Integrated ecological and chemical food web accumulation modeling explains PAH temporal trends during regime shifts in a shallow lake

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Abstract

Shallow lakes can switch suddenly from a turbid situation with high concentrations of phytoplankton and other suspended solids to a vegetated state with clear water, and vice versa. These alternative stable states may have a substantial impact on the fate of hydrophobic organic compounds (HOCs). Models that are fit to simulate impacts from these complex interactions are scarce. We developed a contaminant fate model which is linked to an ecosystem model (PCLake) for shallow lakes. This integrated model was successful in simulating long-term dynamics (1953-2012) of representative polycyclic aromatic hydrocarbons (PAHs) in the main biotic and abiotic components in a large shallow lake (Chaohu in China), which has undergone regime shifts in this period. Historical records from sediment cores were used to evaluate the model. The model revealed that regime shifts in shallow lakes had a strong impact on the fate of less hydrophobic compounds due to the large storage capacity of macrophytes, which accumulated up to 55.6% of phenanthrene in the clear state. The abrupt disappearance of macrophytes after the regime shift resulted in a sudden change in phenanthrene distribution, as the sediment became the major sink. For more hydrophobic compounds such as benzo(a)pyrene, the modeled impact of the regime shift was negligible for the whole environment, yet large for biotic compartments. This study is the first to provide a full mechanistic analysis of the impact of regime shifts on the fate of PAHs in a real lake ecosystem.

Keywords: Fate models; Food web models; Polycyclic aromatic hydrocarbons; Regime shift; Shallow lakes

1. Introduction

Freshwater shallow lake ecosystems often suffer from water quality deterioration due to eutrophication (Conley et al., 2009). The response of these systems to eutrophication is non-linear, reinforced by multiple feedback mechanisms (Carpenter et al., 1999) that lead to alternative stable ecosystem states, i.e. a clear, macrophyte-dominated state, and a turbid, phytoplankton-dominated state (Scheffer et al., 2001). Such systems are stable in either of the states (Scheffer et al., 1993) but can shift from one state to another surprisingly, when a threshold of a certain external condition is crossed (e.g. nutrient loading), which is generally referred to as a ‘regime shift’ (Scheffer and Jeppesen, 2007). Over the last decades, many shallow lakes around the globe have undergone such a regime shift from clear to turbid state mainly due to severe pressure from anthropogenic activities (Kong et al., 2017; Wang et al., 2012).

Meanwhile, freshwater lake ecosystems in highly populated areas are increasingly contaminated with hazardous chemicals such as hydrophobic organic contaminants (HOCs) (Schwarzenbach et al., 2006). These HOCs are distributed among various compartments in the lake environment and accumulate in the aquatic food web, thereby threatening ecological functions due to toxicity to organisms (Qin et al., 2013b), and potentially undermining human health if contaminated aquatic products are consumed (Wu et al., 2007). The coincidence of eutrophication and contamination in shallow lakes calls for a better understanding of the impacts of regime shifts on the behavior and potential risks of HOCs in such aquatic ecosystems. This is a prerequisite for ecological and human health risk assessment, which in turn may inform environmental management and policy. The ecological structures in the alternative states have been shown to be largely different (Kong et al., 2016), while the fate of HOCs in aquatic ecosystems is largely associated with the biogeochemical cycle of carbon

and the dynamics of primary producers (Koelmans et al., 2001; Nizzetto et al., 2012). Consequently, the changes in ecological structure due to the aforementioned regime shifts will drastically alter the fate and bioaccumulation of certain HOCs in lakes. This issue has been addressed by indoor experiments (Roessink et al., 2010) and field studies with several lakes in distinctive ecological states (Moermond et al., 2005), yet illustrative cases of regime shifts within one ecosystem are scarce. This can be explained by lack of long-term observations in lake ecosystems that may have suffered from regime shifts, and by the difficulty in detecting the occurrence of these regime shifts. However, studying regime shifts in one specific system has the advantage that confounding factors due to differences among lakes play no role.

The mechanistic understanding of the implications of regime shifts for HOC fate and effects requires the integration of ecosystem and contaminant fate models (Koelmans et al., 2001). Integrated models can be used to test new hypothesis at the interface of the disciplines of aquatic ecology and environmental chemistry. However, such integrated models are poorly available. Furthermore, there is room for improvement with respect to some of the process descriptions, for instance those capturing the dynamics of the food web (Gouin et al., 2013), and those quantifying sorption to condensed carbon phases (Koelmans et al., 2006; Koelmans et al., 2001), which we aim to address in the present study. Finally, whether such models can adequately represent the implications of a regime shift for the cycling of HOCs in a real system remains to be demonstrated.

We applied the model to polycyclic aromatic hydrocarbons (PAHs), which is a class of HOCs that can exert a wide range of different chemical behaviors (Shen et al., 2013). The objectives of the present study are: 1) to develop an integrated ecological and chemical fate model for PAHs that describes their fate in both abiotic and biotic compartments in a lake

area at short- and long- term temporal scales, which can include regime shifts, and 2) to investigate the impact of regime shifts on the fate of PAHs in shallow lake ecosystems. To achieve these aims, we coupled a food web accumulation model based on existing approaches for accumulation of PAHs in aquatic organisms (Arnot and Gobas, 2004; Campfens and Mackay, 1997; Gandhi et al., 2006), to a previously developed contaminant fate model (Kong et al., 2014), which is applicable to multiple groups of HOCs, like for instance PAHs (Xu et al., 2013). The model was further integrated with a well-calibrated ecosystem model (PCLake) (Kong et al., 2017), which accounted for the ecological processes. To evaluate the model, we used the data from a large shallow lake in China (Lake Chaohu), which had been subject to a regime shift from clear to turbid state over the last several decades (Kong et al., 2017). Contemporary data of PAHs observations in various biotic and abiotic components in Lake Chaohu were available. To further evaluate the model performance in the long-term, we used the information on historical pollution of PAHs over a time span of 60 years from 1953 to 2012, measured in two sediment cores from the lake. To our knowledge, this is the first study that aims at modeling regime shifts in PAH cycling, and that combines historical information and modeling results to illustrate the long-term dynamics of PAHs fate in an aquatic environment.

