



Using a dynamic mass balance model to predict fate and transport of PCBs in a polluted boreal lake in Sweden

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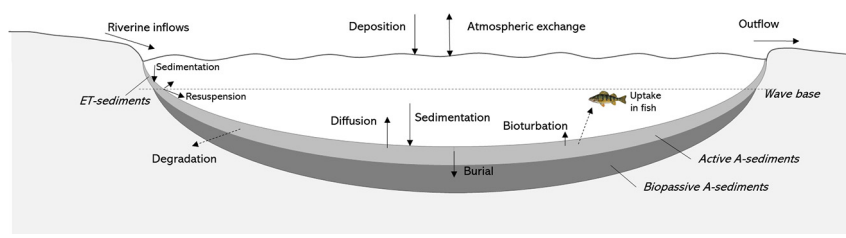


HIGHLIGHTS

- PCBs in different aquatic matrices in a severely contaminated lake system are presented.
- A unique dataset of PCBs was used to test and develop a mathematical model for PCBs.
- The model was used to identify crucial transport processes for PCBs.
- Long-term recovery of PCBs was modeled for three remediation scenarios.

GRAPHICAL ABSTRACT

Dynamic mass balance model to predict fate and transports of PCBs



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ABSTRACT

In 2013, a screening survey including fish (European perch, *Perca fluviatilis*) from 20 locations in the Stockholm region of Sweden indicated exceptionally high levels of PCBs (>450 ng ΣPCB₇/g ww) in Lake Oxundasjön. An extensive sampling program was launched to define the magnitude and area of impact of PCBs. Moreover, a dynamic mass balance model approach was applied to identify and quantify key transport processes and predict the long-term turnover of PCBs given various remediation scenarios.

Based on the dating of sediment profiles, primary emissions of PCBs to Lake Oxundasjön have likely occurred from the end of the 1940s until 1980, reaching the lake via one of its tributaries. Presently, the main source of PCBs is diffusion from the lake sediments. From the lake outlet, >400 g ΣPCB₇/yr are transported to Lake Mälaren (the third largest lake in Sweden), supplying drinking water for parts of the Stockholm area. Remediation actions are necessary to reduce the PCB levels in fish below today's marketing limits and environmental quality standards. With natural recovery, our results indicate that the PCB levels in non-migratory fish from Lake Oxundasjön will be elevated for decades to come. The mass of PCBs stored in the lake sediments was estimated, and to our knowledge, Lake Oxundasjön is the most heavily PCB contaminated lake in Sweden. The system constitutes a unique opportunity to test and develop a mathematical mass balance model for PCBs, with substantial data acquired from different aquatic matrices. The model presented in the paper is applicable for risk assessments of PCBs, and the results contribute to the general understanding of the transport and turnover dynamics of PCBs in aquatic ecosystems.

1. Introduction

Since the mid-1960s, when polychlorinated biphenyls (PCBs) were detected in environmental matrices (Jensen, 1966, 1972), the substance group has been a subject for extensive environmental research. PCBs is a

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group of synthetic organic substances, theoretically 209, manufactured as industrial chemicals mainly in the 1930–1980s. The industrial use was wide, and PCBs was included in, e.g., heat exchange fluids, electric transformers and capacitors, additives in paints, carbonless copy paper, and plasticizers. Global production of PCBs most likely peaked in 1970, and then presumably ceased in 1993 (Breivik et al., 2002; AMAP, 2000). Due to its persistent properties, PCBs is still found in considerable amounts in the environment and remote areas such as the Arctic (AMAP, 1998). PCBs bioaccumulates in food chains due to their persistency and lipophilic properties. Consequently, fractions found in water are small (Josefsson et al., 2011). PCBs has been found in human tissue, blood, and breast milk, where consumption of meat, fish, and dairy products as well as PCBs in air constitutes important exposure routes (Van den Berg et al., 2006; Lehmann et al., 2015; Wang et al., 2016).

The first large-scale sediment remediation of PCBs in Sweden was conducted in Lake Järnsjön in 1993–1994, where about 400 kg was found in lake sediments originating from paper mills upstream of the lake (Larsson et al., 1990). World-wide, a great number of PCB-contaminated sites have been remediated during the last four decades. One example is the PCB-contamination of the Hudson River in the United States, contaminated with PCBs originating from two capacitor manufacturing plants (Thomann et al., 1991). Extensive remediation actions in the river, including sediment removal, have continued into the twenty-first century (Chitsaz et al., 2020). Environmental monitoring can be combined with mass balance modeling to investigate the fate of chemicals in environmental systems and identify important transport processes. The literature available on modeling of PCBs in aquatic environments are extensive (e.g., Thomann et al., 1991; Kim et al., 2017; Guo et al., 2018; Sun et al., 2018).

In a study of a wide range of contaminants in non-migratory fish in the Stockholm region in Sweden (Karlsson and Viktor, 2014), exceptionally high PCB levels was found in Lake Oxundasjön, a boreal low-land lake some 25 km north of Stockholm. To find the cause of the severe pollution of Lake Oxundasjön, extensive investigations on PCBs in different matrices, including sediment, fish, water, and air in the lake, its surroundings and tributaries, were performed. The aim of this paper has been to:

- i) Compile and evaluate empirical datasets from seven years of measurements in various matrices (water, air, sediment, fish) from upstream, downstream and within Lake Oxundasjön.
- ii) Define the magnitude, and area of impact of the present pollution situation.
- iii) Develop a mass balance model to identify crucial processes for the fate of PCBs in the lake system.
- iv) Identify present exposure routes of PCBs to non-migratory fish in the lake system.
- v) Predict temporal recovery aspects regarding PCBs in the lake system e.g., in sediment and non-migratory fish, applying different scenarios of remediation actions.

Historical background information and source apportionment of the contaminants are just briefly presented to limit the scope of this paper. Since the contamination situation was discovered in 2013, extensive investigations were made to find the source/sources. This included bottom sonar mapping to investigate potential dumping areas in the lake as well as water and sediment sampling in the tributaries to Lake Oxundasjön. The results indicated that dumping could not explain the situation. However, sampling in one of the tributaries to Lake Oxundasjön, River Väsbyån, resulted in elevated PCB levels in sediment and water which originated from an area in the central parts of Upplands Väsby municipality. In this area, located some 100–200 m from River Väsbyån, a metal industry was operating from the early 1900s until the factory was demolished in the 1990s. Extremely high levels of PCBs, with similar congener composition as the sediments of Lake Oxundasjön, were found in the soil at the former industrial site. Presently, procedure is ongoing to identify parties legally responsible for the contamination. Performed investigations to find and define the source area will be presented in full in Johansson et al. (in prep.).

2. Material and methods

2.1. Study area

Lake Oxundasjön is a boreal clay plain lake of 1.5 km² located in Upplands Väsby and Sigtuna municipalities in the greater Stockholm region of Sweden (Fig. 1). The catchment area is 270 km² and mainly consists of forested land, farmland, and semi-urban areas where about 100,000 persons are living. Lake Oxundasjön is eutrophic, with phosphorus concentrations typically around 75 µg/l, nitrogen concentrations varying between 730 and 1700 µg/l, Secchi depths of 1.0–2.6 m, and chlorophyll levels around 13 µg/l (Lindqvist, 2020). The physical characteristics of Lake Oxundasjön are summarized in Fig. 1. Fish species include perch (*Perca fluviatilis*), asp (*Aspius aspius*), northern pike (*Esox lucius*), pikeperch (*Sander lucioperca*), as well as carp species typically found in Swedish eutrophic lakes. There are two main tributaries; River Väsbyån (0.84 m³/s) and River Verkaån (0.64 m³/s). The lake outlet in the northern part of the lake forms a short river, River Marängsåån, discharging in a small bay (Rosersbergsviken Bay, 0.6 km²) in the northern part of Lake Mälaren. Lake Mälaren is the third largest lake in Sweden, supplying raw water for drinking water production for about 2 million persons in the Stockholm area. Lake Mälaren discharges into the Baltic Sea at the central parts of Stockholm.

2.2. Sampling and analysis

Sampling and analysis procedures are presented briefly below. See Table S1 for description of limits of quantification (LOQ) and uncertainties of the performed chemical analyses.

2.2.1. Biota

PCBs in biota was mainly investigated in non-migratory fish, including European perch and northern pike. Sampling was performed in 2013–2017 primarily using bottom nets, with a complete sampling procedure described in Karlsson and Viktor (2014). Samples consisted of perch in standard Swedish environmental monitoring size (15–20 cm, $n = 24$, 2+ years), analyzed as collective samples of 5–7 individuals. Northern pike (47–106 cm, $n = 29$) was analyzed individually. IVL Swedish Environmental Research Institute (IVL) performed chemical analysis of PCBs (Σ PCB₇, i.e., congeners PCB₂₈, 52, 101, 118, 153, 138 and 180) according to SS-EN ISO/IEC 17025:2018.

