Human Health Risk Assessment in Relation to Environmental Pollution of Two Artificial Freshwater Lakes in The Netherlands

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A human health risk assessment has been performed in relation to recreational activities on two artificial freshwater lakes along the river Meuse in The Netherlands. Although the discharges of contaminants into the river Meuse have been reduced in the last decades, which is reflected in decreasing concentrations of pollutants in surface water and suspended matter, the levels in sediments are more persistent. Sediments of the two freshwater lakes appear highly polluted and may pose a health risk in relation to recreational activities. To quantify health risks for carcinogenic (e.g., polycyclic aromatic hydrocarbons) as well as noncarcinogenic compounds (e.g., heavy metals), an exposure assessment model was used. First, we used a standard model that solely uses data on sediment pollution as the input parameter, which is the standard procedure in sediment quality assessments in The Netherlands. The highest intake appeared to be associated with the consumption of contaminated fish and resulted in a health risk for Pb and Zn (hazard index exceeded 1). For the other heavy metals and for benzo(a)pyrene, the total averaged exposure levels were below levels of concern. Secondly, input data for a more location-specific calculation procedure were provided via analyses of samples from sediment, surface water, and suspended matter. When these data (concentrations in surface water) were taken into account, the risk due to consumption of contaminated fish decreased by more than two orders of magnitude and appeared to be negligible. In both exposure assessments, many assumptions were made that contribute to a major degree to the uncertainty of this risk assessment. However, this health risk evaluation is useful as a screening methodology for assessing the urgency of sediment remediation actions. *Key words*: exposure assessment, freshwater lakes, health risk assessment, sediment pollution. *Environ Health Perspect* 107:27–35 (1999). [Online 7 December 1998]
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The river Meuse is located in the northwestern part of Europe and flows through three countries. It arises in France, proceeds through Belgium, and has its entrance into the North Sea in The Netherlands. The river Meuse is a pluvial river, characterized by low base flows in summer and relatively high peak flows in winter. The river responds very quickly to rainfall in its catchment area. High peak flows may result in flooding of the river, which is mainly due to the geomorphological composition of the river system. The river Meuse has many important functions, e.g., drinking water supply, water supply for industry and agriculture, shipping, power generation, recreation, and sustaining the ecological value (1,2). Furthermore, by excavation of minerals, e.g., gravel and sand, artificial freshwater lakes have been created along the river. These lakes are mainly situated in The Netherlands; some are in open connection with the river and others are disconnected.

The Meuse basin is contaminated by different pollutants, both of organic and inorganic nature. The most problematic are heavy metals, some organic micropollutants in relation with suspended matter [polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and pesticides], nitrogen and phosporus compounds, fluorides, cyanides, and chlorides (1,3,4). Only a small amount of the heavy metals originates from natural processes like weathering and erosion (5). For many metals and also for PAHs, emissions from anthropogenic sources exceed natural sources by far (5–8). Communal waste water and discharges by industry are major anthropogenic sources, but probably most important is diffuse pollution resulting from atmospheric deposition, shipping, river bank protection, road traffic, and agriculture.

In general, monitoring data on the river Meuse, generated by the Association of Rhine and Meuse water supply companies, indicate an improvement of the water quality over the last decades, in particular over the period 1974–1982 and 1983 (9). However, concentrations of various compounds still exceed Dutch and European Community guidelines for drinking water quality and the Dutch guideline for general environmental water quality. In river water, compounds such as PAHs, PCBs, pesticides, and heavy metals are primarily adsorbed to suspended matter and subsequently settle to the sediments. The concentrations of these compounds in suspended matter have also decreased over the last decades, although these still do not comply with the Dutch guidelines (1). By contrast, the chemical quality of the sediments has changed only to a minor extent.

The water quality of the river Meuse may influence the environmental quality of the artificial lakes along the river Meuse. Generally, in lake sediments the suspended matter/sediment is stored as a result of a significant decrease in velocity at the point of inflow into the lake and by the large water storage capacity relative to the inflow rate. The fine-grained suspended matter/sediment deposits in the deeper water region (5). Furthermore, as a result of high flow rates in the river, and consequently of the floodings, sediments may accumulate in the lakes. In 1989, various connected as well as disconnected lakes have been investigated. Of the 15 lakes investigated, in 8 lakes the sediments were highly contaminated by heavy metals (e.g., Ni, Cd, Zn, Cu, As) and organic micropollutants (e.g., PCBs and PAHs) (10). Additionally, the European Community guideline for bathing water quality (11) was frequently exceeded in these artificial lakes. These guidelines are based on physical–chemical parameters such as pH, aroma, oil, color and clarity, and bacteriological parameters such as thermotolerant coliforms and fecal streptococci. Mainly, standards for clarity and pH were exceeded.

The artificial lakes along the river Meuse are increasingly used for recreational purposes. In this respect, the pollution levels of these lakes may pose a risk to human health. During recreational activities, human exposure may occur via three different pathways: ingestion, inhalation, and dermal contact. Human exposure through dermal contact includes direct contact of the skin with contaminated sediments, with riverbank soils, or with water. Exposure through ingestion occurs through contaminated sediment, surface water during swimming, and via consumption of fish (12). Inhalation of contaminants volatilized from surface water and sediment may also occur. However, this pathway is of minor importance in relation to recreational activities (13,14). In general,

only a few studies, predominantly conducted in the area of the U.S. Great Lakes, have been performed to estimate human health risks in relation to recreational activities on freshwater lakes (13–18). Various studies have estimated health risks in relation to fish consumption from different lakes (13,19–22). Recreational or subsistence anglers may be at higher risk because they probably consume more fish, and consequently fish contaminants, than the general population (13). Next to ingestion of fish, the dermal route represents an important exposure pathway, in particular for organic compounds like DDT and most of the PAHs (12,14,23).

In this paper we describe a human health risk assessment as a function of recreational activities in relation to environmental pollution (e.g., by the heavy metals As, Cd, Cu, Pb, and Zn and 16 EPA-listed PAHs) of two artificial freshwater lakes, one connected to the river Meuse and the other currently disconnected from the river. An existing exposure assessment model, SEDISOIL, was applied to estimate human health risks (12). This model incorporates all relevant exposure pathways and is based on an exposure model to quantify health risk in relation to contaminated soils; it is currently used by the Dutch government as a guidance model to determine the urgency of remediation of sediment pollution. Two different methods were applied to estimate human exposure risks in relation to recreational activities: 1) human exposure risks were estimated by means of the standard SEDISOIL model, which solely uses data on sediment concentrations of relevant pollutants; and 2) location-specific input data derived from actual measurements of samples of surface water and suspended matter were incorporated in the model to refine the exposure model and to replace some default assumptions. In addition, we compare the measured concentrations of contaminants in sediment in both lakes to the previous study performed in 1989 and discuss the differences between the lakes in relation to the connection of the river Meuse.