2. Materials and methods

2.1 Study area

Lake Chaohu (31°25'28"-31°43'28"N, 117°16'54"-117°51'46"E) is the fifth largest freshwater shallow lake in China with a surface area of 760 km² and an average depth of 3 m, located in the most developed region among the lower reaches of the Yangtze River floodplain (Fig. 1). The lake used to provide important ecological services to the catchment

area, e.g. drinking water supply for large cities Hefei and Chaohu. However, the lake ecosystem has undergone regime shifts between clear and turbid states since the 1950s (Kong et al., 2017). The heavy flood occurring in 1954 induced a regime shift into a turbid state in this lake, which shifted back to a clear state before 1960 (Kong et al., 2017). The onset of the sluice in 1963 (Fig. 1), however, tipped the lake into a turbid state again, and increasing nutrient loadings (Kong et al., 2015) triggered the nuisance of phytoplankton blooms since 1980, which has lasted until now. The lake is no longer serving as the drinking water source to adjacent cities. Great effort has been invested into ecological restoration and management of this lake with however limited improvement in lake water quality and ecological status at the current stage (Kong et al., 2017). Meanwhile, the lake has been subjected to severe PAHs pollution (as well as black carbon) due to increasing energy consumption by biomass fuel combustion for heating in rural areas, vehicle emissions in urban areas and coal combustion at nearby power plants in the catchment (Qin et al., 2014). Reduction in PAHs input to the lake in recent years is not likely to happen due to intensified human activities in the lake catchment (Giesy et al., 2016).

2.2 Data Collection

Phenanthrene (Phe), pyrene (Pyr) and benzo(a)pyrene (BaP), were selected to represent low-, moderate- and high-molecular-weight PAHs. Monthly concentrations of these PAHs were available for gaseous, aerosol particles and for suspended solids from May 2010 to April 2011, and aqueous phase from May 2010 to April 2012 (Qin et al., 2013a; Qin et al., 2014), at multiple sampling sites (Fig. 1). Other field data included PAHs in multiple groups of biota for human consumption in 2009 (Qin et al., 2013c) and in phytoplankton in 2012 (provided as Supporting Information). We further collected data from literature, including

PAHs in surface sediment in 2011 (Li et al., 2014) and in soil close to the study site in 2007 (Wang et al., 2010). Additionally, we used PAHs concentrations in vegetation for Lake Taihu in 2014 (Tao et al., 2015) as a proxy for those in Lake Chaohu. This is based on the fact that: 1) both lakes are located in the lower Yangtze River floodplain, where the ecosystems are subjected to very similar (intense) human disruptions and pollutions (Dearing et al., 2012); 2) Both lakes are large shallow lakes with similar eutrophication status since the 1980s (Janssen et al., 2014; Kong et al., 2017); 3) Both lakes are located in the area with similar intensity of PAHs emission (Zhang et al., 2007). Overall, PAH observations were available for model evaluation in all modeled compartments except for zooplankton. For all organisms, data on lipid fractions were available from our previous measurements (Qin et al., 2013c). In addition, PAH profiles were measured in the first 30 cm of two sediment cores (Fig. 1), which were collected with a Kajak gravity corer from the centers of the west and the east part of Lake Chaohu on August 23rd, 2011 (details for PAHs measurement provided as Supporting Information). The time series of historical PAHs records were used for the evaluation of the long-term model simulation. Chronologies were obtained by measuring ²¹⁰Pb and ¹³⁷Cs radionuclide activities in contiguous samples in the cores (details provided as Supporting Information). More details regarding analytical procedures for ²¹⁰Pb and ¹³⁷Cs radionuclide activities in the sediment were reported before (Kong et al., 2017).

2.3 Model development

The model framework is composed of an ecological module (PCLake model) and a contaminant fate model that includes an abiotic module and a biotic (foodweb) module (Fig. 2). Model equations are provided as Supporting Information. Important environmental processes (e.g. water-sediment interaction) and dynamics of the food web components in

Lake Chaohu are simulated using PCLake (Fig. 2). PCLake is a well-developed ecosystem model for shallow lakes in the context of the alternative stable states theory (Janse, 2005). This model has been adapted for Lake Chaohu in our previous study and fitted to field observations (including nutrient levels, water quality indicators and biomass of various biota components) in both short-term (2008-2013) and long-term (1953-2012) simulations (Kong et al., 2017). PCLake has a food web module that is similar to the biotic module in the contaminant fate model in the present study. Therefore, the biomass for different organism groups as simulated by PCLake (Fig. S1) is used as input for the simulations of the food web PAH bioaccumulation model. This is based on the assumption that PAH toxicity had no influence on the biomass and abundance of species, which is reasonable for the present case study because the PAH concentrations (total and individual PAH) in both sediment cores were generally lower than the threshold effect concentration (TEC), and were always below the probable effect concentration (PEC) (MacDonald et al., 2000) (Fig. S2).

For the contaminant fate model, the abiotic module is a fugacity-based level IV fate model for lakes based on previous studies (Kong et al., 2014; Xu et al., 2013), with an additional soil compartment in the lake basin. The water compartment is assumed well-mixed in this study as the lake is shallow with an average depth of only 3 m. Following earlier modeling (Mackay and Paterson, 1991), equilibrium is assumed within each main abiotic compartment. The biotic module is a food web accumulation model (Campfens and Mackay, 1997; Gandhi et al., 2006), with an extended description of the PAH exchange between water and phytoplankton (Fig. 2; SI text). Based on our previous study on the food web structure in Lake Chaohu (Kong et al., 2016), we defined a simplified food web structure for the model in the present study that comprises seven functional groups: piscivorous fish, zooplanktivorous fish, benthivorous fish, zoobenthos, zooplankton, phytoplankton, and macrophytes.

Suspended solids in the water compartment (i.e. detritus) serve as an additional group in the food web. Modeled processes include exchange through gills from water or pore water in sediment (D_W), uptake from food (D_A), loss by fecal egestion (D_E), loss by growth dilution (D_G), loss by metabolism (D_M) and loss by predation (D_P). For the three fish groups, we added the process of production (D_{2f}) representing the influence of fishery. The exchange with water for fish groups is modeled following Arnot and Gobas (2004), which is also applied for invertebrates (zooplankton and zoobenthos). Growth dilution and metabolism for both fish and invertebrates are modeled as first-order kinetic processes, while the PAH metabolic transformation rates are approximated following Moermond et al. (2007). Note that metabolism is found to dominate PAH elimination from fish and invertebrates and is thus important for the trophic transfer of PAHs in aquatic ecosystems (Wan et al., 2007). For modeling PAHs in macrophytes and phytoplankton, we used similar principles as those for fish and invertebrates, with modifications based on the uptake model for phytoplankton from Dachs et al. (1999) (SI text). Bioaccumulation is modeled based on the foodweb interactions (Arnot and Gobas, 2004) and the dietary composition defined in Table S3. Finally, we modeled elimination by egestion as a constant fraction of the uptake from food, quantified as the limiting biomagnification factor (Q) (Campfens and Mackay, 1997).