2.2.2. Sediment

During 2014–2017, 163 sediment samples, of which 39 surface sediment samples (0–5 cm) and 22 sediment cores (up to 60 cm depth) from Lake Oxundasjön were analyzed. Sampling also included surface sediments upstream of Lake Oxundasjön ($n = 18$) and downstream in Lake Mälaren ($n = 7$). Sediment samples were collected by grab samplers (Ponar) and core samplers (Kajak and Niemistö). Sampling stations are presented in Fig. 2.

Σ PCB₇, organic material, and water content were analyzed in homogenized material from 5 cm-intervals. Additionally, three deeper cores (0–60 cm) sampled in 2017 were also analyzed for metals (As, Ba, Cd, Co, Cu, Cr, Ni, Pb, V, Zn) in 2 cm intervals. The chemical analysis was performed by Eurofins Scientific (according to SS-EN 16167:2018 + AC:2019 for PCBs and SS 028150:1993/SS-EN ISO 17294-2:2016 for metals).

The total mass (kg) of PCBs in the sediments was estimated from the average content of Σ PCB₇ in 5 cm vertical layers in ten horizontal sub-compartments from the northwestern to the southern part of the lake. Each sub-compartment represented 10 % of the total area of accumulation sediments (1.36 km²). Measured Σ PCB₇ content (mg/kg dw) in each sediment layer/sub-compartment was then transformed to mass Σ PCB₇ (kg) from the sediment density. The sediment density was calculated from water and organic content (Håkanson and Jansson, 1983).

Historically, emissions from a municipal sewage water treatment plant as well as emissions from a metal industry located in Upplands Väsby

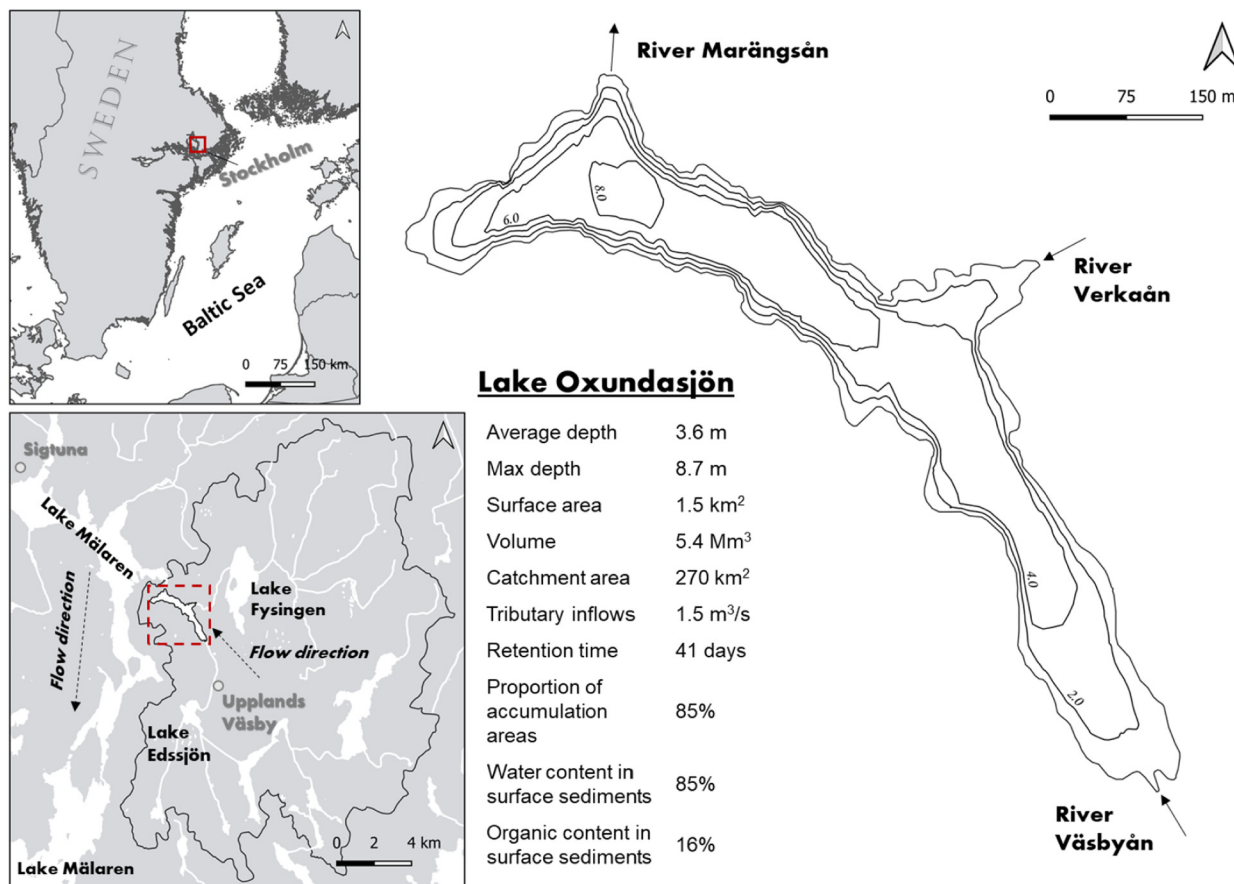


Fig. 1. Study area including catchment area of Lake Oxundasjön (bottom left), physical characteristics (middle) and lake bathymetry (right).

were released into River Väsbyån, causing eutrophication issues and elevated levels of heavy metals in Lake Oxundasjön. From the following known historical stages in industrial production and operation, three sediment cores (0–60 cm) from the southern end of Lake Oxundasjön were dated:

- i) The metal industry was started in 1903 – emissions of copper (Cu) and zinc (Zn) from brass production. The industry was closed during most of the 1920s but reopened in 1928 with increased production in the following years.
- ii) Galvanic surface treatment in the metal industry was introduced in 1933 – emissions of chrome (Cr) and nickel (Ni) from galvanization and chrome-plating.
- iii) Sewage water from the municipal treatment plant was rerouted to a centralized treatment plant outside Stockholm in 1970, resulting in significantly better conditions in River Väsbyån and Lake Oxundasjön in terms of nutrients and oxygen-consuming materials.
- iv) The metal industry reduced its operation during the 1980s and closed in 1990, resulting in significant reduction of heavy metals emissions into Lake Oxundasjön.

2.2.3. Water

Water sampling was performed from 2016 to 2020 in River Verkaån ($n = 11$), River Väsbyån upstream ($n = 22$) and downstream ($n = 23$) the urban area of Upplands Väsby, Lake Oxundasjön ($n = 19$), lake outlet/River Marängsån ($n = 23$), Rosersbergsviken Bay ($n = 13$) and about 8 km downstream in Lake Mälaren ($n = 12$). Chemical analysis was performed by IVL Swedish Environmental Research institute according to SS-EN ISO/IEC 17025:2018. The sampling scheme consisted of 4–6 samples yearly (2×2.5 l in glass bottles) with an analysis of ΣPCB_7 . Sampling stations are presented in Fig. 2.

Flow measurement data for the tributaries and the lake outlet was supplied by the Swedish Meteorological and Hydrological Institute (SMHI) from a national river-monitoring program (<https://www.smhi.se/data/hydrologi/vattenwebb>, 2021-03).

2.2.4. Air and deposition

Air and atmospheric deposition sampling were performed in the vicinity of Lake Oxundasjön (Fig. 2) on two occasions in 2016, each lasting one month (March/April, August/September). Air sampling was also performed in the central parts of Upplands Väsby in December/January 2016/2017 and in July 2017. Air sampling was performed using a low-volume sampler, equipped with glass-fiber filters to collect PCBs in particle-phase and two glass columns with an adsorbent of polyurethane foam (PUF) to collect PCBs in gas-phase. The pump used had a pump capacity of 35–40 l/min, giving a total air volume through the filters and PUF of about 1500–1700 m³ during each measurement period. The results are presented as sum of particle and gas phase, but filters (particle-phase) and PUF (gas) were analyzed separately.

Atmospheric deposition was sampled using a bulk sampler, consisting of a 1 m² Teflon-covered surface with 10 cm high edges. Dry deposition on the surface was collected with ethanol-moist glass-fiber filters and the wet deposition was collected in a container with a PUF-adsorbent. IVL performed both air and deposition analyses.

