Journal of Hazardous Materials 171 (2009) 200–207

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Exposure to contaminated sediments during recreational activities at a public bathing place

Monika Filipsson, Marianne Lindström, Pasi Peltola, Tomas Öberg*

School of Pure and Applied Natural Sciences, University of Kalmar, SE-391 82 Kalmar, Sweden

ARTICLE INFO

Article history:
Received 25 November 2008
Received in revised form 25 May 2009
Accepted 27 May 2009
Available online 6 June 2009

Keywords:
Exposure assessment
Exposure factors
Recreational activities
Variability
Uncertainty

ABSTRACT

More and more time is spent on recreational activities, but few risk assessments focus specifically on these situations and exposure factor data are often scarce. To assess exposure to contaminants at a public bathing place in an urban environment, we have compiled literature data, conducted observation studies, and analyzed water and sediment samples. The levels of anthropogenic contaminants are high in urban environments and traffic frequently plays an important role. In this study, to characterize variability and uncertainty, the deterministic exposure calculations for metal pollutants were supplemented by a probability bounds analysis for the polycyclic aromatic hydrocarbons (PAH). The results from these calculations show that oral intake is the major exposure route for metals, while skin absorption, with present assumptions, is more important for the PAH. The presently measured levels of contaminants, at this public bathing place, cannot be anticipated to cause any significant adverse influence on public health. This assessment methodology is easy to adapt and can be used routinely in other situations with more heavily contaminated surface sediments and lake water.

1. Introduction

Lake Trekanten in Stockholm, the capital of Sweden, is an example of a heavily polluted lake where recreational activities could potentially be harmful, due to the contaminants present in sediments. The City of Stockholm and the District Administration of Liljeholmen have therefore commissioned a risk assessment for bathing activities in the lake. A previous interview investigation by the Stockholm Environment and Health Administration showed that pollution from traffic, due to surface water runoff, was perceived by stakeholders as the largest risk for the lake.

Recreational activities, although limited in time, are usually associated with strong emotional aspects and children are often the most exposed target group. However, only a few risk assessments have been reported in the literature that specifically deals with bathing, swimming, and other recreational activities at polluted shores and beaches.

The wreckage of the oil tanker ‘Erika’ in 1999 polluted 400–500 km of the coast of Brittany (France). Two risk assessments have been published from this incident, and both assessed cleaning as well as tourist activities [1,2]. The calculations were in both cases deterministic (point estimates) and based on exposure factors from the literature, supplemented by assumptions about exposure time (duration), frequency, intakes of water and sediment, and skin contact. Although the accident resulted in heavy pollution, the clean-up was successful and exposure from subsequent recreational activities was insignificant.

Health risks may also occur—during recreational activities—from direct or indirect exposure to contaminated sediments. Elevated levels of metals and organic pollutants are often found in lake, river, and marine sediments in urban and industrialized areas. In a probabilistic health risk assessment for exposures to estuary sediments contaminated by polychlorinated biphenyls, the general exposure factors from literature were supplemented by site-specific information [3]. The probability distributions, however, were in most cases derived from limited data or based on judgment, and uncertainty was not separated from variability. This study included several exposed population groups and the pathways associated with the highest intake in this study were dermal contact with the contaminated sediments and ingestion of contaminated biota. Similar results were obtained in another study, with recreational beach activities, but where swimming was considered unlikely [4]. In this deterministic assessment, fish consumption and dermal contact were the major exposure routes.

Another human health risk assessment of recreational activities, along two contaminated lakes in the Netherlands, also indicates that fish consumption is a major exposure route [5]. Furthermore, applying a standard model indicated that the contamination could present a health hazard, but this was subsequently contradicted when location-specific data were taken into account. The need for location-specific data in our study of Lake Trekanten was therefore apparent.

* Corresponding author. Tel.: +46 480 44 62 47.
E-mail address: tomas.oberg@hik.se (T. Öberg).