2.4 Parameter determination

Definition, statistical data and sources for all the parameters in the contaminant fate model are listed in Tables S4-S6. There are 159 parameters in the model, including 46 environmental parameters, 21 chemical-specific parameters (mass transfer and physicochemical parameters) and 92 food web-related parameters. Twenty-one parameters vary seasonally, whereas the other parameters remain constant throughout the simulation. The

parameters were obtained from the relevant literature or calculated based on the conditions in Lake Chaohu (Table S4-S6). Temperature corrections were applied to both subcooled liquid vapor pressure (P_{s25} ; Pa) and Henry's law constant (H_{25} ; Pa·m³/mol) at 25°C (Lun et al., 1998; Paasivirta et al., 1999). Note that vapor pressure for the solid substance of the chemical, which is generally lower than that for the subcooled liquid of the chemical, can lead to an overestimation of the fugacity capacity of aerosol particles and thus to inaccurate predictions (Paasivirta et al., 1999). Sorption of PAH to black carbon (BC) in sediment was taken into account. Following previous approaches (Hauck et al., 2007; Koelmans et al., 2009), a whole-lake median literature value for the fraction of BC in sediment was applied ($f_{BC}=0.002$). Other parameters for strong sorption of PAHs to carbonaceous materials and for metabolic transformation were based on Moermond et al. (2007). We used the bioconcentration factors for phytoplankton's matrix and surface (BCF_M and BCF_S , respectively; m³/kg) reported by Del Vento and Dachs (2002), adjusted by the factors $k_d/(k_d + k_G)$ and $k_{des}/(k_{des} + k_G)$, respectively, to account for the dilution effect of growth (Koelmans, 2014; Koelmans et al., 1995). The parameters k_d (1/h), k_{des} (1/h) and k_G (1/h) are the depuration rate, the desorption rate of surface, and the growth rate of phytoplankton, respectively. Uncertainly exists when these parameters are extrapolated to other phytoplankton species (Del Vento and Dachs, 2002). However, as the experimentally determined values were not available for species in Lake Chaohu, we used literature data (Del Vento and Dachs, 2002). We assumed that species-dependent kinetic factors play a secondary role in determining the fate of POPs in aquatic environments, which has been proved valid for modeling other HOCs (Dachs et al., 1999). For the remainder, the food web accumulation model was parametrized according to previous studies (Arnot and Gobas, 2004; Campfens and Mackay, 1997; Di Paolo et al., 2010). In the present modeling approach, none of the parameters was optimized by fitting to measured data.

2.5 Model simulations

Two simulations were conducted. The short-term simulation covered the period from May 2010 to April 2012. The long-term simulation covered the period from January 1953 to December 2012, i.e. 60 years in total. The short-term simulation aimed at evaluating the robustness of the model and the ability of the model to predict the seasonality of PAH concentrations in Lake Chaohu. The long-term simulation was designed to investigate the impact of the catastrophic regime shift on the fate and dynamics of PAHs in the catchment of Lake Chaohu. Note that our model is 0-dimensional, which suffices for the present study because the lake is assumed to be well-mixed both horizontally and vertically. The external conditions were derived from our previous study (Kong et al., 2017), including water inflow and outflow, water depth, wind speed, water temperature and precipitation. Other boundary conditions such as emission inventories are provided as Supporting Information. For the short-term, initial values were the observations from March 2010 for concentrations in air and water, complemented with the available field observations for the other compartments. For the long-term, however, no data were available before the starting date. We assumed initial values to be two orders of magnitude lower than the average values for 2008-2013 based on the concentrations measured in the deepest (oldest) layers from sediment cores (Fig. S3). The model was implemented in Matlab (MathWorks, 2002). The differential equations were solved using a fourth-order Runge-Kutta method as available in Matlab (ode45), with a simulation time step of 1 hour.

3. Results and discussion

3.1 Model evaluation in the short-term simulation

In the short-term simulation, our yearly (2-year for dissolved phase in water) average results fit well to the measured data for all modeled compartments (Fig. 3); two-thirds of the outcomes with a deviation smaller than a factor of 3, one-sixths with a deviation between a factor 3 and 5, and one sixths with a deviation between a factor 5 and 10. An acceptable deviation between measured and modeled values for contaminant fate models is 0.7 logarithm units or lower, i.e. a factor smaller than 5 (Cowan et al., 1995). Thus, five-sixths of the modeling results fall in the acceptable range. We found that model performance for suspended solids, phytoplankton and macrophytes are relatively less robust (with larger deviation), particularly for Phe and BaP. Moreover, previous studies showed that the underestimation of sorption of PAHs to sediment can be due to the neglect of strong nonlinear sorption to condensed carbonaceous materials such as BC in the sediment (Moermond et al., 2007). Here, the inclusion of sorption to BC significantly improves model performance, particularly for the more hydrophobic PAHs, as the deviation between measured and modeled concentrations of Pyr and BaP in the sediment reduces by approximately one order of magnitude (not shown), to only a factor of 2 (Fig. 3). The importance of BC in modeling PAHs sorption in sediment has been previously demonstrated (Hauck et al., 2007). The typical value of the fraction (0.002) of BC in aquatic sediments, which is applied in the present study, seems to be a reasonable estimation when field data are not available. In addition, it has been shown that metabolic transformation at higher trophic levels may cause trophic dilution of PAHs in aquatic food webs (Wan et al., 2007). Here, the inclusion of metabolic transformation has a significant positive effect on the outputs of PAHs concentrations in fish and invertebrates (not shown), which is crucial to model the bioaccumulation of PAHs in food web (Moermond et al., 2007). Overall, our model reconfirms the importance of incorporating the processes of BC sorption and metabolic

transformation for PAH modelling (Di Paolo et al., 2010; Hauck et al., 2007; Moermond et al., 2007). On a seasonal scale, the model provides a reasonable match with the observations for the three PAHs in the gaseous phase and aerosol particles in air. However, the model shows a relatively limited ability to predict seasonal variations of PAH concentrations in the water column (dissolved and suspended solids) (Fig. S4). More details are provided as Supporting Information. The seasonal validation provides more detail on model performance, detail that cannot be clearly demonstrated from annual average results. For example, better model performance for aerosol particles than for the gas phase can be observed from the seasonal simulation data (Fig. S4), but cannot be easily seen on an annual average scale (Fig. 3). Overall, the results imply that within short temporal scales, the developed model, with all the mechanisms above incorporated, is capable of predicting the magnitude in most compartments (less than 0.7 logarithm units deviation between measured and modeled values for five-sixths of outputs) and seasonal patterns in abiotic compartments (air and water) for the three PAH residual concentrations within Lake Chaohu. The model was subsequently evaluated using long-term data.