Methods

Study Area

The location of the study area is shown in Figure 1. The artificial freshwater lake, Lake Eijsden, actually is a branch of the river Meuse. Lake Dilkensplas is an artificial freshwater lake that is currently disconnected from the river. In the mid-1960s at the beginning of the enclosure works, the lake had been connected to the river. Since 1974/1975, no further enclosure works have

been performed, but the lake was still connected to the river via a channel until the mid-1980s when the lake was disconnected from the river Meuse. At present, during high flow rates and consequently by the flooding of the river, polluted river sediment may still enter the lake. The maximum depth of Lake Dilkensplas is 14 m and the surface area is 14.8 ha. The surface area of Lake Eijsden is about 156 ha, and the mean depth is 6 m, with a maximum of 8 m. The lake has been in existence for approximately 20 years, and until 1995, enclosure works have been executed in a small part of the lake (24). Various recreational activities take place at both lakes. At Lake Eijsden, various beaches and swimming areas exist, and the lake is used by boaters, windsurfers, and recreational anglers. Yacht clubs and camping sites are located around the lake. Swimming, windsurfing, and fishing are the recreational activities that occur at Lake Dilkensplas, where a camping site is also located. During the summer season, 2,000 to 5,000 people visit Lake Eijsden and about 500 people visit Lake Dilkensplas. Furthermore, both lakes are eutrophic and have an ecological function (24).

Sampling

Sediment, surface water, and suspended matter samples were collected in the spring of 1995. Sediment samples (top 10 cm) were taken at the beach zone and from a boat at deeper parts of the lakes by a Van Veen grab. At each sample location within 1 m^2, three subsamples were taken and homogenized. A total of 27 sediment samples was collected from the freshwater Lake Eijsden and 16 samples from Lake Dilkensplas. The homogenized samples

were stored in clean polyethylene jars (2 liter). Upon arriving at the laboratory, the sediment samples were separated into subsamples for the respective pretreatment procedures.

The surface water samples were taken at the same location as the sediment samples and stored in polyethylene bottles (100 ml), which were previously cleaned with diluted nitric acid (5%). Water samples were acidified to pH <2 with concentrated nitric acid and centrifuged for 15 min at 2,500 rpm. The freshwater samples were stored at 4°C for further analysis.

Collection of suspended matter was only performed at the beach zone of the freshwater lakes. Suspended matter was sampled using polyethylene buckets. Five liters of surface water were collected. After the suspended matter settled, the water layer was filtered. In order to concentrate the amount of suspended matter, this procedure was repeated. In total, 50 l of surface water were sampled (10 buckets per site). The concentrated samples were stored in clean polyethylene jars (2 liter). Samples were centrifuged for 15 min at 3,000 rpm. After the supernatant was removed, the residue was dried with nitrogen.

Chemical Analysis

Heavy metals. The sediment samples were dried at 105°C, sieved (2 mm), and subsequently ground in an agate mortar. From each dried sample collected at two different zones at Lake Eijsden (beach zone and total lake) and Lake Dilkensplas, 30 g of dried sediment were taken, bulked, and successively passed through different sieves. The sediment and suspended matter samples (0.5 g) were boiled under
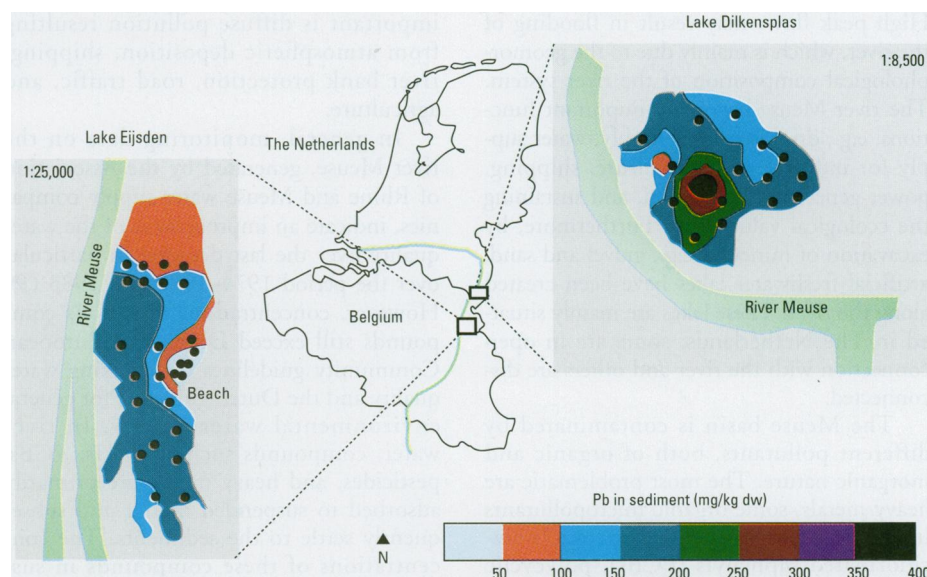

Figure 1. Map of the area showing the geographical variation of Pb in sediment of both lakes.

reflux for 2.5 hr in Aqua Regia and then filtered. The Cd, Cu, and Pb concentrations in sediment, suspended matter, and water were measured by graphite furnace atomic absorption spectrometry with Zeeman background correction, with the Zn concentration being determined by flame atomic absorption spectrometry. Determination of the arsenic content in water samples was performed by atomic absorption spectrometry applying the hydride generation technique. The As content of sediments and suspended matter was also determined by graphite furnace atomic absorption spectrometry with Zeeman background correction.

The detection limit of heavy metals in sediment/suspended matter was 0.18 mg/kg dry weight (dw) for As, 0.02 mg/kg dw for Cd, 0.36 mg/kg dw for Cu, 0.48 mg/kg dw for Pb, and 7.8 mg/kg dw for Zn. The detection limits in water were estimated to be 0.9 µg/l, 0.1 µg/l, 1.8 µg/l, 2.4 µg/l and 3.9 µg/l for As, Cd, Cu, Pb and Zn, respectively.