0304-3894/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2009.05.128

The initial step in our study was to frame and delimit the risk assessment task, and define the data requirements. Previous investigations of the pollution in Lake Trekanten have focused on sampling and analysis of contaminants in sediments from deeper parts of the lake. Additional sampling at the public bathing place and of lake water therefore seemed necessary.

The exposure to chemicals is, apart from level of contamination and release, controlled by human behaviour, physiological characteristics, and environmental factors. Data on variability and uncertainty in the physiological characteristics and more general exposure factors were gathered from compilations in the literature. Complementary data on time use and the behaviour at the public bathing place were collected through observation studies and questionnaires to the public. The exposure was subsequently quantified to characterize the potential health risks involved, and to evaluate whether any additional risk management efforts were needed.

2. Materials and methods

The exposure assessment involved five distinct tasks: environmental sampling and chemical analysis, choice of exposure model, collection and evaluation of exposure factor data from the literature, observation study and questionnaires on recreational and bathing behaviour, and risk characterization.

2.1. Sampling and chemical analysis

Lake Trekanten is a small lake with a surface area of only 13.5 hectare (33.4 acre), an average depth of 4 m, and a maximum depth of 7 m. The lake is situated close to the main E4 highway in the southern part of Stockholm, the capital of Sweden (Fig. 1). There are four outlets into the lake for the discharge of stormwater runoff.

Water and sediment samples were collected on two occasions, in August 2006 and 2007, but the first set of samples were used only to screen for toxic elements (Sb, As, Ba, Be, Cd, Cr, Co, Cu, Pb, Mo, Ni, Se, Sn, V, Zn, and Hg) and organic contaminants (aliphatic hydrocarbons, pesticides, phenols, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, phtalates, other semi-volatile and volatile organic compounds, in total more than 200 compounds). Most contaminants were below the detection level in the first set and the samples from 2007 were therefore analyzed with more sensitive methods and selected as the quantitative basis for the exposure assessment.

Four sediment grab samples were collected at 1–2 m water depth in a 10 m radius around the bathing place, at the north-western corner of the lake. An additional sediment sample was collected closer to the highway. Six water samples were also collected manually and below surface from the same positions, with an additional sampling site at the eastern part of the lake. The samples prepared for elemental analysis were collected in acid-washed 0.5 L polypropylene vessels. The samples for organic trace analysis were collected in 3-L glass vessels that had been washed and dried, and subsequently heated to 500 °C for 5 h.

The sediment and water samples were analyzed for 56 elements by an accredited laboratory: Actlabs (Ancaster, ON, Canada). The sediment samples were sieved to <2 mm and prepared for elemental analysis by drying at 35 °C until constant weight (48–72 h), followed by strong leaching of 2 g dried sediment sample in 10 ml aqua regia at 110 °C for 1.5 h. The water samples were prepared by addition of concentrated HNO_3 to 2% (w/w) in the unfiltered sample. This releases elements bound to organic matter and exchangeable sites, but not from resistant minerals. The samples were subsequently analyzed by ICP-MS and ICP-AES.

The sediment and water samples were analyzed for polycyclic aromatic hydrocarbons (PAH) by another accredited laboratory:

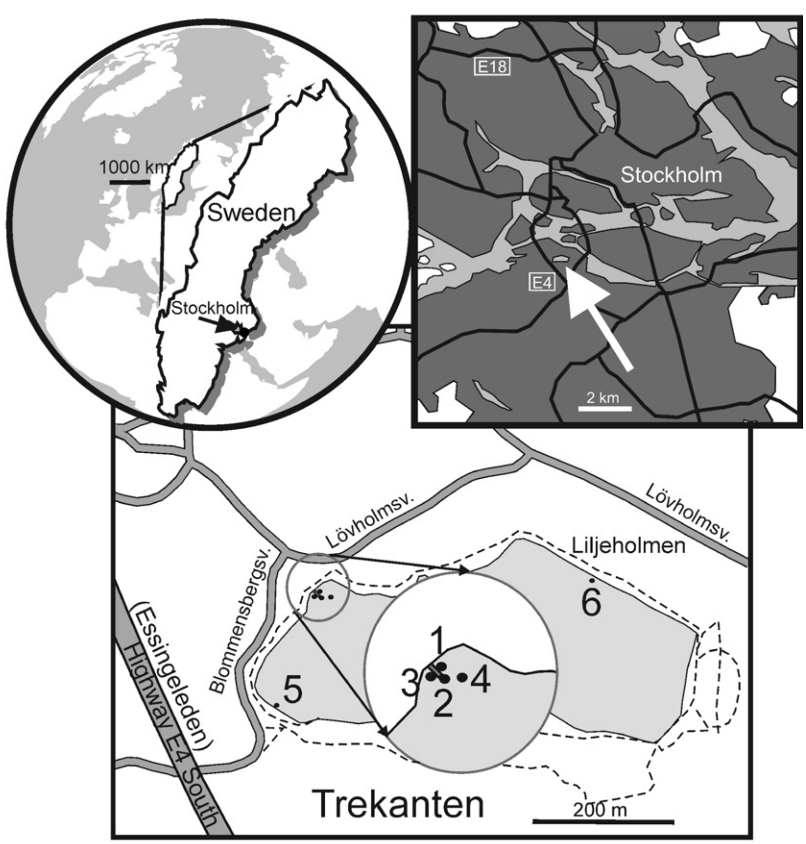

Fig. 1. Lake Trekanten, 4 km from the city centre of Stockholm and close to the main highway E4.

ALcontrol (Linköping, Sweden). Sediment and water samples were Soxhlet and liquid–liquid extracted, respectively, following US EPA methods 3540 and 3520 [6,7]. Polycyclic aromatic hydrocarbons (PAH) were separated and quantified by high-resolution gas chromatography and selected ion monitoring mass spectrometry (HRGC-MS/SIM). Deuterium-labelled standards were added to the samples prior to extraction and analytical results were reported for 16 PAH species.

For the purpose of this risk assessment the toxic equivalent concept of Nisbet and LaGoy was adopted, and the analytical results for PAH were recalculated to benzo[a]pyrene-equivalents [8]. Several other weighting scales have also been reported and a revised version has been announced by the US EPA, but is currently not yet available [9].

2.2. Exposure model

Three exposure pathways were considered in this assessment; oral intake of water, oral intake of sediment, and dermal uptake. The exposure equations conform to the recommended US EPA multiple pathway model for Superfund risk assessments [10]. These calculations, however, do not take bioavailability by oral intake into account. Such differences add to the overall uncertainty in intake estimates, but data were not available to characterize this additional uncertainty.

The oral intake of contaminants from lake water (I_w, mg/kg-day) is determined by: the chemical concentration in water (CW, mg/L), the amount of water swallowed while swimming (CR, L/h), exposure time (ET, h/day), exposure frequency (EF, days/year), exposure duration (ED, years), body weight (BW, kg), and the period over which exposure is averaged (AT, days), Eq. (1).

$$I_w = \frac{CW \times CR \times ET \times EF \times ED}{BW \times AT} \quad (1)$$

The oral intake from sediment (I_s, mg/kg-day) is determined by: the chemical concentration in sediment (CS, mg/kg), the intake of sediment from the contaminated source (IR, mg/day), exposure frequency (EF, days/year), exposure duration (ED, years), body weight (BW, kg), and the period over which exposure is averaged (AT, days), Eq. (2). The term sediment here encompasses all solid fractions irrespective of grain size. A conversion factor (CF, 10^{-6} kg/mg) is also included in the equation.

$$I_s = \frac{CS \times IR \times CF \times EF \times ED}{BW \times AT} \quad (2)$$