3.2 Model evaluation using long-term simulation

The 60-year simulation was evaluated against data using the historical records in the sediment cores (Fig. 4). The model predictions for the sediment show a good match to the long-term historical records, whereas the prediction power in other compartments remains untested. Based on the model predictions, emission in the lake catchment (Fig. S3) drives the accumulation of PAHs in the sediment of Lake Chaohu since the 1950s, which is consistent with the positive correlations between PAH emissions and residue levels in sediments of Lake Chaohu (Ren et al., 2015). However, the model tends to overestimate the concentrations of

Phe before 1990 (Fig. 4A). This discrepancy may be attributed to the underestimation of biodegradation of low-molecular-weight PAHs, the effect of which may be much stronger in earlier time. The general decreasing trend of PAHs in sediment after the 2000s is in line with the emission data (Fig. S3), which was however not fully captured in the model outputs. Multiple factors may contribute to this deviation, such as the absence of long-term data regarding BC contents in the sediment. A better model prediction may be achieved by using time explicit data of BC content in sediment cores. This BC content may have decreased after the 2000s due to a higher energy use efficiency (Wang et al., 2014) and enhanced burial. Nonetheless, the general agreement between measured and modeled PAH concentrations in sediment with respect to both magnitude and temporal dynamics implies that long-term model simulation outputs can be used for further evaluation.

3.3 Impact of the regime shift on the modeled fate of PAHs

The regime shift in this lake was characterized by a loss of macrophytes, rapidly developing blooms of phytoplankton, a switch in fish community towards domination of small zooplanktivorous fish, a strong enhancement in sediment resuspension and a substantial increase in suspended solid concentration (Fig. S1). The lake state dynamics are nicely represented by the vegetation coverage from the PCLake model (Fig. 5A), which agrees well with the field data (Kong et al., 2017).

Long-term dynamics of the mass distribution for the three modeled PAHs among various compartments are simulated for the same period (Fig. 5). In general, our modeling results demonstrate that the mass distribution of less hydrophobic PAH in the lake is more susceptible to changes in ecological structure than that of more hydrophobic PAH (Fig. 5B-D). During the clear state (1958-1962), the model shows that a large fraction of Phe in the

lake area is distributed in the food web (55.6% on average), while the fractions are much lower for Pyr and BaP (2.6% and 0.3% on average, respectively). On the other hand, during the turbid state after the regime shift (1963-2012), the corresponding average values become 0.050%, 0.010% and 0.008% for Phe, Pyr and BaP, respectively, and the dominant sinks of the three modeled PAHs in the total environment of the lake catchment are soil and sediment. This finding agrees with results from indoor mesocosm experiments (Roessink et al., 2010), which show that the less hydrophobic and more mobile HOCs are more susceptible to ecological changes than the more hydrophobic and less mobile HOCs.

The model further indicates that the higher susceptibility to ecological changes for less hydrophobic chemicals is attributable to their higher tendency to get absorbed by macrophytes (Fig. 5B) before they get a chance of being adsorbed to BC. The model confirms that macrophytes are one of the dominant sinks for Phe in addition to soil and sediment in a clear lake basin, which is quantitatively in accordance with earlier results from laboratory mesocosm experiments mimicking conditions of shallow lakes (Roessink et al., 2010). In addition, the fractions of total mass in macrophytes for three modeled PAHs decrease drastically with increasing molecular weight (Fig. 5B-D), resulting in a lower susceptibility to ecological changes for Pyr and BaP. Both Pyr and BaP are primarily bound in soil and sediment rather than in the macrophytes because of their stronger sorption to BC than that of Phe (Koelmans et al., 2006). Therefore, different sorption abilities to BC of chemicals determine their behaviors during regime shifts. More hydrophobic PAHs may be bound primarily to soil and sediment due to stronger BC sorption, thereby being less affected by a regime shift within biotic compartments.

As for modeling PAH bioconcentration by macrophytes, our model may leave some room for improvement. Based on previous modeling work (Janse, 2005), the equation for

macrophytes (see the Supporting Information) describes the exchange of substances with water and sediment separately, whereas it ignores the transport process between root and shoot, and it considers the chemicals to be evenly distributed in the macrophyte biomass. However, recent work has shown that chemicals were more slowly translocated from root to shoot than the other way around (Diepens et al., 2014). Consequently, including heterogeneity within the macrophytes and accounting for translocation between shoot and root in our model, may further increase ecological realism and provide more accurate simulations, especially when partitioning to macrophytes is important like during the clear state before 1963 (Fig. 5). However, parameters regarding uptake and elimination kinetics of PAH in sediment-rooted macrophytes are still uncertain and scarce (Diepens et al., 2014). More field data, as well as a more comprehensive submodel for macrophytes involving heterogeneity, are expected in future research.

The capacity of macrophytes to store considerable amounts of PAHs, i.e. ‘biomass dilution’ (Roessink et al., 2010), results in the depletion of PAHs in other environmental compartments, most importantly in biota. The model shows that in a clear lake, macrophytes account for 99.7% of Phe mass, 99.2% of Pyr mass and 83.1% of BaP mass in the water column above the sediment (Fig. 5E). Further indoor experiments have focused on investigating the potential of certain macrophytes to become a major storage reservoir for HOCs (Schneider and Nizzetto, 2012). In addition, macrophytes are modeled to stabilize the sediment, thereby reducing resuspension fluxes due to fish bioturbation and wind shear stress in shallow lakes (Janse, 2005). Consequently, the concentrations of suspended solids and PAH in the dissolved phase are lower with macrophytes present, due to lower desorption of PAHs from the suspended solids and to higher accumulation in macrophytes. We can infer that ecological restoration of a turbid lake back to a clear, macrophyte-rich state will not only

improve water quality, ecological functioning and services, but also lead to the redistribution of PAHs in the lake ecosystem where higher proportions of PAHs will end up in macrophytes. As a consequence, PAH concentrations in suspended solids and biota compartments, as well as the toxic effects and ecological risks of these contaminants, may be reduced on an annual scale.