To prevent contamination, all used glass and plastic labware was previously washed in diluted nitric acid and deionized water.

Polycyclic aromatic hydrocarbons. The PAH concentration of the sediments was determined by a method described by Schilderman et al. (25). The method has been partly modified to quantify PAHs in sediments. The 16 EPA-listed PAHs [naphthalene, acenaphthylene, acenaphtene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene (BaP), benzo(ghi)pyrene, dibenzo(ah)anthracene and indeno(1,2,3-cd)pyrene] were measured except for acenaphthylene, which cannot be measured by fluorescence detection. Briefly, after filtration of the water layer, the wet sediment (10 g) was mixed with a suitable amount of anhydrous Na_2SO_4 and extracted with petroleum ether (40–60°C; 120 ml) in a

soxhlet apparatus for 4 hr. After the extract was concentrated under reduced pressure on a rotary evaporator at 45°C to approximately 50 ml, petroleum ether was added to a total of 100 ml. From this extract, 0.5 ml was dissolved in 1 ml acetonitrile and concentrated under nitrogen until the upper petroleum ether layer was evaporated; the sample was then analyzed by high performance liquid chromatography with fluorescence detection (25). The PAH concentration was quantified by comparison with selected standards. The detection limit of the individual PAHs in sediment was 8 µg/kg dw.

Soil characteristics. All sediment samples were analyzed for organic matter as loss on ignition by 550°C for 4 hr. The clay content of the sediment (particulate fraction <2 µm) was determined in seven samples at both artificial lakes. The samples were treated with H_2O_2 and HCl and then sieved over a 38-µm sieve, after which the fraction (<2 µm) was separated by a settling technique.

Risk Assessment

For quantification of exposure in relation to sediment contamination by heavy metals and PAHs, a multiple pathway exposure model (SEDISOIL) was used (12). The model was developed by the National Institute of Public Health and Environmental Protection and includes six exposure routes: the ingestion of sediment, surface water including suspended matter, dermal contact via surface water and sediment, and fish consumption. The following equations are incorporated into the model to calculate total exposure.

Ingestion of contaminated sediment (mg/kg/day)

$$= \frac{CS \times IRs \times EF \times AF}{BW}, \qquad (1)$$

where CS = concentration of the contaminant in sediment (mg/kg dw), IRs = ingestion rate of sediment (kg dw/exposure day),

EF = exposure frequency (days/365 days), AF = absorption factor (unitless), and BW = body weight (kg).

Ingestion of surface water (mg/kg/day)

$$= \frac{CW \times IRw \times EF \times AF}{BW}, \qquad (2)$$

where CW = concentration of the contaminant in surface water (mg/liter) and IRw = ingestion rate of surface water (liter/exposure day).

Ingestion of suspended material (mg/kg/day)

$$= \frac{CM \times CMW \times IRw \times EF \times AF}{BW}, \qquad (3)$$

where CM = concentration of the contaminant in suspended matter (mg/kg dw) and CMW = suspended matter content of surface water (kg/liter).

Dermal contact with contaminated sediment (mg/kg/day)

$$= \frac{CS \times SAs \times AD \times ASs \times Mf \times EDs \times EF \times AF}{BW}, \qquad (4)$$

where SAs = dermal surface area for sediment exposure (m^2), AD = dermal adherence rate for sediment (mg/cm^2), ASs = dermal absorption rate for sediment (liter/hr), Mf = matrix factor (unitless), and EDs = exposure duration from dermal exposure to sediment (hr/day).

Dermal contact with contaminated surface water (mg/kg/day)

$$= \frac{CW \times SAw \times ASw \times EF \times EDw \times AF}{BW} \qquad (5)$$

$$ASw = \frac{5000 \times (0.038 + 0.153 \times K_{ow})}{5000 + (0.038 + 0.153 \times K_{ow})}$$

$$\frac{\times \exp(-0.016 \times M)}{1.5}, \qquad (6)$$

where SAw = dermal surface area for exposure in surface water (m^2), ASw = dermal absorption rate for exposure in surface water [(mg/m^2)/(mg/liter)/hr], EDw = exposure duration from dermal exposure to surface water (hr/day), K_{ow} = octanol/water partition coefficient, and M = molecular weight (g/mol).

Ingestion of fish (mg/kg/day)

$$= \frac{CF \times IRf \times FI \times AF}{BW}, \qquad (7)$$

where CF = concentration of the contaminant in fish [mg/kg fresh weight (fw)], IRf = ingestion rate of fish (kg fw/day), and FI = fraction contaminated (unitless).

Table 1. Parameters used in the exposure model (12)

Parameter	Child	Adult	References
Ingestion rate of sediment (IRs) (kg dw/exposure day)	10^{-3}	0.35×10^{-3}	Assumption
Ingestion rate of fish (IRf) (kg fw/day)	0.015	0.055	(26,27)
Ingestion rate of surface water (IRw) (liter/exposure day)	50×10^{-3}	50×10^{-3}	(28)
Absorption factor (AF) (unitless)	1	1	Assumption
Dermal absorption rate (ASs) (liter/hr)	0.01	0.005	(29)
Dermal surface area for sediment exposure (SAs) (m^2)	0.17	0.28	(29)
Dermal surface area for exposure in surface water (SAw) (m^2)	0.95	1.80	(30)
Dermal adherence rate for sediment (AD) (mg/cm^2)	0.51	3.75	(29,30)
Matrix factor (Mf) (unitless)	0.15	0.15	(29)
Exposure frequency (EF) (days/365 days)	30	30	Assumption
Body weight (BW) (kg)	15	70	(30)
Suspended matter content of surface water (CMW) (kg/liter)	30×10^{-6}	30×10^{-6}	(31)
Exposure duration to sediment (EDs) (hr/day)	8	8	(29)
Exposure duration in surface water (EDw) (hr/day)	2	1	Assumption
Fraction contaminated (FI) (unitless)	0.5	0.5	(32)

Abbreviations: dw, dry weight; fw, fresh weight.

The default parameters listed in Table 1 (*12,26–32*) are those adopted by the Dutch National Institute of Public Health and the Environment and are incorporated in the standard exposure model (*12*). This model, in current use by the Dutch government, uses data on sediment pollution as input. The concentrations of contaminants in surface water and suspended matter are derived from these sediment concentrations by application of known equilibrium partitioning coefficients (K_d values). The concentration ratio between suspended matter and sediment is 1.5 for heavy metals and 2.0 for organic compounds (*12*). Furthermore, for the location-specific calculation procedure, data on heavy metal concentrations (mean values) from surface water and suspended matter (only Lake Eijsden) were directly used to calculate total average exposure. In general, amounts of sampled suspended matter were insufficient for reliably measuring PAH contents, which indicated that for PAH exposure assessment, only the standard model could be applied. For both exposure assessments, exposure via individual pathways and total exposure were calculated for both children and adults.