2.3. The mass balance approach

A dynamic mass balance model, initially developed for phosphorus turnover (Malmaeus and Håkanson, 2004), was used to evaluate PCB turnover and fate transports in the lake system. The model has also been derived and tested in Baltic coastal areas for PCDD/Fs (Karlsson et al., 2010).

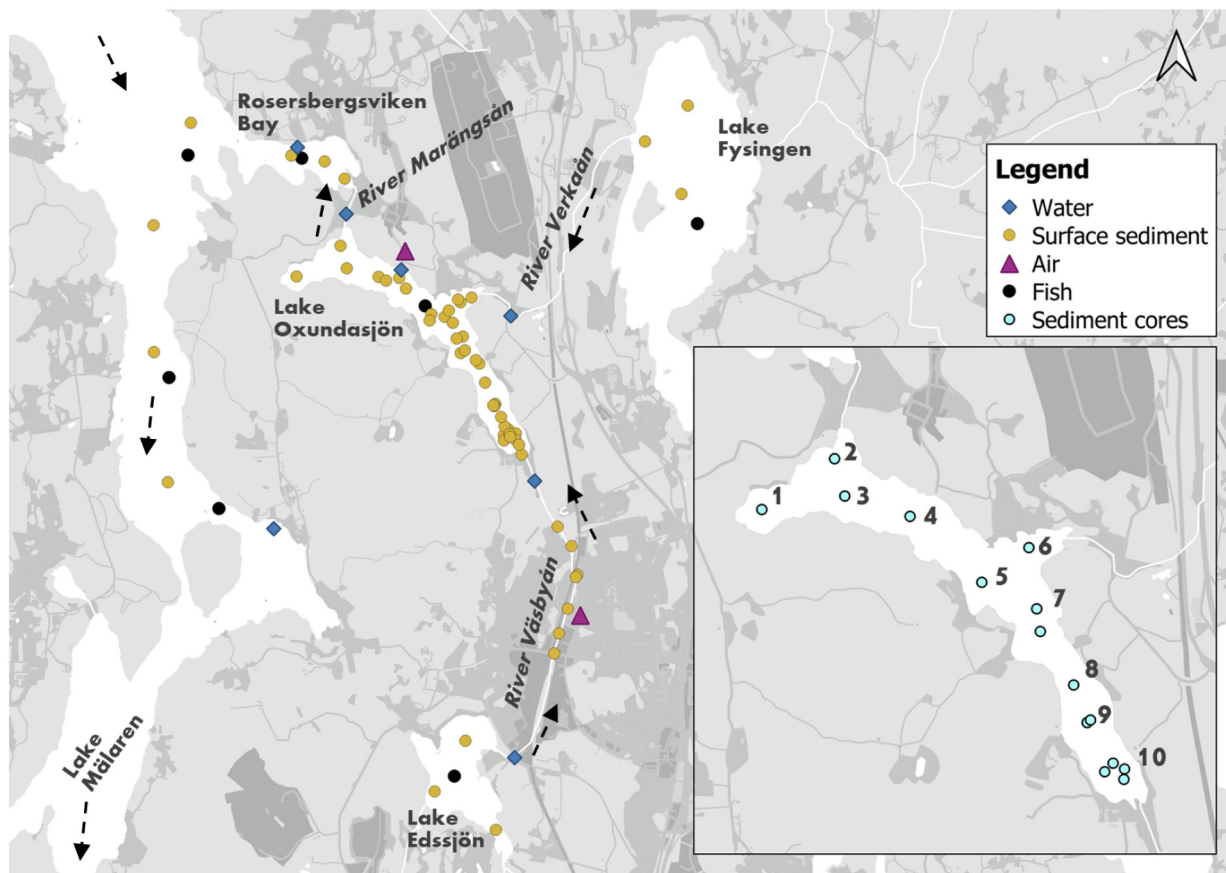


Fig. 2. Sampling stations for PCBs in water, sediment, fish, and air in the Lake Oxundasjön area. The inset map shows sampling stations for sediment cores in Lake Oxundasjön, which is divided into ten sub-compartments. The arrows show the flow direction.

A conceptual model describing the dynamic transport processes for PCBs is presented in Fig. 3. The model consists of three dynamic compartments: surface water and two sediment compartments. Erosion and transport (ET) sediment are found above the wave base, where wind and current-driven erosion and transport processes dominate. Accumulation (A) sediments are below the wave base, where an accumulation of fine particles is allowed (Håkanson and Jansson, 1983). A-sediments are furthermore divided into two sub-compartments: bio-active and bio-passive A-sediments. The bio-active compartment is defined as the top 10 cm of the sediments, whereas sediments deeper than 10 cm are considered as permanently buried.

PCBs is among the most studied persistent organic pollutants, and it is well known that the congeners differ substantially in their partitioning

characteristics and environmental fate (Li et al., 2003). Therefore, the model was constructed, calibrated, and run separately for the seven PCB congeners included in ΣPCB_7 . Finally, the results for each separate congener were summed up and presented as ΣPCB_7 . Consequently, it should be stressed that the model is valid for the seven congeners included in ΣPCB_7 .

Dynamic processes were implemented in STELLA Professional®, version 1.9.1. All model equations with including parameter values and initial conditions are presented in the Supplementary Material (Section 2). Each process is briefly described below. The model was run monthly, and the simulation period was set to 25 years. Transport processes were computed iteratively in STELLA, where the time step was set to 0.05 months. All compartments were considered homogenic and that PCBs was evenly distributed inside each compartment.

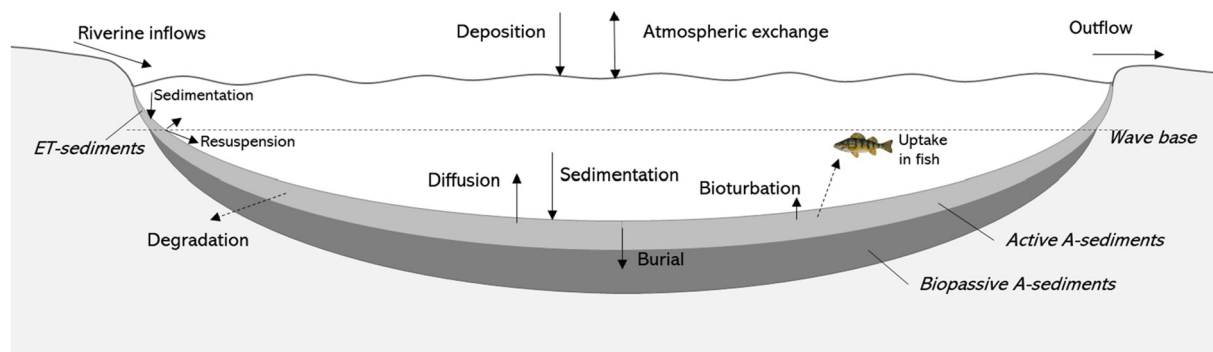


Fig. 3. A conceptual model to illustrate dynamic transport processes of PCBs in Lake Oxundasjön. The model consists of three compartments: surface water, ET-sediments and A-sediment (which is divided by the wave base). The arrows show direction of transports of PCBs between the different compartments. The model was implemented in STELLA Professional®, version 1.9.1.

2.3.1. Riverine inflows and outflows

Monthly mass transports (g/month) from riverine inflows (River Väsbyån and River Verkaån) were based on measured ΣPCB_7 concentrations in water during 2016–2020 and modeled monthly discharge during 2010–2020 from SMHI. Empirical water concentrations in the lake outlet were used to calibrate the model.

2.3.2. Air-water exchange and deposition

The air-water exchange of PCBs from water to air (dissolved and gas phases) was estimated by applying the two-film theory (Whitman, 1923; Liss and Slater, 1974), where concentrations in air and water were replaced by fugacity according to the procedure described in Mackay (2001). Default parameters, e.g., mass transport coefficients (m/h) for PCB homologs as well as Henry's Laws constants (Mackay, 2001; Li et al., 2003), were applied (see Section 2 of the Supplementary Material). Site-specific parameters included lake surface area (1.5 km²) and monthly empirical ΣPCB_7 concentrations in water and air. Air temperature data were acquired from a national monitoring station in Stockholm-Arlanda: N59.6269, E17.9545 (<https://www.smhi.se/data/meteorologi/temperatur,2021-03>) and modeled water temperature data from SMHI (<https://www.smhi.se/data/hydrologi/vattenwebb,2021-03>).

Atmospheric deposition of ΣPCB_7 was acquired from measurements in 2016.