After the lake ecosystem tips into a turbid state, the dominant sinks of the three modeled PAHs are primarily soil and sediment, whereas phytoplankton being the primary producer accumulates only a negligible fraction of PAH mass in the whole environment (Fig. 5B-D). The predominant roles of soil and sediment to determine PAH distribution in an aquatic ecosystem are in agreement to earlier studies (Liu et al., 2007; Roessink et al., 2010). However, within the water column above the sediment (the soil and sediment compartments were both excluded), the model reveals the significant role of phytoplankton in the distribution of PAHs, particularly for more hydrophobic chemicals such as Pyr and BaP, because 39.7% of Pyr mass and 83.8% of BaP mass are associated with the phytoplankton (Fig. 5F). A similar pattern was observed in an earlier laboratory study, in which periphyton dominated the mass distribution of PAHs in indoor model ecosystems when macrophytes were not present (Roessink et al., 2010). This effect also has been recognized for pelagic ecosystems (Nizzetto et al., 2012), in which phytoplankton is referred to as the ‘biological pump’ (Jurado and Dachs, 2008). PCLake shows that biomass of phytoplankton is much higher without macrophytes (Fig. S1), primarily due to the increase of available nutrients from the dead macrophytes and the destabilized sediment. The rapidly increased phytoplankton biomass may bind large amounts of PAHs from the dissolved phase, which may promote the transfer of PAH from the atmosphere to the water as was suggested before (Dachs et al., 1999). In addition, the mortality of phytoplankton may enhance the vertical flux

to sediment, whereas the absence of macrophytes may also result in a higher resuspension intensity. Consequently, the interaction between water and sediment possibly is much stronger in turbid than in clear lakes, which may enlarge the pool of sediment acting as a sink of PAHs. Our study reveals that the capacity of phytoplankton as a storage reservoir of PAHs is much lower than that of macrophytes, but the influence of phytoplankton on the fate of PAHs in the components of the food web and the aqueous phase can be substantial.

3.4 Integrated modeling approach: merits and limitations

The model developed in the present study has several advantages over earlier models. By forcing the contaminant fate model with outputs from the PCLake ecosystem model, our model accounts for processes including organic carbon cycling, transport and accumulation of PAHs in the foodweb, and limnologic processes such as bottom up and top down control, which are essential to evaluate the fate of contaminants in lake ecosystems with changing nutrient loading (Koelmans et al., 2001). In addition, long-term and intensive time series data from field observations in lake ecosystems are usually scarce, particularly those covering a time span where regime shifts occur. Complex aquatic ecosystem models describing the main biotic and abiotic components, such as PCLake, can be considered as a ‘virtual mesocosm’, the output of which can subsequently serve as the supplement of sparse field observations. Our model may provide benefits for lake management, for instance to base a trigger for sediment PAH remediation on anticipated consequences of abatement of eutrophication, which in part have been covered at length in earlier literature (Koelmans et al., 2001).

The toxic effect of PAHs on organisms was assumed to be negligible here, because observed PAH concentrations are below the PEC levels (Fig. S2). Here the thresholds for PAHs (TEC and PEC) are both consensus-based sediment quality guidelines for freshwater

ecosystems (MacDonald et al., 2000). This, however, may not be true in other cases. After all, PCLake predictions without accounting for the toxic effects may be biased because toxicity may affect abundances of certain sensitive species (e.g. arthropods), which play key roles in food web interactions (Koelmans et al., 2001). Including the toxic effect of PAHs in the model would permit to investigate if high PAHs concentrations could trigger regime shifts on the ecosystem level. Like all other ecological stressors, toxicity of HOCs is known to cause gradual changes on the level of individuals (e.g. impairment of individuals), populations and communities (e.g. the abundance and diversity of species) (Diepens et al., 2016). Several studies have further indicated adverse effects of chemicals, e.g. tributyltin and organochlorine pesticides, that break down the feedback mechanisms that promote the dominance of macrophytes (Sayer et al., 2006; Stansfield et al., 1989). The possibility of HOCs to cause regime shifts in lakes may largely depend on the systems' stability, i.e., how far the system is from the tipping point, which is generally determined by multiple factors, such as food web interaction, trophic state and chemical pollution.

4. Conclusion

We provided an integrated ecological and BC-inclusive chemical transport and food web accumulation model, which predicted concentrations of three different PAH compounds that are consistent with short-term and long-term measured data. Model simulations revealed a full picture of the impact of demonstrated regime shifts in the shallow lake ecosystem on the fate of the PAHs. Also for the first time, historical records from sediment cores were used for model evaluation, serving as an alternative way to compensate data deficiency.

The model revealed that regime shifts in shallow lakes have a stronger impact on the fate of less hydrophobic compounds than on more hydrophobic compounds due to the large

storage capacity of macrophytes, which in the studied case accumulated up to 55.6% of Phe in the clear state. The abrupt disappearance of macrophytes after the regime shift resulted in a sudden change in Phe distribution. For more hydrophobic compounds such as BaP, the impact of the regime shift was negligible for the whole environment, yet large for biotic components. The present study demonstrates how integrated modeling can assist in identifying the central roles of both abiotic components (soil and sediment) and biota at the base of the food web (phytoplankton and macrophytes) in driving the distribution of PAHs in shallow lakes.

The present results may have implications for lake management, as many shallow lakes around the globe are simultaneously polluted by excess nutrient loads and HOCs such as PAHs. A better understanding of their interactions may enhance our prediction power on the fate of HOCs, which in turn may facilitate the development of sound lake management strategies.

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Appendix A. Supplementary data

Additional Information includes: SI Text for model equations, description of modelling water-phytoplankton interactions, methods for PAHs measurement in phytoplankton and sediment cores, methods for chronologies determination in sediment cores, boundary conditions, and model evaluation of seasonal patterns in the short-term simulation. SI Figures include Fig. S1-S5. SI Tables include Table S1-S8.