The dermal uptake (I_{du}, mg/kg-day) is determined by the chemical concentration in sediment (CS, mg/kg), skin surface area available for contact (SA, cm^2/day), sediment to skin adherence factor (AF, mg/cm^2), absorption factor (ABS), exposure frequency (EF, days/year), exposure duration (ED, years), body weight (BW, kg), and the period over which exposure is averaged (AT, days), Eq. (3). A conversion factor (CF, 10^{-6} kg/mg) is also included.

$$I_{du} \frac{CS \times CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (3)$$

The exposure pathways (I_w, I_s and I_{du}) were summarized. The equations were reorganized to the following equation, in order to avoid repetition of the exposure factors in the probabilistic calculation, Eq. (4).

$$I_{tot} = \frac{EF \times ED}{BW \times AT} \times ((CW \times CR \times ET) + (CS \times CF \times (IR + (SA \times AF \times ABS)))) \quad (4)$$

2.3. Exposure factor data

Data on body weights and skin surface areas were taken from a recent compilation of data on the Swedish population [11]. These data are given as multiple percentiles (1–99%) and other statistics, with uncertainty ranges (95% confidence intervals) bootstrapped. The skin surface areas were calculated from body weights and heights using the formula by Gehan and George [12].

Accidental intake of water by swimmers was investigated in a pilot study and a full field sampling study by the US EPA [13–15]. The data from the full field sampling study were used to estimate percentiles and statistics with bootstrapped uncertainty ranges.

Data on soil intake (assumed similar for sediment), sediment adherence and absorption factors were taken from the literature: the US EPA's Exposure Factors Handbook, Child-Specific Exposure Factors Handbook, Risk Assessment Guidance for the Superfund, and a Swedish draft guideline on contaminated soil [16–21]. However, the uncertainty is substantial and the reviewed literature seems to suggest that the absorption factors for PAH are overestimated [22–26].

2.4. Observation study and questionnaires

Reliable information on bathing behaviour was not found in the literature and a separate study of recreational behaviour at the bathing place was therefore conducted. In a pilot study, 38 participants visiting the lake were interviewed between May and June 2006. In a structured interview format questions were answered regarding recreational use of the lake and bathing place. Supplementary questions covered age, sex, children, residential address, and residence time in the vicinity. The pilot study was subsequently used for designing the questionnaire used for the main study.

The main study was completed during June–August 2006, with 67 interviews and 101 self-reported questionnaires (distributed at a nearby kiosk). Observation studies of bathing behaviour were conducted during seven days within the same time period. The observation study focused on assessing the number of visitors at the bathing place, the sex and age distribution, and bathing behaviour. People at the beach and in the water were counted every hour during daytime. Bathing behaviour among some of the children was observed for several hours. Observation protocols were supplemented with photographic documentation (Fig. 2).

Results from the observation study and questionnaires were subsequently used to estimate exposure time and frequency, and residence time in the area.

2.5. Data analysis and exposure calculations

Statistics for the concentration of contaminants and exposure factors were calculated with the software SPSS v15.0 (SPSS Inc., Chicago, IL, USA). The confidence intervals for these statistics and percentiles were estimated by re-sampling ("bootstrapping") with 10 000 iterations [27]. The bootstrapping calculations were made with the software Crystal Ball v7.2.2 (Decisioneering Inc., Denver, CO, USA).

Initially, mean and reasonable maximum exposures (RME) were calculated for the contaminants considered (selected elements and PAH). The RME were estimated either from the 95 or 5 percentiles (depending on factor) or maximum values (when percentiles could not be calculated). There is, however, a growing interest in applying probabilistic methods to handle variability and uncertainties [28]. Subsequently, a probabilistic calculation was performed for long time exposure to PAH. The probabilistic calculations were

Fig. 2. Photographic documentation of the bathing place.

performed with the software RAMAS Risk Calc v4.0 (Applied Biomathematics, Setauket, NY, USA) [29].

The uncertainty in assigning parametric distributions to observation data has been highlighted previously [11,28,30]. We have therefore chosen to use probability bounds analysis (PBA), to avoid unwarranted assumptions about distribution form and dependencies. PBA is founded on the use of probability boxes (*p*-boxes) rather than probability distributions to describe model inputs. In contrast to Monte Carlo simulations, PBA can also be performed without assuming independence between exposure factors [29,31].