2.3.3. Sedimental processes

2.3.3.1. Sedimentation. Sedimentation of particulate PCBs was estimated using equations specified in Karlsson et al. (2010), with settling velocity set to 6 m/month.

The fraction of particulate PCBs (i.e., PCBs bound to particles) was calculated from equations described in Burkhard (1998). Average total organic carbon (TOC) concentration in the outlet of Lake Oxundasjön was 10.4 mg/l between 2017 and 2019 (Lindqvist, 2020). Here, we assumed the fraction dissolved organic carbon (DOC) to be 90 %. Carbon/water partitioning coefficients (K_{OC}) were estimated from K_{OW} -values given in Shiu and Mackay (1986) and transformed to K_{OC} by using $K_{OC} = 0.41 * K_{OW}$ (Karickhoff, 1981). Particulate fractions varied from 32 % for lower chlorinated congeners (PCB28) to 96 % for higher chlorinated congeners (PCB180). Calculations and estimated particulate fraction of all the included congeners are presented in Table S2.

2.3.3.2. Resuspension. Resuspension of particulate PCBs from ET-areas to surface water or directly to A-sediments was estimated from equations given in Karlsson et al. (2010). The turnover rate of ET-sediments was set to a default value of 1 month.

2.3.3.3. Burial. Burial from bio-active A-sediments (0–10 cm) to deeper, bio-passive sediments (>10 cm) is described in Karlsson et al. (2010). The turnover rate of A-sediments (Age_A) was calculated to be 308 months (see Section 2 in Supplementary Material). The gross sedimentation was derived from field measurements in Lake Oxundasjön (~700 mg/m² * year).

2.3.3.4. Diffusion. The fugacity approach (Mackay, 2001) was applied to model the diffusive transports of PCBs from active A-sediments to surface water. Site-specific parameters included the area of A-sediments, sediment density, and modeled water and sediment concentrations in any given timestep (see Section 2 in Supplementary Material). Potential transport caused by bioturbation of benthic fauna in sediment was assumed to be included in the diffusion rate.

2.3.4. Degradation/dechlorination

Contained evidence regarding degradation/dechlorination of PCBs in aquatic environments is conflicting, and limited, especially in northern conditions (Sinkkonen and Paasivirta, 2000; Aronson et al., 2006). However, a degradation rate was estimated for the modeled PCB congeners in the A-sediment compartment, derived from half-life ($t_{1/2}$) of PCB_{tot} (i.e., all 209

theoretical congeners) in sediments given in Mackay (2001); 700 months. The degradation rate (month⁻¹) was then calculated from $\ln(2)/t_{1/2}$ and multiplied by the mass of PCBs in the A-sediment compartment. No consideration was taken for degradation/dechlorination in the water compartment due to the limited turnover rate (~40 days) compared to the estimated half-life of PCB_{tot} in water (6.3 years) (Mackay, 2001).

2.3.5. Predicting levels of PCBs in non-migratory fish

Using data from regional monitoring programs in the Stockholm region, a log-linear correlation (p -value < 0.001) was found between levels of ΣPCB_7 in surface sediments and non-migratory fish (perch, 15–20 cm) from 25 different locations in the Stockholm region, including the Lake Oxundasjön area (Fig. S1). From the log-linear correlation, PCB levels in perch (y) from Lake Oxundasjön were predicted in the model using modeled content in surface sediment (x). Congener specific correlations were used in the sediment-fish module (see Fig. S2). To clarify, seven separate models were set up in STELLA, one for each congener, and the resulting congener specific levels in fish were summed up and presented as ΣPCB_7 (ng/g ww).

2.3.6. Initial conditions, model calibration and sensitivity

To set the initial conditions of the model, averaged data from surface sediments (2014–2017), surface water (monthly, 2016–2020), air (2016–2017) and fish (2014–2018) were used. The initial values used (concentrations and mass of all included congeners) are presented in Section 2 of the Supplementary Material. Since the data used to set the initial conditions spans over the period 2014–2020, we can roughly estimate that the initial year in the model represents year 2017. With this estimation, the last year in the model (i.e., model year 25) represents year 2042.

Calibration of the mass balance model was performed by adding a dimensionless scaling factor to the sediment diffusion rate. This does not imply that all other processes are considered more certain but was motivated by sediment diffusion being the main transport process. By tuning the diffusion rate, modeled water concentrations were calibrated to match data from five years of water sampling (2016–2020) in the Lake Oxundasjön outlet.

The robustness of the model was tested with uncertainty analysis according to Håkanson and Bryhn (2008). The test was performed with Monte Carlo simulations ($n = 1000$), with simulated ΣPCB_7 -levels in perch in 25 years as target value. The test was based on generated random value series with assigned coefficient of variations (CV values) for five transport processes: air-water exchange (CV = 0.5), sediment diffusion (CV = 0.5), sediment degradation (CV = 0.5), sedimentation (CV = 0.1), and burial (CV = 0.2). Note that the CV values were applied to the transport processes above, but not for each parameter within in each process eq. E.g., when testing the air-water exchange, the CV value were applied for the resulting transport of each congener, and not for each underlying parameter used to calculate the transport (e.g., Henry's Laws constant). In addition, the sediment-fish correlation was also tested with an assigned CV value of 0.3. Riverine inflows and outflows were not tested since being based on a robust set of data. Resuspension and atmospheric deposition were also excluded due to the limited impact on the mass balance.

2.3.7. Remediation scenarios

Generally, risk management strategies for aquatic environments with contaminated sediments are based on monitored natural recovery (MNR), dredging (removal), or isolation (capping). Three different scenarios for PCBs in Lake Oxundasjön were evaluated with the model:

1. Natural recovery, driven by sedimentation of new “clean” material, causing burial of polluted sediments over time (i.e., no active removal of PCBs).
2. Dredging all contaminated A-sediments (1.36 km²), resulting in a 99 % reduction of PCBs in active A-sediments. Assuming that the transport of PCBs from River Väsbyån is reduced to background levels (i.e., concentrations measured in River Verkaån).
3. Dredging the southern end of Lake Oxundasjön (20 % of the area of A-sediments, reducing the total mass of PCBs in active A-sediments by

75 %), combined with in-situ capping or using activated carbon over the entire lake area, resulting in reduced diffusion from sediment to surface water by 75 %. Assuming that the transport of PCBs from River Väsbyån is reduced to background levels.

Model results are presented as initial and final concentrations of PCBs in different matrices, given the three different scenarios. Scenarios 2 and 3 include dredging, which inevitably disturbs the bottom sediment and resuspends part of it. We assume that the effect from resuspension, or other spills during the dredging procedure, can be neglected in this case. This is motivated using modern technology, as well as strong Swedish legislation regarding the limiting of such effects.

There are several other remediation technologies for PCBs, e.g., described in [Jing et al. \(2018\)](#). However, few have been implemented on a large scale and were therefore not evaluated in this study.

3. Results

3.1. Measured levels in various matrices

In the following sections, data from Lake Oxundasjön and its surrounding areas are presented. Data are mainly presented as ΣPCB_7 , however congener specific data from all measurements included in this paper are presented in Section 3 of the Supplementary Material.

3.1.1. Water

Concentrations of ΣPCB_7 in River Väsbyån, Lake Oxundasjön, and downstream the lake in Lake Mälaren are presented in [Fig. 4](#). Concentrations in River Verkaån were generally below the limit of quantification for all congeners, and therefore, not included in the figure. Concentrations averaged 0.16 ng/l in the upstream part of River Väsbyån, while concentrations increased tenfold to 1.4 ng/l downstream the Upplands Väsby municipality. In Lake Oxundasjön and its outlet River Marängsån, concentrations increased another ten times, averaging 12 ng/l. Seasonal variations in PCB concentrations at the outlet of Lake Oxundasjön are presented in [Fig. 5](#), combined with water temperature and discharge. The data clearly indicates higher concentrations during the warmer months (Apr–Oct/Nov), compared to the winter months (Nov–March).

3.1.2. Sediment

ΣPCB_7 -content in surface A-sediments from Lake Oxundasjön generally varied from 1.5 to 4.5 mg/kg dw ([Fig. 4](#)). However, the levels were higher in deeper layers (~10–40 cm), with up to 280 mg/kg dw in the lake's southern end (Table S3). In upstream lakes, Lake Edssjön and Fysingen, the average ΣPCB_7 -content in surface sediments were 0.067 and 0.0011 mg/kg dw, respectively. In Lake Mälaren, downstream Lake Oxundasjön, levels gradually decreased from about 0.3–0.5 mg/kg dw in Rosersbergsviken Bay to 0.016 mg/kg dw about 5 km further downstream.