Calculated total heavy metal exposure levels were compared with the tolerable daily intake (TDI). The TDI refers to the reference dose of a substance that can be taken in daily without identifiable risk at lifetime exposure. Additionally, the hazard index was calculated, which refers to the ratio of the calculated lifetime daily exposure divided by the reference dose (TDI) (*16*). Daily exposure (milligrams per kilogram per day) averaged over a lifetime (e.g, 70 years) was calculated:

$$\frac{6 \times daily\ exposure_{child}}{70}$$

$$+ \frac{64 \times daily\ exposure_{adult}}{7}. \qquad (8)$$

If the hazard index is below 1, no health risk may occur. For Pb, a distinction was made between the TDI for adults and children because children are more sensitive to Pb than adults. The TDIs for heavy metals were derived from Bocking et al. (*12*).

For quantification of health risks in relation to PAH exposure, the TEF (toxic equivalency factor) approach was used (*33,34*). The TEF approach was designed to estimate the toxicity of complex mixtures for risk assessment purposes (*33,35*). PAH compounds were separated into two subclasses: carcinogens and noncarcinogens. Benzo(*a*)pyrene was assumed to be one of the most potent carcinogenic
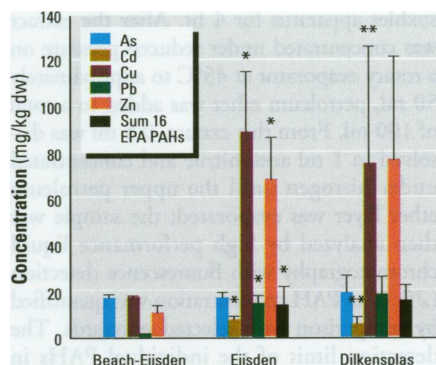

Figure 2. Concentrations (mean ± standard deviation) of some heavy metals and the sum of EPA-listed PAHs (mg/kg dw) in sediment samples from the artificial lakes Eijsden and Dilkensplas, The Netherlands. dw, dry weight. The concentrations of Pb and Zn were multiplied by 10. The statistical significance of differences were evaluated by the Mann-Whitney U-test.
*Significantly different from beach zone ($p<0.001$).
**Significantly different from Lake Eijsden ($p<0.05$).

PAH compounds. The cancer potencies of the various PAH compounds were compared with the cancer potency of BaP, after which a scaling factor was established. By this method, the evaluation of PAH mixtures can be expressed as BaP equivalents. Nisbet and Lagoy (*33*) proposed the following TEF factors for the 16 EPA-listed PAHs: dibenzo(*ah*)anthracene, 5; BaP, 1; benzo(*a*)anthracene, 0.1; benzo(*b*)fluoranthene, 0.1; benzo(*k*)fluoranthene, 0.1; indeno(*1,2,3-cd*)pyrene, 0.1; anthracene, 0.01; benzo-(*ghi*)perylene, 0.01; chrysene, 0.01; acenaphthene, 0.001; acenaphthylene, 0.001; fluoranthene, 0.001; fluorene, 0.001; naphthalene, 0.001; phenanthrene, 0.001; and pyrene, 0.001. The BaP equivalent concentration is the sum of the weighted potency factor of each compound in the mixture multiplied by the concentration of the compound in the mixture. The carcinogenic risks associated with exposure to BaP equivalents averaged over a lifetime (e.g., 70 years) were calculated and compared to the Dutch maximum permissible individual risk level (MPR), which corresponds to an annual probability to die as a consequence of cancer of 10^{-6}.

Statistical Analyses

The Spearman rank correlation coefficient was used to determine relationships between heavy metal, PAH contents in sediment, and bottom soil characteristics, and to correlate mutual data. A $p<0.05$ was considered statistically significant.

Statistical differences between the lakes with respect to bottom soil characteristics, heavy metal content in sediment and surface water, and PAH concentration in sediments were analyzed by the Mann-Whitney U-test. Again, $p<0.05$ was considered statistically significant. For statistical purposes, all values
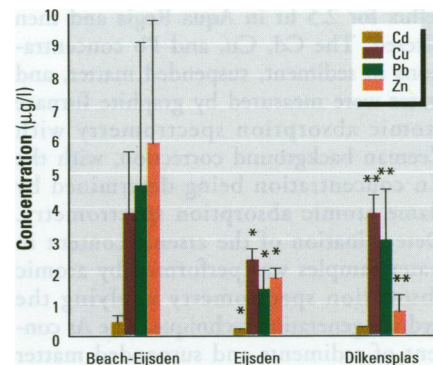

Figure 3. Concentrations (mean ± standard deviation) of some heavy metals (µg/l) in surface water samples from the artificial freshwater lakes Eijsden and Dilkensplas, The Netherlands. The concentration of Zn was multiplied by 10. The statistical significance of differences have been evaluated by the Mann-Whitney U-test.
*Significantly different from beach zone ($p<0.001$).
**Significantly different from Lake Eijsden ($p<0.05$).

below the detection limit were replaced by half the detection limit.

Results

Chemical Analysis

Figure 2 shows As, Cd, Cu, Pb and Zn contents as well as the sum of the 16 EPA-listed PAHs in sediment samples collected at the two freshwater lakes. Results from the beach zone and the total lake with respect to Lake Eijsden are shown separately. The heavy metal and PAH concentrations varied between sampling sites, but in general, comparable levels were observed in sediment samples taken from Lake Eijsden (total) and Lake Dilkensplas.

Considerable variations in sediment heavy metal and PAH contents existed in each lake. The sum of the EPA-listed PAHs in Lake Eijsden (total) and Lake Dilkensplas varied from 2.6 to 29.9 mg/kg dw and 0.89 to 25.5 mg/kg dw, respectively. In Lake Eijsden (total), the sediment values were 14–22 mg/kg dw for As, 2–10 mg/kg dw for Cd, 29–115 mg/kg dw for Cu, 39–191 mg/kg dw for Pb, and 149–970 mg/kg dw for Zn. In Lake Dilkensplas, the concentrations of heavy metals were 11–47 mg/kg dw for As, 1–20 mg/kg dw for Cd, 15–237 mg/kg dw for Cu, 48–430 mg/kg dw for Pb, and 188–2,080 mg/kg dw for Zn. The highest heavy metal concentrations were measured in sediment samples collected from the center of Lake Dilkensplas, and were approximately 2–4 times above the mean levels in the lake. For example, the Pb variation between sampling sites is shown in Figure 1.