A *p*-box is a class of distribution functions $F(x)$ bounded by two cumulative distribution functions $F_1(x)$ and $F_2(x)$ such that $F_1(x) \leq F(x) \leq F_2(x)$ for all x. When precise information about distribution form is unavailable, other available pieces of information (min, max, statistics, and percentiles) can be put together to construct these constraints for the class of possible distributions. For example, a *p*-box for the concentration of benzo[*a*]pyrene-equivalents in sediment was constructed from the minimum, mean, and standard deviation (Fig. 3).

3. Results and discussion

The chemical analyses showed very low concentrations of most pollutants in both sediment and lake water samples at the bathing place. Only chromium was found at levels above the European drinking water criteria (50 μg/L) and arsenic was the only element of concern in any of the intake estimates. Here we therefore limit our evaluation to these two elements and polycyclic aromatic hydrocarbons, reported as benzo[a]pyrene-equivalents (BaP-equivalents).

3.1. Chemical analysis

Table 1 summarizes the analytical results for arsenic, chromium, and PAH as BaP-equivalents. Non-detects were treated as one-half of the detection limit in calculating the statistics.

3.2. Exposure factors and observations

Observation data and questionnaire responses were available for exposure time (ET), exposure frequency (EF), and exposure duration (ED). The average exposure time estimates were in an interval 20–30 min for children 1–6 years and 30–40 min for children 7–14

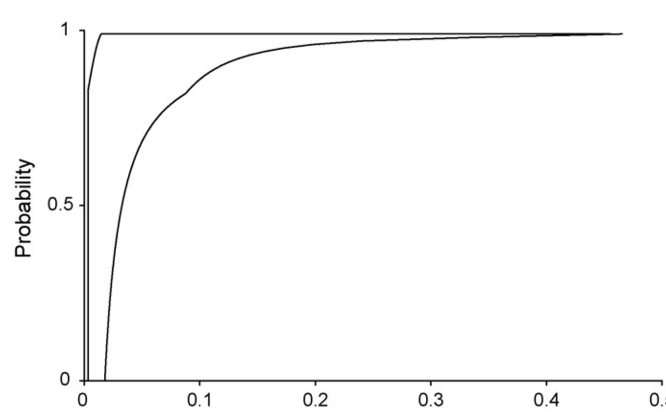

Fig. 3. A probability box (“*p*-box”) described by minimum, mean, and standard deviation of benzo[*a*]pyrene-equivalents in sediment (mg/kg).

Table 1
Concentrations of arsenic, chromium, and PAH as BaP-equivalents in sediments (mg/kg) at the bathing place and in lake water (mg/L).

	n	Mean	SD	Min	Median	Max
Sediments						
As	5	0.5	0.071	0.4	0.5	0.6
Cr	5	13	2.0	9.9	13	16
BaP	5	0.018	0.032	0.0033	0.0034	0.074
Water						
As	6	<0.01	0	<0.01	<0.01	<0.01
Cr	6	0.11	0.033	0.050	0.11	0.15
BaP	6	1.0E−5	3.5E−6	7.0E−6	9.2E−6	1.5E−5

n = number of samples and SD = standard deviation.

years. These intervals were adjusted upwards to 1 and 1.5 h/day for these two age groups, to account for repeated exposure the same day. The small children (2–3 years) went up and down from the water about 9–10 times while they were observed (3.5–4 h), sometimes playing with sand in the water and sometimes just sitting in the water playing. The somewhat older children, about 10–12 years, were diving, picking up sand from the bottom, playing and swimming. For adults, the questionnaire response 0.5 h/day was used unadjusted. Some children were observed to bath for several hours. These observations, and literature data [4,18,19], were used to support an estimated RME of 4 h/day for children and 2 h/day for adults.