The mass of ΣPCB_7 in the lake sediments was calculated to 1100 kg (Table S3). The Swedish EPA suggests that ΣPCB_7 represents about 20 % of the total amount of PCBs, PCB_{tot} , in soils ([SEPA, 2016](#)). From this approximation, the mass of PCB_{tot} in the sediments of Lake Oxundasjön estimates to 5500 kg. However, as [Megson et al. \(2019\)](#) pointed out, the correlation of ΣPCB_7 and PCB_{tot} varies and should be interpreted cautiously, especially in sites where PCBs are likely to undergo dechlorination due to microbial activity.

3.1.2.1. Sediment dating. Using historical stages in urban emissions to River Väsbyån and Lake Oxundasjön (see [Section 2.2.2](#)), three sediment cores from the lake's southern end were dated. In [Fig. S3](#), levels of heavy metals are presented in one of the dated sediment cores (Ox 10:4) as an example. An increase from background levels of Cu and Zn were seen in the deeper sediment layers, which we assumed relates to the year 1903 when the metal industry in Upplands Väsby was started. We then assumed a constant yearly deposition of dry sediment (see [Fig. S3](#)). The increasing levels of Cu and Zn at the start of the 1900s are followed by decreasing levels in the

1920s, matching limited industrial activity during this period. The levels then increased around 1930, which relates to the re-opening of the metal industry in 1928. Galvanic surface treatments were introduced in 1933, assumably causing emissions of Cr and Ni to the receiving waters. This is also reflected in the dated sediment cores, where levels of Cr and Ni increased in the 1930s. Furthermore, concentrations of Cu, Zn, Cr, and Ni simultaneously declined in the 1980s, correlating to reduced industrial activity and the closing of the metal industry around 1990. Visual inspection of the dated sediment cores also indicated improving oxygen conditions in the lake bottom water from 1970 to 1980, which may be explained by the rerouting of wastewater from the municipal treatment plant in 1970.

From the dated sediment cores, sedimentation rates (mm/yr) were calculated from accumulated deposition of dry sediment ($\text{g}/\text{m}^2 \text{ yr}$). Sediment accumulation varied from 600 to 2300 $\text{g}/\text{m}^2 \text{ yr}$, corresponding to sedimentation rates of 3–7 mm/yr. The highest sediment accumulation was found close to the river mouth of River Väsbyån, with decreasing accumulations further out in the lake.

By matching the dated sediment cores against PCB levels in each core, we found that the PCB emissions to Lake Oxundasjön could have begun in the late 1940s and then gradually increased until towards the (early) 1970s, followed by an inflow decrease around 1980 ([Fig. 6](#)).

3.1.3. Biota

In [Fig. 4](#), levels of ΣPCB_7 in fish (European perch and northern pike) from Lake Oxundasjön are compared to fish from upstream and downstream lakes and the Stockholm city and archipelago regions ([Karlsson and Viktor, 2014](#)). Results showed the highest levels in fish from Lake Oxundasjön and Rosersbergsviken Bay, with gradually decreasing levels downstream in Lake Mälaren. Levels in perch from Lake Fysingen were comparable to those from Lake Mälaren as well as the outer parts of the Stockholm archipelago, indicating a limited urban impact. The levels in Lake Edssjön were similar to those in urban affected waters, i.e., upstream in Lake Mälaren, Stockholm city, and its inner archipelago.

3.1.4. Air and deposition

Air concentrations of ΣPCB_7 (sum of gas and particle phase) near Lake Oxundasjön averaged 39 pg/m^3 and peaked in late summer with 72 pg/m^3 . Winter concentrations in the urban area of Upplands Väsby were measured to 842 pg/m^3 and summer concentrations to 424 pg/m^3 .

Atmospheric deposition of ΣPCB_7 at Lake Oxundasjön was measured to 0.54 $\text{ng}/\text{m}^2 \text{ day}$ in March/April and 1.0 $\text{ng}/\text{m}^2 \text{ day}$ in August/September (i.e., 0.78 $\text{ng}/\text{m}^2 \text{ day}$ on average).

3.1.5. Congener distribution in different matrices

In [Fig. 7](#), the relative congener distribution of PCB28, 52, 101, 118, 153, 138 and 180 in European perch and surface sediments from the Lake Oxundasjön area is presented. Fish and sediment from “Upstream Lake Oxundasjön” represent Lake Fysingen and Lake Edssjön and “Lake Mälaren” are samples from Lake Mälaren downstream the area of impact of Lake Oxundasjön. Both fish and sediment samples from Lake Oxundasjön are dominated by lower chlorinated PCBs, while upstream and downstream samples is dominated by higher chlorinated congeners.

3.2. Model output

3.2.1. Model calibration and sensitivity

The model was calibrated by increasing the diffusion from sediments by 100 % during summer (April–September) and reducing it by 50 % during the winter (October–March) ([Fig. S4](#)). Prior to calibration, the diffusion was relatively constant over the year.

Assigned CV values for included variables in the model sensitivity analysis are shown in [Table S4](#). The target variable was ΣPCB_7 -levels in perch in 25 years. Model uncertainty and sensitivity were tested with Monte Carlo simulations ($n = 1000$) in two steps: first, all selected variables were included (i.e., air-water exchange, sediment diffusion, sediment degradation, sedimentation, burial, and sediment-fish correlation), second, each variable was

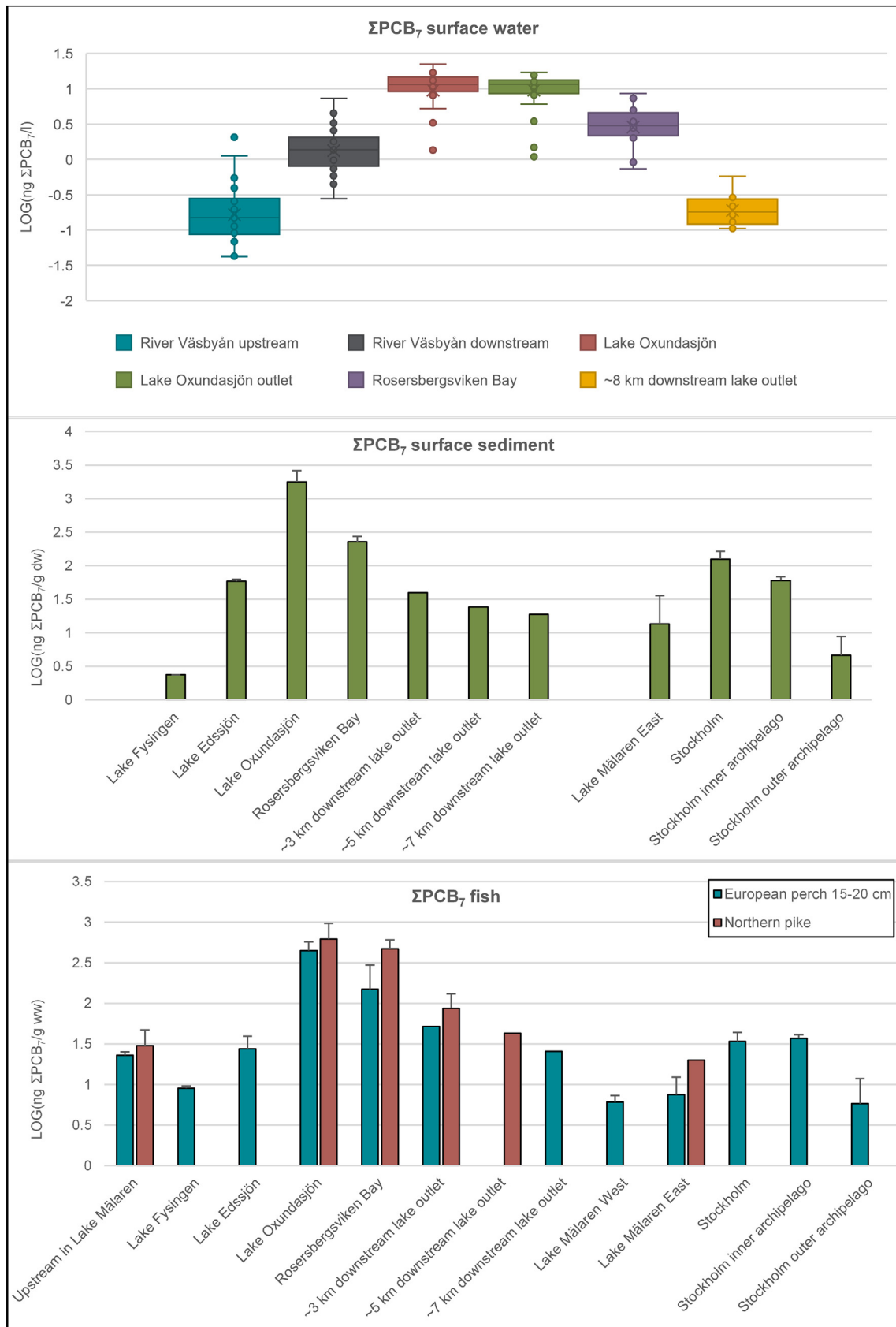


Fig. 4. ΣPCB₇ in water (2016–2020), surface sediments (2014–2017), and fish (European perch and Northern pike, 2013–2017) in Lake Oxundasjön compared to upstream and downstream waters in the Stockholm area. The general water flow is from left to right in the graphs.