At the beach zone of Lake Eijsden, the heavy metal content and the sum of EPA-listed PAHs in sediment were remarkably

Table 2. Carcinogenic risk and average total exposure levels (μg/kg/day) for benzo(a)pyrene (BaP) equivalents

BaP equivalent	Lake Eijsden Standard		Lake Dilkensplas Standard	
	Adult	Child	Adult	Child
Ingestion of sediment	1.2×10^{-3}	1.6×10^{-2}	1.4×10^{-3}	1.9×10^{-2}
Ingestion of surface water	3.3×10^{-6}	1.5×10^{-5}	3.9×10^{-6}	1.8×10^{-5}
Ingestion of suspended matter	1.5×10^{-5}	5.4×10^{-5}	8.3×10^{-6}	3.9×10^{-5}
Dermal contact with sediment	2.2×10^{-4}	1.7×10^{-4}	2.6×10^{-4}	2.0×10^{-4}
Dermal contact with surface water	6.9×10^{-3}	3.4×10^{-2}	8.2×10^{-3}	4.0×10^{-2}
Ingestion of fish	1.1	1.3	1.3	1.6
Total	1.1	1.4	1.3	1.7
Carcinogenic risk	5.6×10^{-7}		6.6×10^{-7}	

low (Fig. 2). Mann-Whitney U-test analysis showed a significant difference between sediment PAH and heavy metal content (except for As) at the beach zone of Lake Eijsden and Lake Eijsden (total). Differences in PAH and heavy metal concentrations in sediment between Lake Eijsden (total) and Lake Dilkensplas were minimal; only Cd and Cu concentrations were significantly lower in Lake Dilkensplas.

The individual PAH levels (except for fluorene) in sediment at the beach zone of Lake Eijsden were significantly lower in comparison to the sediment levels in Lake Eijsden (total) (Mann Whitney U-test, $p<0.05$). The profile of the individual PAHs in sediment of Lake Dilkensplas was similar to that of Lake Eijsden (total). Relatively high concentrations of 2-ring PAHs were observed. The mean levels of the individual PAHs in sediment from Lake Dilkensplas were higher compared to the sediments in Lake Eijsden. Only sediment levels of fluorene, phenanthrene, benzo(a)anthracene, dibenzo(ah)anthracene, and chrysene were statistically different between the lakes.

The levels of heavy metals in surface water are shown in Figure 3. No detectable concentrations of As were found in the water samples. Furthermore, in some water samples the Pb and Zn concentrations were below the detection limit. The highest levels were found in surface water samples taken at the beach zone of Lake Eijsden. Mann-Whitney U-test analysis demonstrated a significant difference between Cd, Cu, Pb, and Zn levels in Lake Eijsden (total) and the surface water levels at the beach zone ($p<0.05$).

Additionally, the Cu, Pb and Zn levels in surface water from Lake Dilkensplas were significantly different from the surface water concentrations in Lake Eijsden (total) ($p<0.05$). No significant correlation was observed between the heavy metal contents in sediment and surface water in either lake.

The heavy metal contents in suspended matter were only determined at Lake Eijsden. Two samples were taken and the mean concentrations were 18 ± 2, 6 ± 1, 65 ± 5, 130 ± 7, and 806 ± 31 mg/kg dw for

As, Cd, Cu, Pb, and Zn, respectively. Due to the limited sample yield, it was not possible to analyze EPA-listed PAHs and soil characteristics such as organic matter and clay fraction in the suspended material. Furthermore, the amounts of suspended matter collected at Lake Dilkensplas were not sufficient for analysis. The total amount of suspended matter was 33 mg/l in Lake Eijsden and 20 mg/l in Lake Dilkensplas.

The Dutch intervention value for Zn was exceeded in more than 80% of the sediment samples from both lakes. The sediment levels of other heavy metals exceeded the intervention value incidentally. The intervention value indicates an unacceptable risk to man or environment due to soil or sediment contamination and is normalized for the clay (25%) and organic matter (10%) content of the soil/sediment. In most surface water samples collected at the beach zone of Lake Eijsden, the Dutch short-term quality standard for Cd, Cu, and Zn was exceeded. For the total lake, the concentrations of heavy metals in surface water met the long-term quality standard (e.g., Cu and Pb) and were below the short-term quality standard for Zn and Cd. The concentrations of Cu and Cd (in 30% of the samples) in surface water from Lake Dilkensplas exceeded the short-term quality standard. The Pb and Zn concentrations in Lake Dilkensplas met the long-term quality standard. The short- and long-term quality standards represent the general environmental water quality. The long-term quality standard refers to an acceptable or natural concentration, and the short-term standard directs to an acceptable risk level.

For Lake Dilkensplas, a significant correlation was observed between the organic matter and clay fraction in sediment and the heavy metal content ($p<0.05$). Furthermore, sediment heavy metal levels correlated significantly to each other. No significant correlation was found between sediment levels of EPA-listed PAHs and the organic matter and clay fraction in sediment. Additionally, for Lake Eijsden (total), the data showed a significant correlation between heavy metal

content in sediment and organic matter (except for As). Furthermore, the heavy metal levels in sediment correlated to each other (except for As/Cu, As/Pb, and As/Zn). Moreover, the sum of EPA-listed PAHs in sediment significantly correlated to the organic matter fraction in sediment. For the beach zone of Lake Eijsden, no appreciable correlations were found.

Risk Assessment

The BaP equivalent concentrations [mean ± standard deviation (SD)] in sediments from the different lakes were 3.0 ± 1.7 mg/kg dw [Lake Eijsden (total)], 0.01 ± 0.03 mg/kg dw (beach zone of Lake Eijsden), and 3.5 ± 1.3 mg/kg dw (Lake Dilkensplas). Spearman rank correlation coefficient (R_s) analysis demonstrated a significant correlation between the sum of EPA-listed PAHs and the BaP equivalent concentration in sediments of Lake Dilkensplas ($p<0.01$; R_s = 0.82) and in sediments of Lake Eijsden (total) ($p<0.0001$; R_s = 0.90). For the beach zone of Lake Eijsden, no significant correlation was observed.

