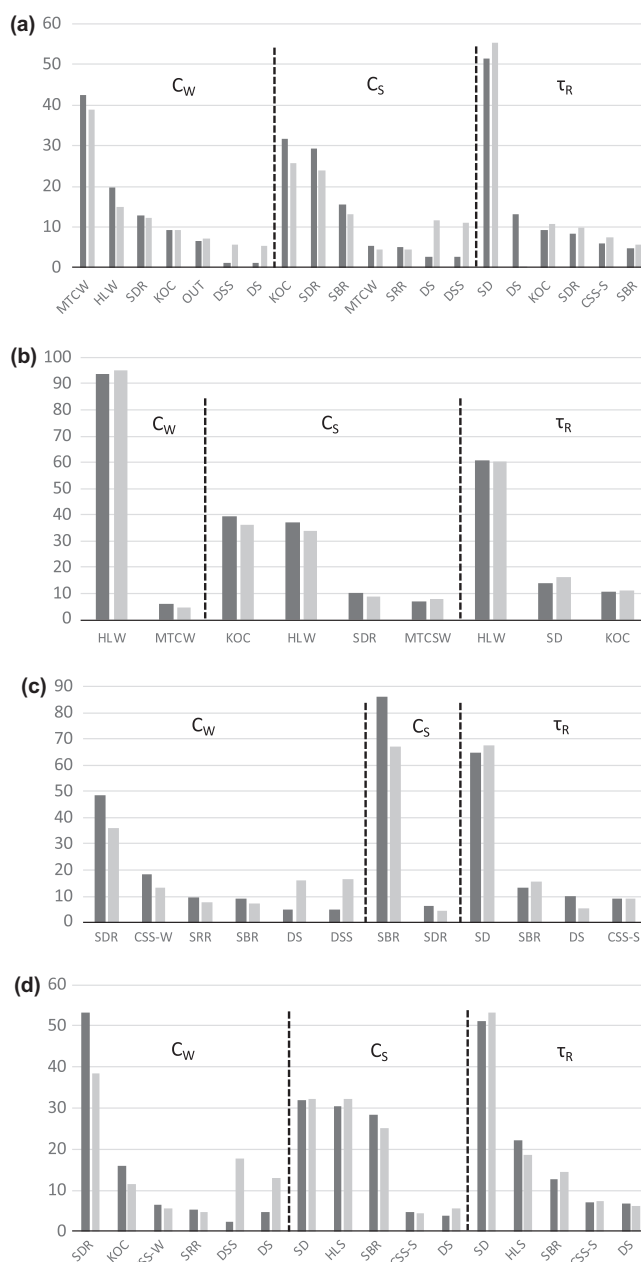


Fig. 5. Percent contribution of input parameters to concentration in water (C_w), concentration in sediment (C_s) and overall residence time (τ_R) for (a) D5 in Lake Pepin, (b) D5 in Lake Ontario, (c) PCB-180 in Lake Pepin and (d) PCB-180 in Lake Ontario. Dark grey bars represent results from the random sampling analysis, light grey those of the OAT analysis.

This yields an x - y sensitivity or S -matrix, each entry of which is the partial derivative of each value of $\ln(y)$ to each $\ln(x)$:

$$S = \delta \ln(y) / \delta \ln(x) = (dy/y) / (dx/x) \sim (\Delta y/y) / (\Delta x/x)$$

where $\Delta x/x$ is a small fractional perturbation in the input parameter and $\Delta y/y$ is the fractional change in the output quantity.

The value of S_{xy} can be negative or positive and its absolute value is usually less than 1.0. It answers the following questions: Does an increase in x cause an increase or decrease in y ? What is the inherent model sensitivity of the column of y values to each x and how do these sensitivities rank?

The natural logarithm of the dispersion factors in the row for each x are then multiplied by each sensitivity S in that row to calculate a matrix of $\ln(k)S$ factors that express the relative magnitude of

the likely dispersion in each y to each x taking into account both the inherent sensitivity and the dispersion in each x . This matrix provides an insight into how dispersion and sensitivity combine to estimate the overall variation in each output quantity.

Finally, a total sensitivity matrix is compiled. The terms in each column of $\ln(k)S$ are squared and totaled. This is equivalent to squaring standard deviations to give variances. Each value of $(\ln(k)S)^2/\Sigma(\ln(k)S)^2$ is calculated to give a fractional or percentage estimate of the individual and total variances. This enables the relative contributions of each x to the likely variances in each y to be compared. The square root of $\Sigma(\ln(k)S)^2$ is a total dispersion for each y and should be similar in magnitude to the value generated by the random sampling analysis. OAT analysis may not give identical contributions because it does not account for interactions between processes, and RS results should be considered more reliable. This method is also suspect for non-linear output quantities such as percent in sediment that are constrained to values less than 100% because the assumption of log-normality does not apply.

6. Discussion

The estimated uncertainties by the two methods are comparable but not identical. Both methods identify the same key influential factors that contribute to the uncertainty but the order of importance is different between the two methods. There are merits to both approaches. The random sampling approach is more transparent and credible and may be preferred for regulatory purposes, it can quantify variance sources from influential factors, and it can better treat constrained quantities such as percentages. The OAT direct inspection approach enables the user to easily probe whether the uncertainty is derived mainly from the inherent sensitivity in the model or from the assigned dispersion factor. This can assist in interpretation of the results and justify the acquisition of more accurate parameter values.

It is hoped that this study will serve as an example for similar studies of the fate of other chemicals in other lakes and thus contribute to the risk assessment. Ultimately there is a compelling incentive to obtain reliable emissions and monitoring data. Models such as QWASI are fundamentally linear in structure, thus the estimated masses, fluxes and concentrations are proportional to the selected emission rate. The importance of obtaining reliable emission rate estimates cannot be over-estimated, especially if comparisons are to be made between model estimates and monitoring data to confirm (or otherwise) that the model assertions are in reasonable accord with reality prior to undertaking a full risk assessment and more extensive monitoring of water, sediments and biota.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.chemosphere.2014.04.033>.

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