The exposure frequencies were estimated from the questionnaires and an assumed bathing season of 8 weeks as best estimate and 10 weeks as RME. The frequencies were estimated to 32 and 70 days/year for children 1–6 years, 24 and 70 days/year for children 7–14 years, and 16 and 40 days/year for adults, respectively. The exposure duration was also estimated from the questionnaire data, where the average residence time in the vicinity was 13 years and the 95 percentile was 42 years. The exposure duration for the two age groups of children was estimated at 6 and 13 years of exposure, respectively, and for all groups the lifetime was assumed to be 80 years. The averaging time (AT) for lifetime exposure is thus $80 \times 365 = 29\,200$ days.

The water intake during swimming activities in a pool has been studied by the US EPA [13–15], and we have assumed that this exposure situation is similar to the one we have evaluated. The data on individual intake estimates from the full field sampling study (FFSS) were used as a basis for the intake calculations [13]. The estimated statistics and percentiles for children and women, with bootstrapped uncertainty intervals, are summarized in Table 2. The average was used as best estimate and the 95 percentile as RME.

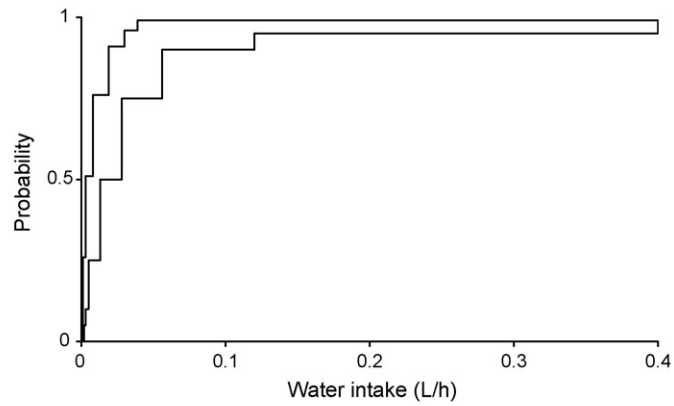

Fig. 4. *P*-box described by uncertainty ranges of the mean, the standard deviation, and the percentiles of water intake for women (L/h).

The exposure factors used in the deterministic estimate are summarized in Table 3. The majority of adults at the bathing place were women (60–70%). Exposure factors for adults are therefore given only for females.

The probabilistic exposure estimate was calculated by probability bounds analysis and *p*-boxes for the exposure factors are given in Table 4. We also had access to data to develop more accurate *p*-boxes for three exposure factors: body weight, skin surface area, and intake of water [11,13]. For example, the available information about the intake of water by female adults (CRadult, L/h) included the statistics reported in Table 5, with single values or interval estimates. The synthesis of this information is the *p*-box shown in Fig. 4, which is the envelope of all the cumulative probability distributions that are consistent with the given information. The other *p*-boxes for these three factors were constructed similarly.

3.3. Exposure assessment

The daily intakes of chromium for female adults and children have been estimated using the deterministic exposure model, the measured concentrations (Table 1), and the estimated exposure factors (Table 3). A best estimate (mean) and a reasonable maximum exposure estimate for oral intake and absorbed dose are given in Table 6. The oral intake then includes intakes of both lake water and sediments. The total intake is calculated as the sum of all the three exposure routes.

Arsenic and some polycyclic aromatic hydrocarbons are both carcinogenic and genotoxic, and hence the risk calculations are

Table 2
Water intake while swimming (mL/h), statistics and percentiles with uncertainty estimates.

	Statistics			Percentiles					
	n	Mean	SD	5	10	25	50	75	90	95
Children 6–12 years	90	54 (44–64)	48 (36–59)	4 (1–9)	8 (4–13)	20 (14–25)	44 (31–54)	75 (56–84)	128 (82–157)	159 (102–198)
Women 18– years	180	28 (16–45)	99 (23–164)	1 (1–2)	2 (1–3)	4 (3–5)	10 (8–13)	24 (19–28)	39 (30–56)	72 (39–120)

Table 3
Exposure factors used in the deterministic exposure calculations.