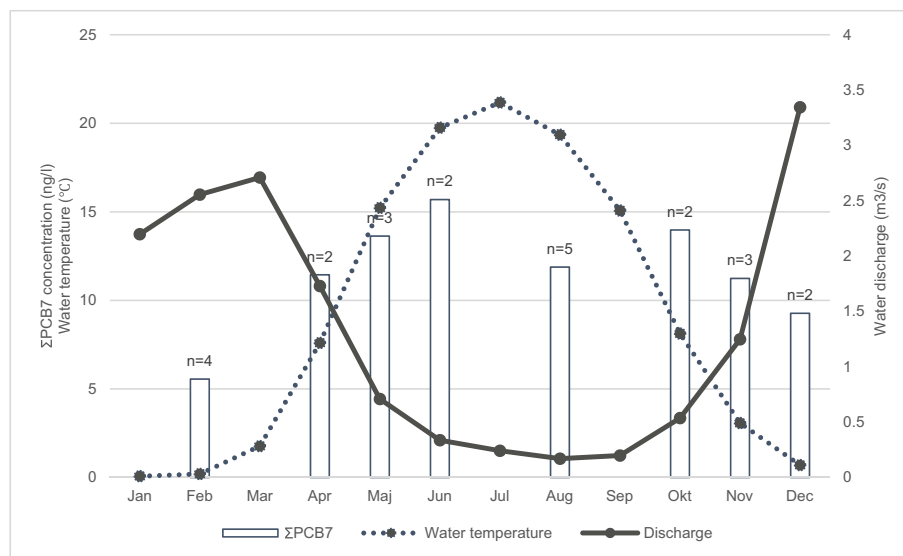


Fig. 5. Monthly (average 2016–2020) Σ PCB₇ concentrations, water temperature and discharge in the outlet of Lake Oxundasjön. Modeled temperature and discharge data from SMHI.

tested independently. With all variables included, the simulation resulted in CV value = 0.35 for the target variable, with average value 83 ng/g ww, min- and max values with 95 % confidence interval of 38 and 150 ng/g ww (Fig. S5). By independently testing each variable, we found the highest CV values for the sediment-fish correlation (0.24), sediment diffusion (0.2), burial (0.14), and sediment degradation (0.10), see Table S4.

3.2.2. Transport and fate of PCBs

A mass balance for present transports of Σ PCB₇ in Lake Oxundasjön is presented in Fig. 8. Main transport processes were air-water exchange (1600 g/yr), sedimentation (170 g/yr), sediment-water exchange (2100 g/yr), burial (2800 g/yr), degradation (870 g/yr) and riverine outflow to Lake Mälaren (430 g/yr).

3.2.3. Remediation scenarios

Results from remediation scenarios 1, 2 and, 3 are presented in Table 1. With natural recovery (scenario 1), Σ PCB₇-content was about 455 ng/g dw in A-sediments and 83 ng/g ww in perch after 25 years. PCBs declined by 7.4 %/year in A-sediments, and 5.4 %/year in perch, which corresponds to environmental half-life (i.e., the total turnover rate of PCBs) of 9.3 years in A-sediments and 13 years in perch. Confidence errors of predicted values were obtained from the Monte Carlo analysis.

Dredging all contaminated sediments (scenario 2) reduced the initial PCB levels in the lake sediments by 99 %, resulting in levels in perch on 3.5 ng/g ww after 25 years. Dredging the southern end of the lake and reducing diffusion from sediments with either in-situ capping or activated carbon (scenario 3) reduced initial sediment surface concentrations by about 75 %. In 25 years, predicted levels in sediments were 180 ng/g dw and 42 ng/g ww in perch.

Given natural recovery, about 5 kg Σ PCB₇ is transported from the lake outlet to Lake Mälaren over the simulation period (25 years). During the same period, about 18 kg Σ PCB₇ is transported to the air due to air-water exchange. However, in scenarios 2 and 3, transports to Lake Mälaren are reduced to 0.13 and 0.45 kg Σ PCB₇, respectively. The air-water exchange over 25 years is reduced to 0.30 kg in scenario 2 and 1.5 kg in scenario 3.

4. Discussion

4.1. Area of impact

From performed sampling in non-migratory fish, water, and sediments, an area of impact for the present contamination situation of PCBs in the

Lake Oxundasjön area was defined, both geographically (Fig. 4) and temporally (Fig. 6).

The extensive sediment sampling performed in Lake Oxundasjön has shown that the main transport route of PCBs to the lake originates from River Väsbyån, which has created a hot spot area in the southern end of the lake. Due to more particulate material settling in this area close to the river mouth, the contamination reaches deeper in the sediments than in the lake's middle and northern parts (Table S3). The relatively smooth distribution of Σ PCB₇ in the sediment cores indicates decades of continuous emissions of PCBs to the lake. Visual analysis of sediment cores (Fig. 6) also indicates anoxic conditions and limited bioturbation in deeper layers, which would have resulted in more distinct “spikes” of PCBs if the emissions had originated from, e.g., single accidents or dumping. The levels gradually decrease towards the surface sediments and are relatively equal in the surface sediments (1.5–4.5 mg/kg dw), indicating an ongoing natural recovery where the present load of PCBs to the lake is limited compared to historical emissions. However, the levels in surface sediments still are about a factor 200 above the limit of “very high levels” given in the Swedish EPA classification system for PCBs (SEPA, 1999).

Sediment dating indicated that the main load of PCBs to Lake Oxundasjön likely occurred from the end of the 1940s until about 1980. Since PCBs were not manufactured in Sweden and the large-scale global export of PCBs started around 1950 (Breivik et al., 2002), it makes sense that no substantial emissions of PCBs occurred to the lake before that. The decrease of PCBs in the 1970s also makes sense, mainly due to the gradual implementation of restrictions on PCBs in the 1970s, which coincides with a drastically reduced global production (de Voogt and Brinkman, 1989).

Levels in non-migratory fish, sediment, and water (Fig. 4) all show gradually decreasing Σ PCB₇ levels with increased distances downstream Lake Oxundasjön and further downstream in Lake Mälaren. The main area of impact seems to be limited to the outlet of Rosersbergsviken Bay, with levels in all matrices reflecting urban affected waters about 3–5 km downstream the outlet of Lake Oxundasjön. There are no indications of substantial impact from PCB emissions upstream in Lake Mälaren. Additionally, the congener patterns of PCBs observed in Lake Oxundasjön differ from surrounding areas, thus indicating a unique source (Fig. 7). Congener patterns from different matrices in the Lake Oxundasjön area have been evaluated and will be presented in Johansson et al. (in prep.).