References

- Arnot, J.A., Gobas, F. (2004) A food web bioaccumulation model for organic chemicals in aquatic ecosystems. *Environmental Toxicology and Chemistry* 23(10), 2343-2355.
- Campfens, J., Mackay, D. (1997) Fugacity-based model of PCB bioaccumulation in complex aquatic food webs. *Environmental Science & Technology* 31(2), 577-583.
- Carpenter, S.R., Ludwig, D., Brock, W.A. (1999) Management of eutrophication for lakes subject to potentially irreversible change. *Ecological Applications* 9(3), 751-771.
- Conley, D.J., Paerl, H.W., Howarth, R.W., Boesch, D.F., Seitzinger, S.P., Havens, K.E., Lancelot, C., Likens, G.E. (2009) Controlling eutrophication: nitrogen and phosphorus. *Science* 323(5917), 1014-1015.
- Cowan, C.E., Mackay, D., Feijtel, T.C.J., van de Meent, D., Di Guardo, A., Davies, J., Mackay, N. (1995) *The multi-media fate model: a vital tool for predicting the fate of chemicals*, SETAC Press, Pensacola, FL.
- Dachs, J., Eisenreich, S.J., Baker, J.E., Ko, F.C., Jeremiason, J.D. (1999) Coupling of phytoplankton uptake and air-water exchange of persistent organic pollutants. *Environmental Science & Technology* 33(20), 3653-3660.
- Dearing, J.A., Yang, X.D., Dong, X.H., Zhang, E.L., Chen, X., Langdon, P.G., Zhang, K., Zhang, W.G., Dawson, T.P. (2012) Extending the timescale and range of ecosystem services through paleoenvironmental analyses, exemplified in the lower Yangtze basin. *Proceedings of the National Academy of Sciences, USA* 109(18), E1111-E1120.
- Del Vento, S., Dachs, J. (2002) Prediction of uptake dynamics of persistent organic pollutants by bacteria and phytoplankton. *Environmental Toxicology and Chemistry* 21(10), 2099-2107.

507 Di Paolo, C., Gandhi, N., Bhavsar, S.P., Van den Heuvel-Greve, M., Koelmans, A.A. (2010)
508 Black carbon inclusive multichemical modeling of PBDE and PCB biomagnification and
509 transformation in estuarine food webs. *Environmental Science & Technology* 44(19), 7548-
510 7554.

511 Diepens, N.J., Arts, G.H.P., Focks, A., Koelmans, A.A. (2014) Uptake, translocation, and
512 elimination in sediment-rooted macrophytes: A model-supported analysis of whole sediment
513 test data. *Environmental Science & Technology* 48(20), 12344-12353.

514 Diepens, N.J., Beltman, W.H., Koelmans, A.A., Van den Brink, P.J., Baveco, J.M. (2016)
515 Dynamics and recovery of a sediment-exposed *Chironomus riparius* population: A modelling
516 approach. *Environmental Pollution* 213, 741-750.

517 Gandhi, N., Bhavsar, S.P., Gewurtz, S.B., Diamond, M.L., Evenset, A., Christensen, G.N.,
518 Gregor, D. (2006) Development of a multichemical food web model: Application to PBDEs
519 in Lake Ellasjøen, Bear Island, Norway. *Environmental Science & Technology* 40(15), 4714-
520 4721.

521 Giesy, J.P., Tang, Z., Zhao, X. (2016) Historical record of effects of human activities on
522 absolute and relative concentrations of Polycyclic aromatic hydrocarbons (PAHs) in Lake
523 Chao, China. *Journal of Environmental Sciences* 46(8), 1-4.

524 Gouin, T., Armitage, J.M., Cousins, I.T., Muir, D.C., Ng, C.A., Reid, L., Tao, S. (2013)
525 Influence of global climate change on chemical fate and bioaccumulation: The role of
526 multimedia models. *Environmental Toxicology and Chemistry* 32(1), 20-31.

527 Hauck, M., Huijbregts, M.A.J., Koelmans, A.A., Moermond, C.T.A., van den Heuvel-Greve,
528 M.J., Veltman, K., Hendriks, A.J., Vethaak, A.D. (2007) Including sorption to black carbon in
529 modeling bioaccumulation of polycyclic aromatic hydrocarbons: Uncertainty analysis and
530 comparison to field data. *Environmental Science & Technology* 41(8), 2738-2744.

531 Janse, J.H. (2005) Model studies on the eutrophication of shallow lakes and ditches [Doctoral
532 dissertation]. Wageningen University, Wageningen, The Netherlands.

533 Janssen, A.B., Teurlincx, S., An, S., Janse, J.H., Paerl, H.W., Mooij, W.M. (2014) Alternative
534 stable states in large shallow lakes? *Journal of Great Lakes Research* 40(4), 813-826.

535 Jurado, E., Dachs, J. (2008) Seasonality in the "grasshopping" and atmospheric residence
536 times of persistent organic pollutants over the oceans. *Geophysical Research Letters* 35(17),
537 DOI: 10.1029/2008gl034698.

538 Koelmans, A.A. (2014) Limited reversibility of bioconcentration of hydrophobic organic
539 chemicals in phytoplankton. *Environmental Science & Technology* 48(13), 7341-7348.

540 Koelmans, A.A., Anzion, S.P.M., Lijklema, L. (1995) Dynamics of organic micropollutant
541 biosorption to cyanobacteria and detritus. *Environmental Science & Technology* 29(4), 933-
542 940.

543 Koelmans, A.A., Jonker, M.T.O., Cornelissen, G., Bucheli, T.D., Van Noort, P.C.M.,
544 Gustafsson, O. (2006) Black carbon: The reverse of its dark side. *Chemosphere* 63(3), 365-
545 377.

546 Koelmans, A.A., Kaag, K., Sneekes, A., Peeters, E.T.H.M. (2009) Triple domain in situ
547 sorption modeling of organochlorine pesticides, polychlorobiphenyls, polyaromatic
548 hydrocarbons, polychlorinated dibenzo-p-dioxins, and polychlorinated dibenzofurans in
549 aquatic sediments. *Environmental Science & Technology* 43(23), 8847-8853.

550 Koelmans, A.A., Van der Heijde, A., Knijff, L.M., Aalderink, R.H. (2001) Integrated
551 modelling of eutrophication and organic contaminant fate & effects in aquatic ecosystems. A
552 review. *Water Research* 35(15), 3517-3536.