Exposure factor	1–6 years		7–14 years		Adult women	
	Best estimate	RME	Best estimate	RME	Best estimate	RME
Exposure time, h/day (ET)	1	4	1.5	4	0.5	2
Exposure frequency, days/year (EF)	32	70	24	70	16	40
Body weight, kg (BW)	18.2	14.6	38.6	27.9	67.7	52.0
Skin surface area, cm^2/day (SA)	7500	6500	12 500	10 300	17 700	15 300
Water intake while swimming, L/h (CR)	0.054	0.16	0.054	0.16	0.028	0.072
Sediment intake, mg/day (IR)	200	400	100	300	50	100
Sediment to skin adherence factor, mg/cm^2 (AF)	0.70	1.17	0.16	0.39	0.16	0.39

Table 4
P-boxes for concentrations and exposure factors. For each variable, the *p*-box defined by the specified parameters represents all probability distributions having those parameters. Intervals are given within square brackets.

Exposure factor	Probability box	
BaP-equivalent concentration in sediment, CS (mg/kg)	CSBaP: minimum = 0.0033, mean = 0.018, standard deviation = 0.032	
BaP-equivalent concentration in water, CW (mg/kg)	CWBaP: minimum = 7.0E−06, mean = 1.0E−05, standard deviation = 3.5E−06	
Absorption factor, ABS (-)	ABS = [0,0.13]	
Sediment to skin adherence factor, AF (mg/cm^2)	AFchild: minimum = 0.1, maximum = 2, mean = [0.36,1.17]	AFadult: minimum = 0.01, maximum = 1, mean = [0.07, 0.39]
Exposure frequency, EF (days/year)	EFchild: minimum = 3.5, maximum = 70, mean = 32	EFadult: minimum = 2, maximum = 70, mean = 16
Exposure time, i.e., time spent in water, ET (h/day)	ETchild: minimum = 0.1, maximum = 4, mean = 1	ETadult: minimum = 0.1, maximum = 2, mean = 0.5
Intake of sediment, children, IR (mg/day)	IRchild: minimum = 5, maximum = 400, mean = 200	IRadult: minimum = 1, maximum = 100, mean = 50

Table 5
Eleven statistics defining a *p*-box for the intake of water by female adults during swimming.

Statistic	Value or interval
Minimum	0.001
Maximum	0.4
Mean	[0.016, 0.045]
Standard deviation	[0.023, 0.164]
5th percentile	[0.001, 0.002]
10th percentile	[0.001, 0.003]
25th percentile	[0.003, 0.005]
50th percentile	[0.008, 0.013]
75th percentile	[0.019, 0.028]
90th percentile	[0.030, 0.056]
95th percentile	[0.039, 0.120]

evaluated on a lifetime basis. A best estimate (mean) and a reasonable maximum exposure estimate for oral intake and absorbed dose are given in Table 7.

The intakes of arsenic and BaP-equivalents are averaged over an estimated lifetime of 80 years. The calculations have been performed by summing the exposure during the periods evaluated. For example, the RME daily intake of BaP-equivalents for someone living in the vicinity between 1 and 42 years is:

$$\left(\frac{EF \times ED}{BW \times AT} \times ((CW \times CR \times ET) + (CS \times CF \times (IR + (SA \times AF \times ABS))))\right)_{1-6\ \text{years}} +$$
$$\left(\frac{EF \times ED}{BW \times AT} \times ((CW \times CR \times ET) + (CS \times CF \times (IR + (SA \times AF \times ABS))))\right)_{7-14\ \text{years}} +$$
$$\left(\frac{EF \times ED}{BW \times AT} \times ((CW \times CR \times ET) + (CS \times CF \times (IR + (SA \times AF \times ABS))))\right)_{15-42\ \text{years}} =$$
$$\left(\frac{70 \times 6}{14.6 \times 29200} \times ((0.000015 \times 0.16 \times 4) + (0.074 \times 0.000001 \times (400 + (6500 \times 1.17 \times 0.13))))\right) +$$
$$\left(\frac{70 \times 8}{27.9 \times 29200} \times ((0.000015 \times 0.16 \times 4) + (0.074 \times 0.000001 \times (300 + (10300 \times 0.39 \times 0.13))))\right) +$$
$$\left(\frac{40 \times 28}{52 \times 29200} \times ((0.000015 \times 0.072 \times 2) + (0.074 \times 0.000001 \times (100 + (15300 \times 0.39 \times 0.13))))\right) =$$
$$2.1E - 7\ \text{mg/kg-day}.$$