Tables 2 and 3 summarize the results of the health risk assessment for adults and children exposed to contaminants in the freshwater lakes Eijsden and Dilkensplas. The heavy metal and BaP equivalent concentrations in sediment at the beach zone for Lake Eijsden were relatively low, and the results of the risk assessment calculations applying the standard model indicate that there was no potential human health risk (data not shown).

After modeling the standard model, the calculated average exposure levels for Zn and Pb as present in both lakes exceeded the TDI, which implies that chronic health effects may occur. The calculated average carcinogenic risk for BaP equivalents as present in both lakes were below the maximally tolerated level of exposure to genotoxic carcinogens. For both lakes, modeling human exposure, taking location-specific input data into account, indicated that the calculated total average exposure levels for As, Cd, Cu, Pb, and Zn were below the levels of concern for human health (the hazard indices were far below 1). Therefore, no significant health risks are expected in relation to recreational activities on freshwater lakes.

For both lakes, exposure through the ingestion of fish by adults and through the ingestion of sediment by children comprised the majority of exposure risk to heavy metals. Dermal exposure to surface water appeared to be an important exposure route for BaP if exposure through the ingestion of fish was not included in the risk assessment.

The calculated averaged total exposure levels to heavy metals were much higher in

Table 3. Hazard index and average total exposure levels of heavy metals estimated by the standard model and by incorporating location-specific data (μg/kg/day)

	Lake Eijsden				Lake Dilkensplas			
	Standard		Location specific		Standard		Location Specific	
	Adult	Child	Adult	Child	Adult	Child	Adult	Child
As								
Ingestion of sediment	6.7×10^{-3}	9.3×10^{-2}	6.7×10^{-3}	9.3×10^{-2}	7.8×10^{-3}	0.1	7.8×10^{-3}	0.1
Ingestion of surface water	1×10^{-3}	4.8×10^{-3}	5.3×10^{-5}	2.5×10^{-4}	1.1×10^{-3}	5.3×10^{-3}	5.3×10^{-5}	2.5×10^{-4}
Ingestion of suspended matter	4.9×10^{-5}	2.3×10^{-4}	3.5×10^{-5}	1.6×10^{-4}	3.4×10^{-5}	1.6×10^{-4}	3.4×10^{-5}	1.6×10^{-4}
Ingestion of fish	3.4×10^{-2}	4.3×10^{-2}	1.8×10^{-3}	2.2×10^{-3}	3.8×10^{-2}	4.9×10^{-2}	1.8×10^{-3}	2.2×10^{-3}
Total	4.2×10^{-2}	0.14	8.8×10^{-3}	9.6×10^{-2}	4.7×10^{-2}	0.16	9.7×10^{-3}	0.10
Hazard index[a]	0.02		0.008		0.03		0.008	
Cd								
Ingestion of sediment	2.9×10^{-3}	3.8×10^{-2}	2.9×10^{-3}	3.8×10^{-2}	2.1×10^{-3}	2.7×10^{-2}	2.1×10^{-3}	2.7×10^{-2}
Ingestion of surface water	2.2×10^{-3}	1.0×10^{-2}	8.8×10^{-6}	4.1×10^{-5}	1.5×10^{-3}	7.2×10^{-3}	1.1×10^{-5}	4.9×10^{-5}
Ingestion of suspended matter	2.0×10^{-5}	9.5×10^{-5}	1.6×10^{-5}	5.4×10^{-5}	8.8×10^{-6}	4.1×10^{-5}	8.8×10^{-6}	4.1×10^{-5}
Ingestion of fish	0.14	0.18	5.6×10^{-4}	7.5×10^{-4}	0.10	0.13	7.1×10^{-4}	9.0×10^{-4}
Total	0.15	0.23	3.5×10^{-3}	3.9×10^{-2}	0.11	0.17	2.8×10^{-3}	2.8×10^{-2}
Hazard index	0.16		0.006		0.11		0.005	
Cu								
Ingestion of sediment	3.6×10^{-2}	0.48	3.6×10^{-2}	0.48	3.1×10^{-2}	0.41	3.1×10^{-2}	0.41
Ingestion of surface water	9.3×10^{-3}	4.4×10^{-2}	1.4×10^{-4}	6.3×10^{-4}	8.2×10^{-3}	4.1×10^{-2}	2.2×10^{-4}	1.0×10^{-3}
Ingestion of suspended matter	2.6×10^{-4}	1.2×10^{-3}	1.3×10^{-4}	5.9×10^{-4}	1.3×10^{-4}	6.2×10^{-4}	1.3×10^{-4}	6.2×10^{-4}
Ingestion of fish	6.3	8	9.0×10^{-2}	0.11	3.0	3.8	0.15	0.19
Total	6.3	8	0.13	0.60	3.0	4.2	0.18	0.60
Hazard index	0.05		0.001		0.02		0.002	
Pb								
Ingestion of sediment	5.9×10^{-2}	0.79	5.9×10^{-2}	0.79	7.5×10^{-2}	1.0	7.5×10^{-2}	1.0
Ingestion of surface water	3.5×10^{-3}	1.6×10^{-2}	5.3×10^{-5}	3.8×10^{-3}	4.5×10^{-3}	2.1×10^{-2}	1.7×10^{-4}	7.8×10^{-4}
Ingestion of suspended matter	4.2×10^{-4}	1.9×10^{-3}	3.5×10^{-4}	1.9×10^{-3}	3.2×10^{-4}	1.5×10^{-3}	3.2×10^{-4}	1.5×10^{-3}
Ingestion of fish	12	15	0.28	0.35	15	18	0.57	0.73
Total	12	16	0.33	1.14	15	19	0.64	1.7
Hazard index	1.7–4.4		0.04–0.3		2.1–5.3		0.1–0.5	
Zn								
Ingestion of sediment	0.28	3.73	0.28	3.73	0.32	4.2	0.32	4.2
Ingestion of surface water	0.16	0.7	1.0×10^{-3}	4.7×10^{-3}	0.18	0.83	4.3×10^{-4}	2.0×10^{-3}
Ingestion of suspended matter	2.0×10^{-3}	9.2×10^{-3}	1.6×10^{-3}	7.3×10^{-3}	1.3×10^{-3}	6.3×10^{-3}	1.3×10^{-3}	6.3×10^{-3}
Ingestion of fish	1,060	1,360	6.7	8.7	1,200	1,520	2.9	3.7
Total	1,060	1,365	7.1	12	1,200	1,525	3.2	7.8
Hazard index	1.08		0.007		1.2		0.007	

[a]Hazard index refers to the ratio of the calculated lifetime daily exposure divided by the reference dose (tolerable daily intake; TDI). For lead, a distinction has been made between the TDI for adults and children because children are more sensitive to lead than adults.

the standard exposure model, in comparison to the calculated averaged exposure levels by taking location-specific data into account. The difference in exposure levels was mainly due to the ingestion of fish and the ingestion of surface water while swimming.