553 Kong, X., Dong, L., He, W., Wang, Q., Mooij, W.M., Xu, F. (2015) Estimation of the long-
554 term nutrient budget and thresholds of regime shift for a large shallow lake in China.
555 *Ecological Indicators* 52, 231-244.

556 Kong, X., He, Q., Yang, B., He, W., Xu, F., Janssen, A.B.G., Kuiper, J.J., van Gerven, L.P.A.,
557 Qin, N., Jiang, Y., Liu, W., Yang, C., Bai, Z., Zhang, M., Kong, F., Janse, J.H., Mooij, W.M.
558 (2017) Hydrological regulation drives regime shifts: evidence from paleolimnology and
559 ecosystem modelling of a large shallow Chinese lake. *Global Change Biology* 23(2), 737-754.

560 Kong, X., He, W., Liu, W., Yang, B., Xu, F., Jørgensen, S.E., Mooij, W.M. (2016) Changes in
561 food web structure and ecosystem functioning of a large shallow Chinese lake during 1950s,
562 1980s and 2000s. *Ecological Modelling* 319, 31-41.

563 Kong, X., He, W., Qin, N., He, Q., Yang, B., Ouyang, H., Wang, Q., Yang, C., Jiang, Y., Xu, F.
564 (2014) Modeling the multimedia fate dynamics of γ -hexachlorocyclohexane in a large
565 Chinese lake. *Ecological Indicators* 41, 65-74.

566 Li, C., Huo, S., Yu, Z., Xi, B., Zeng, X., Wu, F. (2014) Spatial distribution, potential risk
567 assessment, and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in
568 sediments of Lake Chaohu, China. *Environmental Science and Pollution Research* 21(20),
569 12028-12039.

570 Liu, Z., Quan, X., Yang, F. (2007) Long-term fate of three hexachlorocyclohexanes in the
571 lower reach of Liao River basin: Dynamic mass budgets and pathways. *Chemosphere* 69(7),
572 1159-1165.

573 Lun, R., Lee, K., De Marco, L., Nalewajko, C., Mackay, D. (1998) A model of the fate of

574 polycyclic aromatic hydrocarbons in the Saguenay Fjord, Canada. *Environmental Toxicology*
575 *and Chemistry* 17(2), 333-341.

576 MacDonald, D.D., Ingersoll, C.G., Berger, T. (2000) Development and evaluation of
577 consensus-based sediment quality guidelines for freshwater ecosystems. *Archives of*
578 *Environmental Contamination and Toxicology* 39(1), 20-31.

579 Mackay, D., Paterson, S. (1991) Evaluating the multimedia fate of organic-chemicals - a
580 Level-III fugacity model. *Environmental Science & Technology* 25(3), 427-436.

581 MathWorks (2002) Using MATLAB version 6; The MathWorks, Inc.: Natick, MA, US.

582 Moermond, C.T.A., Traas, T.P., Roessink, I., Veltman, K., Hendriks, A.J., Koelmans, A.A.
583 (2007) Modeling decreased food chain accumulation of PAHs due to strong sorption to
584 carbonaceous materials and metabolic transformation. *Environmental Science & Technology*
585 41(17), 6185-6191.

586 Moermond, C.T.A., Zwolsman, J.J.G., Koelmans, A.A. (2005) Black carbon and ecological
587 factors affect in situ biota to sediment accumulation factors for hydrophobic organic
588 compounds in flood plain lakes. *Environmental Science & Technology* 39(9), 3101-3109.

589 Nizzetto, L., Gioia, R., Li, J., Borga, K., Pomati, F., Bettinetti, R., Dachs, J., Jones, K.C.
590 (2012) Biological pump control of the fate and distribution of hydrophobic organic pollutants
591 in water and plankton. *Environmental Science & Technology* 46(6), 3204-3211.

592 Paasivirta, J., Sinkkonen, S., Mikkelsen, P., Rantio, T., Wania, F. (1999) Estimation of vapor
593 pressures, solubilities and Henry's law constants of selected persistent organic pollutants as
594 functions of temperature. *Chemosphere* 39(5), 811-832.

595 Qin, N., He, W., Kong, X., Liu, W., He, Q., Yang, B., Ouyang, H., Wang, Q., Xu, F. (2013a)
596 Atmospheric partitioning and the air–water exchange of polycyclic aromatic hydrocarbons in
597 a large shallow Chinese lake (Lake Chaohu). *Chemosphere* 93(9), 1685-1693.

598 Qin, N., He, W., Kong, X., Liu, W., He, Q., Yang, B., Ouyang, H., Wang, Q., Xu, F. (2013b)
599 Ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in the water from a
600 large Chinese lake based on multiple indicators. *Ecological Indicators* 24, 599-608.

601 Qin, N., He, W., Kong, X., Liu, W., He, Q., Yang, B., Wang, Q., Yang, C., Jiang, Y., Jorgensen,
602 S.E. (2014) Distribution, partitioning and sources of polycyclic aromatic hydrocarbons in the
603 water–SPM–sediment system of Lake Chaohu, China. *Science of the Total Environment* 496,
604 414-423.

605 Qin, N., He, W., Wang, Y., He, Q., Kong, X., Ouyang, H., Xu, F. (2013c) Residues and health
606 risk of polycyclic aromatic hydrocarbons in the water and aquatic products from Lake

607 Chaohu. *Acta Scientiae Circumstantiae* 33(1), 230-239 (in Chinese).

608 Ren, C., Wu, Y., Zhang, S., Wu, L., Liang, X., Chen, T., Zhu, C., Sojinu, S.O., Wang, J. (2015)
609 PAHs in sediment cores at main river estuaries of Chaohu Lake: implication for the change of
610 local anthropogenic activities. *Environmental Science and Pollution Research* 22(3), 1687-
611 1696.

612 Roessink, I., Moermond, C.T.A., Gillissen, F., Koelmans, A.A. (2010) Impacts of
613 manipulated regime shifts in shallow lake model ecosystems on the fate of hydrophobic
614 organic compounds. *Water Research* 44(20), 6153-6163.

615 Sayer, C.D., Hoare, D.J., Simpson, G.L., Henderson, A.C., Liptrot, E.R., Jackson, M.J.,
616 Appleby, P.G., Boyle, J.F., Jones, J.I., Waldock, M.J. (2006) TBT causes regime shift in
617 shallow lakes. *Environmental Science & Technology* 40(17), 5269-5275.