Σ PCB₇ concentrations in air at Lake Oxundasjön were about tenfold higher than at a national monitoring station on the east coast of Sweden during 1996–2019 (Fredricsson et al., 2021), indicating atmospheric release of PCBs from the lake and/or from the urban area of Upplands

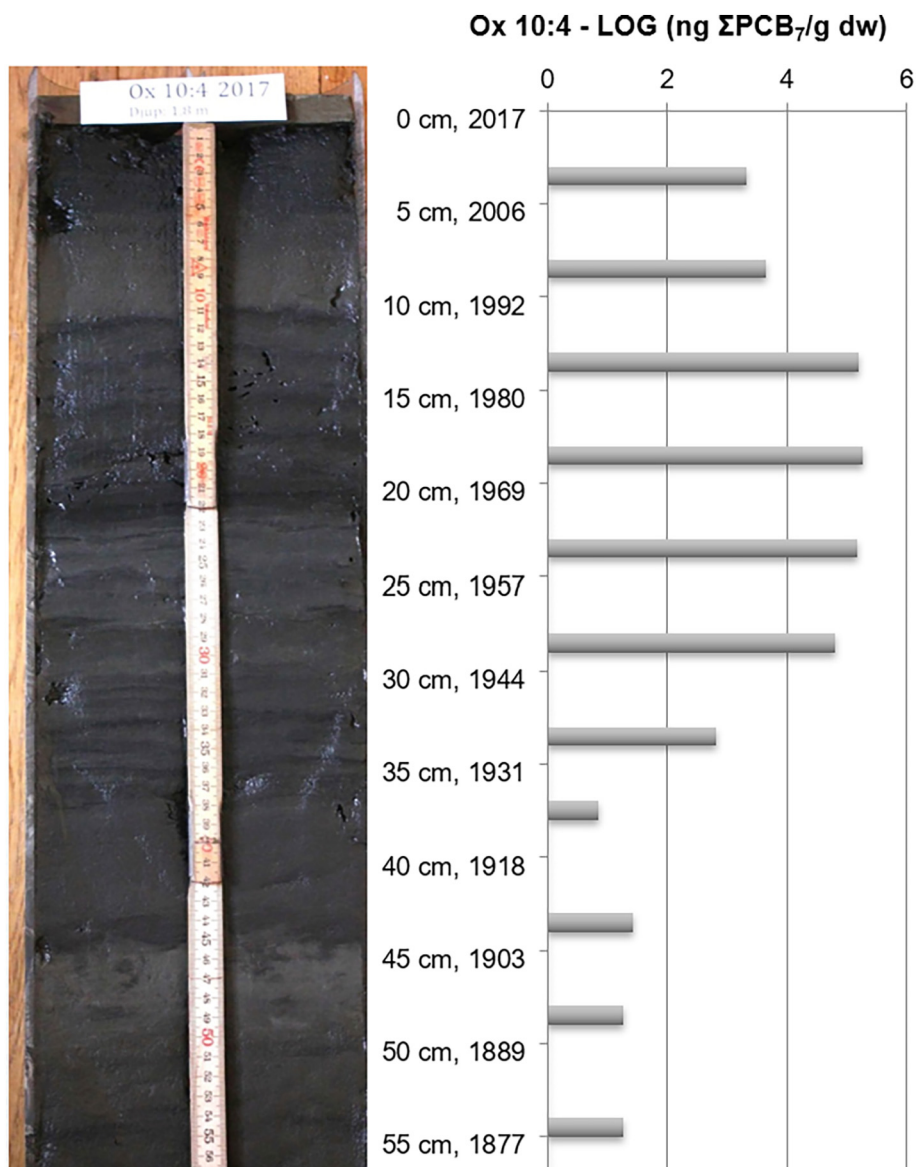


Fig. 6. Content of Σ PCB₇ (mg/kg dw) in a dated sediment core (0–60 cm) from the southern part of Lake Oxundasjön. Photo: Per Jonsson, March 2017.

Väsby municipality. Like our results, Σ PCB₇ concentrations at the national monitoring station varied seasonally, with higher concentrations during summer caused by increased PCB volatilization from soils and waters. In the urban area of Upplands Väsby municipality, air concentrations were about ten times higher than concentrations at Lake Oxundasjön. Here, winter concentrations were probably higher than summer concentrations due to extensive removal of PCB-contaminated soil performed during the measurement period (December 2016–January 2017).

Due to the elevated PCB levels in non-migratory fish, Sigtuna and Upplands Väsby municipalities advise against consuming fish from the area. To put in perspective, the levels in fish from Lake Oxundasjön are comparable to (or higher than) fish from the Swedish Lake Järnsjön prior to remediation in 1993–1994 (Larsson et al., 1992).

4.2. Fate and transports of PCBs

Mass balance modeling identified air-water exchange, sedimentation, sediment-water diffusion, burial, degradation, and riverine outflow to Lake Mälaren as the most important transport processes for PCBs (Fig. 8). Expressed as Σ PCB₇, >400 g was transported yearly to Lake Mälaren, which accounts for about 8 % of the fate of PCBs. The main fate of PCBs was sediment burial,

about 2800 g/yr, i.e., nearly 50 % of the fate transport. The air-water exchange increased in summer to 37 g/month, compared to 7 g/month during the winter. In comparison, Thomann et al. (1991) found that volatilization accounted for 66 % (higher for lower chlorinated congeners) and sedimentation for 6 % of the fate transport of PCBs in the Hudson River estuary in years 1947 through 1987, with remaining transports consisting of burial and dredging. Volatilization of PCBs from previously mentioned Lake Järnsjön was estimated to be only 9 g/yr prior to dredging (Gullbring and Hammar, 1993). The substantially higher volatilization from Lake Oxundasjön might be explained by larger lake area (1.5 km² vs. 0.26 km²) and slower water turnover rates (40 days vs. 4 h) compared to Lake Järnsjön.

The primary source of PCBs is currently diffusion from the lake sediments to water (about 2000 g/year), about 40 times the present load from tributaries. Similarly, the dominating source of PCBs in Lake Järnsjön was diffusion, where 5–8 kg PCBs (PCB_{tot})/yr was estimated to be released from the lake sediments prior to dredging (Larsson et al., 1990; Gullbring and Hammar, 1993).

In Lake Oxundasjön, natural recovery (i.e., scenario 1) indicates an annual decline of PCBs in perch by 5.4 % (Table 1). This agrees with the general trend of PCBs in herring from the Baltic Sea, where levels are decreasing with 2.6–5.5 %/yr (PCB28) and 2.0–5.7 %/yr (PCB153) during

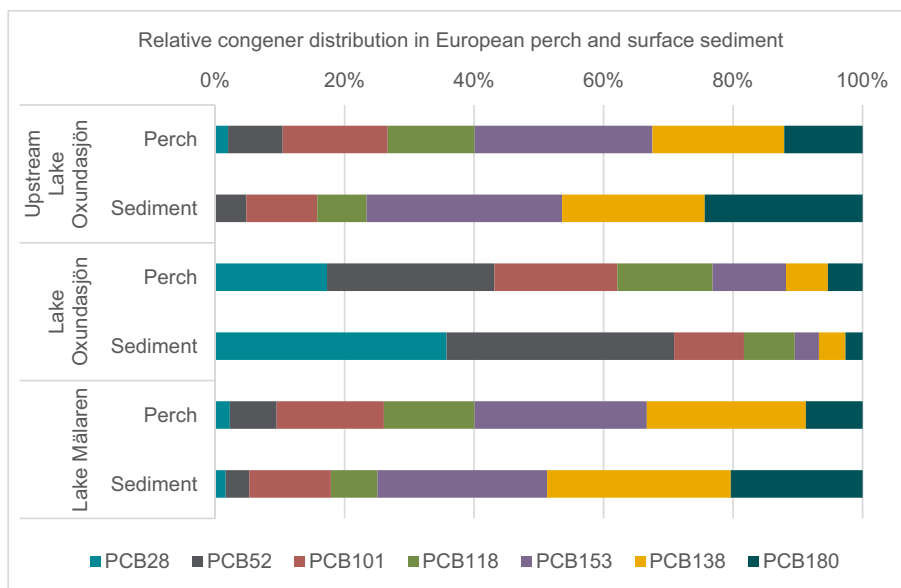


Fig. 7. Relative congener distribution of PCB28, 52, 101, 118, 153, 138 and 180 in European perch and surface sediments from the Lake Oxundasjön area.

1987–2016 (Glynn et al., 2020). Furthermore, between 1960 and 1996, PCB levels in Arctic char from Swedish Lake Vättern were reduced by 5 %/yr and northern pike from Swedish Lake Bolmen by 6 %/yr (Lindell et al., 2001). The decline of PCBs in the A-sediments of Lake Oxundasjön corresponded to an environmental half-life of 9.3 years. The suggested environmental half-life of PCBs in Baltic Proper sediments varied from 3 years for PCB28 to 38 years for PCB180 (Sinkkonen and Paasivirta, 2000). In contrast, suggested global environmental mean residence times of PCB153 and PCB180 are about 110 and 70 years, respectively (Jönsson et al., 2003).

Dredging is historically the most common remediation method for polluted sediments. When Lake Järnsjön was dredged in 1993–1994, a 97 % reduction of PCBs in the sediments was accomplished (Bremle et al., 1998). Since then, with technical improvements, a 99 % reduction of PCB content in A-sediments was believed to be realistic. As expected, dredging all contaminated lake sediments (scenario 2) was most effective for removing PCBs. This also resulted in PCB levels in perch below existing environmental quality standards and marketing limits. However, after remediation, older fish will still be affected by the contaminants while newborn fish gradually will form a population of less contaminated fish. For example, Bremle and Larsson (1998) found that PCB levels were halved in fish from Lake Järnsjön between 1991 and 1996, where dredging occurred in 1993–1994.