Discussion

Environmental Monitoring

Heavy metals and PAH levels determined in sediment in the two artificial freshwater lakes in this study were not significantly different from those determined 7 years previously in the same lakes (10). An exception was Cd. The mean levels in sediment in both lakes were decreased, by 15% for Lake Eijsden and by 50% for Lake Dilkensplas, as compared to 1989. However, at that time, only one sample was taken from Lake Dilkensplas and two samples (beach zone and deeper part) from Lake Eijsden. Although the chemical quality of the surface water, including suspended matter, of the river Meuse has improved over the last

decades, the quality of the lake sediments for both lakes in general has changed only to a minor extent during the previous years.

The quality of the river Meuse may influence the quality of the freshwater lakes along the river. Although Lake Dilkensplas is now disconnected from the river, sediment/sludge may accumulate into the lake as a result of flooding of the river. Lake Eijsden is connected to the river and therefore directly affected by the river. Since 1989, two flooding disasters have occurred along the Belgium and Dutch part of the river, at the end of December 1993 and at the end of January 1995, and sedimentation of polluted suspended matter and sediments have occurred. However, comparable results were found in sediment samples taken from both lakes. This suggests that a direct or indirect connection to the river Meuse appears less important for the pollution of both freshwater lakes; sediment contamination presumably reflects historical pollution.

The concentrations of heavy metals (except for As) and PAHs in sediment were

significantly lower at the beach zone of Lake Eijsden as compared to the total lake. After the flooding of the river Meuse in January 1995, the sludge was removed and new sand was pumped onto the beach zone of Lake Eijsden. The more sandy sediment and low organic matter content (2%) also contributed to the lower heavy metal and PAH concentrations in the sediment at the beach zone (36). The As levels represent background levels for both sites in the lake (beach and total).

When comparing lake sediment concentrations, heavy metals significantly correlated with each other. Diffuse sources (for example, atmospheric deposition, industrial discharges, and road traffic) may contribute to the heavy metal load in both lakes; furthermore, postdepostional remobilization may occur (5). To identify the main source, geochemical and paleolimnological studies must be performed (5). In addition, heavy metals correlated significantly with organic matter and clay fraction of the sediments (only Lake Dilkensplas),

which indicates that heavy metals may be adsorbed by Fe hydroxides, Fe/Mn oxides, or carbonates (37). Surfaces of these particles are negatively charged, and interaction may occur with the positively charged heavy metals. Additionally, other factors may influence sorption and desorption processes related to the properties of sediment (e.g, grain size, pH, temperature, water content, cation exchange capacity, salt concentration), the properties of water (e.g., pH), and the properties of the chemical itself (chemical speciation and water solubility) (38,39). The adsorption to these compounds influences the potential availability to biota (5,39). Because heavy metals tend to adsorb to clay and organic matter in sediment, the Dutch environmental standards for sediment and soil are normalized for the clay content (25%) and organic matter content (10%). Results from this study indicate that in both lakes, the intervention value for Zn (environmental standards) normalized to organic matter and clay fraction was exceeded in more than 80% of the sediment samples. Other heavy metals incidentally exceeded these environmental standards.

PAHs are primarily adsorbed on organic matter (38), and a significant correlation was observed between the organic matter content of the sediments and the sum of EPA-listed PAHs for Lake Eijsden (total). However, for Lake Dilkensplas no significant correlation was found.

No correlation was observed between the heavy metal concentration in sediment and the surface water concentration for both lakes. We measured the total concentration (dissolved and particulate forms) of heavy metals in surface water. In river systems As, Cd, Cu, and Zn appear to occur in 10–50% as the dissolved fraction and Pb appears in 1–10% dissolved (5,39). The monitoring network at the river Meuse, managed by the Association of Rhine and Meuse water supply companies, determines total concentration of heavy metals in surface water (9). During the same sampling period, higher surface water concentrations of Pb and Zn (total concentration) were observed in the river Meuse at the monitoring station at Lake Eijsden in comparison to the total Pb and Zn concentrations measured in the surface water in Lake Eijsden. The concentrations of heavy metals in the river Meuse are more influenced by industrial discharges, which result in an increase of suspended matter concentrations (5).

Concentrations of heavy metals in surface water at the beach zone of Lake Eijsden were significantly higher in comparison to concentrations in the lake itself. This was probably due to the sampling strategy at the beach zone of Lake Eijsden. These samples

were taken while standing in the water (depth, 1 m), and we assume that the surface sediments were disturbed; therefore, relatively more suspended material was sampled. Because the water samples were directly acidified without filtration in the laboratory, consequently higher heavy metal concentrations were found in the surface water from the beach zone of Lake Eijsden.

Risk Assessment

There are only a few reports on possible health risks in relation to recreational activity on freshwater lakes or estuaries, in particular in an estuary in the Tidewater region of Virginia, and from areas of the Great Lakes in the United States and Canada (13–18). These studies identify ingestion of contaminated fish as an important exposure pathway in relation to exposure to PCBs, PAHs, and heavy metals in sediments. In these studies, PCB exposure accounted for the excess cancer risk, which was mainly due to the consumption of crab (Tidewater region) (14) and carp (Great Lakes) (15). The noncarcinogenic risks were of minor importance, except for subsistence anglers in the Great Lakes area, where exposure to heptachlor epoxide (17), dieldrin (18), and methyl mercury and copper contaminations (16,17) exceeded the health-based standards.

The results of this health risk assessment indicate that sediment contamination by Pb and Zn in both lakes may present a health hazard, if the risks are calculated on the basis of the standard exposure model. However, by taking location-specific data into account, including data of actually measured contaminant concentrations in surface water and suspended matter, in the estimation of exposure levels, the health hazards were insignificant. Furthermore, the additional exposure through recreational activities on both freshwater lakes appeared to be much lower than background values; an exception was Pb, for which the additional exposure levels contributed for approximately 100% (adults) and 300% (children) to background exposure levels. Background exposures to As, Cd, Cu, Pb, and Zn have been estimated to be approximately 0.18, 0.26, 21, 0.46, and 190 µg/kg/day, respectively (40).