618 Scheffer, M., Carpenter, S., Foley, J.A., Folke, C., Walker, B. (2001) Catastrophic shifts in
619 ecosystems. *Nature* 413(6856), 591-596.

620 Scheffer, M., Hosper, S.H., Meijer, M.L., Moss, B., Jeppesen, E. (1993) Alternative equilibria
621 in shallow lakes. *Trends in Ecology & Evolution* 8(8), 275-279.

622 Scheffer, M., Jeppesen, E. (2007) Regime shifts in shallow lakes. *Ecosystems* 10(1), 1-3.

623 Schneider, S.C., Nizzetto, L. (2012) Bioconcentration and intracellular storage of
624 hexachlorobenzene in charophytes and their potential role in monitoring and remediation
625 actions. *Environmental Science & Technology* 46(22), 12427-12434.

626 Schwarzenbach, R.P., Escher, B.I., Fenner, K., Hofstetter, T.B., Johnson, C.A., Von Gunten,
627 U., Wehrli, B. (2006) The challenge of micropollutants in aquatic systems. *Science* 313(5790),
628 1072-1077.

629 Shen, H., Huang, Y., Wang, R., Zhu, D., Li, W., Shen, G., Wang, B., Zhang, Y., Chen, Y., Lu,
630 Y. (2013) Global atmospheric emissions of polycyclic aromatic hydrocarbons from 1960 to
631 2008 and future predictions. *Environmental Science & Technology* 47(12), 6415-6424.

632 Stansfield, J., Moss, B., Irvine, K. (1989) The loss of submerged plants with eutrophication
633 III. Potential role of organochlorine pesticides: a palaeoecological study. *Freshwater Biology*
634 22(1), 109-132.

635 Tao, Y., Xue, B., Yao, S. (2015) Using linoleic Acid embedded cellulose acetate membranes
636 to in situ monitor polycyclic aromatic hydrocarbons in lakes and predict their bioavailability
637 to submerged macrophytes. *Environmental Science & Technology* 49(10), 6077-6084.

638 Wan, Y., Jin, X., Hu, J., Jin, F. (2007) Trophic dilution of polycyclic aromatic hydrocarbons

(PAHs) in a marine food web from Bohai Bay, North China. *Environmental Science & Technology* 41(9), 3109-3114.

Wang, R., Dearing, J.A., Langdon, P.G., Zhang, E., Yang, X., Dakos, V., Scheffer, M. (2012) Flickering gives early warning signals of a critical transition to a eutrophic lake state. *Nature* 492(7429), 419-422.

Wang, R., Liu, G., Chou, C., Liu, J., Zhang, J. (2010) Environmental assessment of PAHs in soils around the Anhui Coal District, China. *Archives of Environmental Contamination and Toxicology* 59(1), 62-70.

Wang, R., Tao, S., Shen, H., Huang, Y., Chen, H., Balkanski, Y., Boucher, O., Ciais, P., Shen, G., Li, W. (2014) Time trend of global black carbon emissions from 1960 to 2007. *Environ. Sci. & Technol.* 48(12), 6780-6787.

Wu, N., Herrmann, T., Paepke, O., Tickner, J., Hale, R., Harvey, E., La Guardia, M., McClean, M.D., Webster, T.F. (2007) Human exposure to PBDEs: associations of PBDE body burdens with food consumption and house dust concentrations. *Environmental Science & Technology* 41(5), 1584-1589.

Xu, F., Qin, N., Zhu, Y., He, W., Kong, X., Barbour, M.T., He, Q., Wang, Y., Ouyang, H., Tao, S. (2013) Multimedia fate modeling of polycyclic aromatic hydrocarbons (PAHs) in Lake Small Baiyangdian, Northern China. *Ecological Modelling* 252, 246-257.

Zhang, Y., Tao, S., Cao, J., Coveney, R.M., Jr. (2007) Emission of polycyclic aromatic hydrocarbons in China by county. *Environmental Science & Technology* 41(3), 683-687.

Figure captions:

Figure 1. Lake Chaohu and the catchment area in China, as well as the sampling sites for two gaseous and aerosol particles samples (Qin et al., 2013a), four water and suspended solid samples (Qin et al., 2014) and two sediment core samples (Kong et al., 2017). The locations of major rivers and the sluice built in 1963 are also shown.

Figure 2. Overview of the model. (A) Processes between major compartments of the contaminant fate model. (B) Structure of the food web accumulation model, including piscivorous fish (*PF*), zooplanktivorous fish (*ZF*), benthivorous fish (*BF*), zoobenthos (*Zb*), zooplankton (*Zp*), phytoplankton (*P*), submerged macrophytes (*SP*) and detritus (*D*). (C) Processes between water and phytoplankton/suspended solids in the model. (D) Schematic framework of shallow lake ecosystem model PCLake (redrawn after Janse, 2005).

Figure 3. Comparison of measured and modeled concentrations (on a logarithmic scale) of Phe (A), Pyr (B) and BaP (C) in all the model compartments averaged over the short-term simulation period. Error bars for measured data relate to standard deviation (s.d.) of monthly observations on different sample sites (May 2010 to April 2012 for water phase, and May 2010 to April 2011 for air, aerosol particles and suspended solid), or s.d. obtained from literature (other compartments). Error bars for modeled data relate to s.d. of simulation outputs. Observations in zooplankton are not available.

Figure 4. Comparison of measured and modeled concentrations of Phe (A), Pyr (B) and BaP (C) in sediment solids for the long term simulation (January 1953 to December 2012). Measured data are the average values from the two sediment cores, and modeled data are

plotted as the annual average values of each year.

Figure 5. Long term model simulations from 1953 to 2012. (A) Vegetation coverage from field observations (red dots) and PCLake model simulation (black line) (from Kong et al., 2017), which indicates the lake state (clear or turbid). (B-D) Modeled chemical mass fractions for Phe (B), Pyr (C) and BaP (D) among various compartments in the lake area. (E and F) Modeled average relative mass distributions of Phe, Pyr and BaP in water, suspended solids and all the biota compartments in the food web during the periods of clear (1958-1962; E) and turbid (1980-2012; F) states (note that soil and sediment are not included).

Method

Question

Figure 1

Figure 2

Contaminant fate model

Foodweb accumulation model

Lake ecosystem & Food web model

Figure 3

Figure 4

Figure 5