In situ capping, where contaminated sediments are covered by stable layers of “clean” materials and sediments, has been widely used in the US, Canada, and Norway, while the use in Sweden has been limited (Jersak et al., 2016). More “unconventional” treatment methods include

in-situ sediment treatment by activated carbon, where activated carbon is mixed into the bioactive sediment layer to adsorb and immobilize organic and inorganic contaminants. Pilot tests using this technique have shown varying results, e.g., Patmont et al. (2015) reported that transport of PCBs from sediment to surface water was almost entirely contained and reduced bioavailability by 90 % over three years. An assessment by Jing et al. (2018) showed remediation efficiencies varying from 60 to 92 % on PCB-contaminated soils and sediments treated by activated carbon.

In scenario 3, the southern end of the lake (0.27 km²) was dredged, decreasing the total mass of PCBs in the A-sediments by 75 %. Using in-situ capping or applying activated carbon on the entire A-sediment area after the dredging, we assumed a 75 % decrease in PCB diffusion from the sediments to surface water in the model. Due to the reduced sediment-water diffusion, the environmental half-life of PCBs in A-sediments increased to about 12 years (as compared to 9.3 years in scenario 1). Since the application of activated carbon reduces the bioavailability of PCBs but does not affect the absolute amount of PCBs in the sediments, predicted levels in perch in this scenario were most likely overestimated.

Direct measures, such as dredging or capping, demand lots of energy, material resources, and areas on land for disposal (Suer et al., 2004). In contrast, monitored natural recovery (MNR) relies on natural processes, where ongoing aquatic sedimentary and biological processes contain and reduce the bioavailability of contaminants (NRC, 1997). MNR is considered most applicable to sites, or portions of sites, where human or ecological risks are not immediate or substantial (Magar and Wenning, 2006). However,

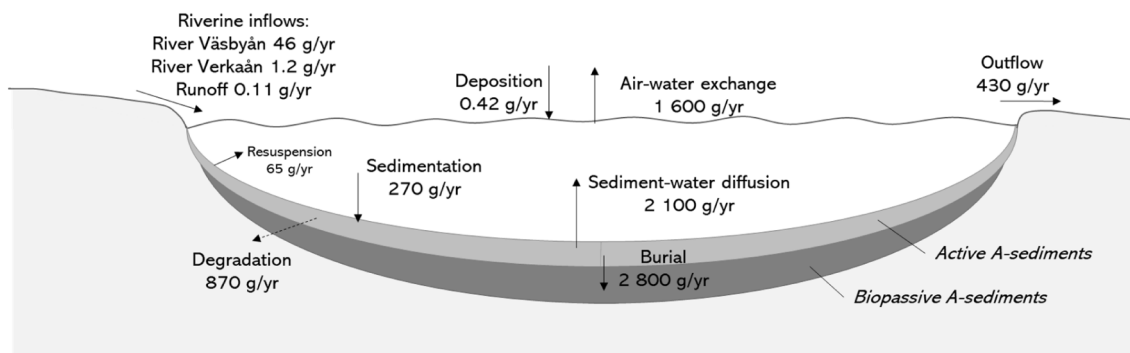


Fig. 8. Present process-related transports of ΣPCB₇ (g/yr) to, from and within Lake Oxundasjön from the mass balance model. The results correspond to the first year simulated in the model (i.e., about 2017).

Table 1

Predicted (25 years) mass and concentrations of ΣPCB_7 in water, active A-sediments and perch given the three different remediation scenarios. In Scenario 1, “Initial” values in the table represent present mass and concentrations. In Scenarios 2 and 3, “initial” values represent mass and concentrations directly after performed remediation actions. Errors of predicted values represents 95 % percent confidence intervals.

Compartment	Mass/concentration	Scenario 1		Scenario 2		Scenario 3	
		Initial	25 yrs	Initial	25 yrs	Initial	25 yrs
Water	Mass (g)	58	9.0 (5.7–14)	4.4	0.24 (0.15–0.38)	7.5	1.1 (0.69–1.7)
	Conc. (ng/l)	11	1.7 (1.1–2.7)	0.82	0.044 (0.028–0.070)	1.4	0.20 (0.13–0.32)
Sediment	Mass (g)	73,000	11,000 (7300–18,000)	750	130 (83–210)	18,000	4600 (3000–7400)
	Conc. (ng/g dw)	2900	460 (290–730)	30	5.2 (3.3–8.2)	730	180 (120–290)
Fish	Conc. (ng/g ww)	310	83 (38–150)	11	3.5 (1.6–6.2)	110	42 (19–76)

we can conclude that with no remediation measures in Lake Oxundasjön, levels in non-migratory fish will exceed existing environmental quality standards and marketing levels for PCBs in 25 years and more. Both in-situ and ex-situ removal or immobilization of PCBs can substantially reduce the amount of bioavailable PCBs in the system.

4.3. Model performance

Seasonal variations in water concentrations correlated to water temperatures and were inversely correlated to water discharge (Fig. 5), thus indicating a concentration of PCBs during periods of limited water turnover and higher temperatures. Similar patterns have been observed at other PCB-contaminated sites (Brown et al., 1985; Larsson et al., 1990). E.g., the sediment-water exchange of PCBs has been found to increase by a factor of 2–4 (Larsson et al., 1990) and 3–7 (Oliver, 1987) when temperatures increased from 10 to 20 °C and from 8 to 20 °C, respectively. Therefore, we consider the used calibration procedure to be validated. For future improvements, bottom water temperatures and concentrations would be helpful to predict sediment-water diffusion better. Field measurements of sediment diffusion (including bioturbation) and volatilization from surface water could also be valuable for model comparison and calibration.

The sensitivity analysis resulted in predicted (25 years) ΣPCB_7 -levels in small perch from 38 to 150 ng/g ww (95 % confidence interval), with an average value of 83 ng/g ww. Performed sensitivity analysis shows that the model is well-balanced and that the process-related assumptions made are not critical for the model's predictive power.

Although there are more sophisticated models for predicting content in biota where, e.g., bioaccumulation mechanisms in food-web interactions are considered (e.g., Thomann et al., 1991; Sun et al., 2018), the derived sediment-perch correlation ($p < 0.001$, Figs. S2 and 3) in the Stockholm region was considered most relevant to apply in the model. However, we stress that the correlation is derived from regional data and that the predicted levels are valid for small perch (15–20 cm). Since PCBs bioaccumulates, larger (and older) perch will assumably be more exposed. The same goes for fish in similar trophic levels as perch, as observed in northern pike from the lake (Fig. 4). Top predators consuming fish from the area, such as mink and birds of prey, are also at risk of being exposed to high levels of contaminants.

5. Conclusions

Based on the results from investigations presented in this paper, we could conclude:

- The contamination situation observed in the Lake Oxundasjön area states an excellent example of the persistent nature of PCBs in the environment. Although the substance group was banned in the 1970s, performed sampling in the area since 2013 has shown extensive contamination in lake water, sediment, biota, and adjacent air. To our knowledge, it is the most heavily PCB-contaminated site in Sweden.
- With the extensive dataset of PCBs acquired from different aquatic matrices, the system constitutes an ideal environment to test and develop a general mathematical mass balance model for PCBs. The presented

model approach is applicable for risk assessments of PCBs and contributes to the understanding of the transport and turnover dynamics of PCBs in aquatic ecosystems. However, the performed calibration is specific for the Lake Oxundasjön dataset and other parameter values might be motivated to change for different sites and conditions.

- The main load of PCBs to Lake Oxundasjön could have occurred from the end of the 1940s until 1980, reaching the lake via one of its tributaries. Mass balance modeling shows that diffusion from the lake sediments is presently the dominating source of PCBs to the system, causing elevated levels in predatory fish.
- Predictive modeling states a valuable concept for evaluating environmental profits from different remediation actions. In the case of Lake Oxundasjön, remediation actions are necessary to reduce the levels of PCBs in fish to or below today's marketing limits and environmental quality standards. With natural recovery only, the levels of PCBs in non-migratory fish from Lake Oxundasjön will be elevated for many decades to come.

CRedit authorship contribution statement

Joakim Hällén: Conceptualization, Methodology, Software, Writing-Original Draft **Mikael Malmaeus:** Writing – Review & Editing **Niklas Johansson:** Writing – Review & Editing **Magnus Karlsson:** Conceptualization, Supervision, Funding acquisition.

Data availability

No data was used for the research described in the article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.158522>.

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