In the present study, measured concentrations of heavy metals in surface water were remarkably lower than the predicted concentrations. Ratios of measured versus predicted concentrations in surface water for Lake Eijsden were 4.2×10^{-3}, 0.014, 0.024, and 6.2×10^{-3} for the heavy metals Cd, Cu, Pb, and Zn, respectively. For Lake Dilkensplas, the ratios were in the same range. The standard model overestimated heavy metal concentrations in surface water

by more than two orders of magnitude. However, the relative contribution of surface water ingestion during swimming to the total average exposure levels of heavy metals was small. The total average exposure was dominated by the expoure through ingestion of fish (adults) or through ingestion of sediment (children).

Various assumptions applied in this risk assessment contribute to uncertainty about the estimated risks: (e.g., exposure frequency and durations, ingestion rate of sediments and contaminated fish, the concentration in surface water and suspended matter, and the fish content and absorption factors) (13,14). For example, it was assumed that exposure levels, exposure duration, and exposure frequency do not change in a lifetime and that human activities remain the same. It is apparent that the probability of this is extremely small. To illustrate this, people spend only a fraction of their lifetime in any one location (41).

In a conservative approach, it is assumed that 100% of the compound in the ingested sediment or fish is sorbed from the gastrointestinal tract (12). Compound-specific absorption factors for ingestion exposure routes in the present study are not available for all compounds (12). Dermal absorption depends on the dermal absorption rate for the individual compounds. For example, in the exposure model a dermal absorption rate of 0.5%/hr (adult) and 1%/hr (child) for organic compounds and 0%/hr for inorganic compounds has been used to estimate dermal exposure through sediment exposure (12).

The contaminant concentration in fish is modeled by the bioconcentration factor (BCF) and the concentration in surface water of the contaminant. The heavy metal concentrations in surface water were overestimated by more than two orders of magnitude; therefore, the concentrations in fish were also overestimated by more than two orders of magnitude, which had a remarkable influence on total average exposure levels in relation to sediment pollution for heavy metals.

Also, different physical, chemical, and physiological parameters influence the BCF. Examples are the age of the fish, the species, chemical speciation, dietary habits of the fish, and the physical and chemical properties of sediments (5,13,42–44). Concentrations in fish estimated by a mean BCF value are therefore of limited accuracy (12).

In addition, no information on the amount and frequency of fish caught at both lakes and consumed by recreational anglers was directly available. For freshwater recreational anglers on lakes and ponds, Finley et al. (45) recommend a fish consumption rate

of 1.7 g/day as the median value and 15 g/day as the 95th percentile as default values for risk assessment. To estimate human health risks for recreational anglers and their families with regard to consumption of contaminated fish from the Great Lakes, a fish ingestion rate of 54 g/day has been assumed, from which 25% is self-caught (16–18). In the exposure model applied in this study, the default fish ingestion rate was 55 and 15 g/day for adults and children, respectively, considering 50% of the fish consumption self-caught (12). This assumption (25 g/day for adults) is at the extreme of the exposure distribution recommended by Finley et al. (45). Furthermore, the concentrations in fish were based on uncooked whole fish instead of the edible parts of the fish, which also represented conservative assumptions (15). Therefore, exposure through ingestion of contaminated fish was highly overestimated by using these conservative assumptions.

Ingestion of sediment was considered an important exposure route if exposure was calculated based on location-specific input data. However, no specific data on the sediment ingestion rate by children and adults were available for risk assessment purposes. Therefore, sediment ingestion rates are based on soil ingestion rates in relation to contaminated sites (12). In the standard exposure model, a sediment ingestion rate of 1 g/day (82 mg/day annual average) for children has been used as a default value. This assumption seems to be plausible, but conservative. Finley et al. (45) recommended a default value of 110 mg/day as the 95th percentile and 16 mg/day as the median value for the child soil ingestion rate.

The other exposure routes were of minor importance in relation to total exposure levels for heavy metals. For example, the ingestion of suspended matter while swimming contributed less than 1% to the total average exposure levels. The concentration of heavy metals in suspended matter was also modeled on the basis of partition coefficients. Equilibrium was assumed between surface water and suspended matter (12). The predicted ratios versus measured concentrations in suspended matter for Lake Eijsden were 0.71, 0.57, 0.49, 0.60, and 0.79 for the heavy metals As, Cd, Cu, Pb, and Zn, respectively. The predicted values were slightly higher, but have no influence on the total average exposure levels.

To estimate human risks in relation to contaminated PAH sediments we used the TEF approach. In this approach, the sum of EPA-listed PAHs in sediments were expressed as BaP equivalents to calculate the carcinogenic risk associated with exposure to a mixture of PAH compounds (33). The ingestion of fish was the predominant

exposure route due to sediment contamination with BaP. Exposure to lower molecular weight PAHs in sediments was due to dermal exposure of surface water while swimming and ingestion of contaminated fish. For higher molecular PAHs (log K_{ow} range 5–6.2), exposure was due to the ingestion of contaminated fish, and for the very high molecular PAHs (log K_{ow} >6.2), exposure was through dermal exposure to sediments and the ingestion of fish (11). The very high molecular PAHs (log K_{ow} >6.2), predominantly dibenzo(ah)anthracene, contributed more than 75% to the total BaP equivalents in sediments. This indicates that the exposure levels of BaP for the pathways of ingestion of contaminated fish and dermal exposure while swimming were overestimated because the very high molecular PAHs are less soluble in water, which is reflected by lower surface water concentrations and lower concentrations in fish.

In summary, the application of the standard exposure model, which is currently used by the Dutch government, leads to the conclusion that sediment contamination by Pb and Zn may cause a health risk in relation to recreational activities on both lakes, which is mainly due to the exposure through ingestion of fish. However, upon inclusion of location-specific data, in particular the concentrations in surface water, the exposure risk estimates are reduced by more than two orders of magnitude. We therefore recommend that location-specific data should be used if exposure through ingestion of contaminated fish is a relevant exposure pathway. Furthermore, calculated risks do not represent actual risks due to the incorporation of conservative assumptions. In general, exposure risk assessment of the present type can be used for screening purposes to identify important exposure pathways and to determine the urgency of sediment remediation actions.

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