In addition, the variability and uncertainty in the intakes of BaP-equivalents were evaluated with probability bounds analysis (PBA), assuming independence and partial dependencies between exposure factors, respectively. Partial dependencies were assumed between the body weight and the various intake estimates, the sediment intake and the skin surface area, and the sediment to skin adherence factor and the absorption factor. The results from the PBA-computations are summarized in Table 8, as median and 95 percentiles with uncertainty intervals.

It should be noted that these intervals for the 95 percentiles also enclose the RME from the deterministic calculation.

3.4. Risk characterization

The exposure estimate for chromium was compared to a tolerable daily intake (TDI) of 1.5 mg/kg-day [21]. The lifetime exposure estimates for arsenic and BaP-equivalents were compared to risk-based daily intakes corresponding to a lifetime cancer risk of 1E−6. These risk-based values were 6.0E−6 mg/kg-day for arsenic and 8.3E−7 mg/kg-day for benzo[a]pyrene, respectively [21].

The RMEs for chromium are far below TDI and the concentrations in sediment are actually at the background level, and thus not cause for any further concern. Similarly, the RMEs for benzo[a]pyrene are below the risk-based daily intake. The probability bounds analysis, accounting for both variability and uncertainty in the exposure estimate, seems to confirm that the intake of BaP-equivalents is below the risk-based daily intake. It is only when partial dependencies are assumed between some exposure factors that the risk-based daily intake is slightly exceeded.

The RMEs for arsenic are twice the risk-based daily intake for those living in the vicinity as children. However, substantial uncertainty in the intake estimates is due to the non-detects in lake water. The RMEs were estimated assuming water concentrations at the detection limit, which is likely to exaggerate the maximum

Table 6
Daily intakes of chromium for female adults and children 1–6 years old (mg/kg-day).

	Oral intake		Absorbed dose		Total intake	
	Best estimate	RME	Best estimate	RME	Best estimate	RME
Children	4.0E−5	1.3E−3	3.3E−6	1.6E−5	4.3E−5	1.4E−3
Female adults	1.4E−6	4.9E−5	2.4E−7	2.0E−6	1.6E−6	5.1E−5

Table 7
Daily intakes of arsenic and BaP-equivalents for female adults and children 1–13 years old, on a lifetime basis (mg/kg-day).

	Oral intake		Absorbed dose		Total intake	
	Best estimate	RME	Best estimate	RME	Best estimate	RME
Arsenic						
Exposure in age 1–13 years	2.0E−7	1.0E−5	3.3E−8	1.8E−7	2.3E−7	1.1E−5
Exposure in age 1–42 years	2.3E−7	1.2E−5	4.3E−8	2.6E−7	2.8E−7	1.2E−5
Exposure during 13 years as adult	1.0E−8	5.1E−7	4.5E−9	3.7E−8	1.4E−8	5.5E−7
Exposure during 42 years as adult	3.2E−8	1.7E−6	1.4E−8	1.2E−7	4.7E−8	1.8E−6
Exposure during a whole lifetime (1–80 years)	2.6E−7	1.4E−5	5.6E−8	3.7E−7	3.2E−7	1.4E−5
BaP-equivalents						
Exposure in age 1–13 years	1.9E−9	5.8E−8	2.5E−9	9.6E−8	4.4E−9	1.5E−7
Exposure in age 1–42 years	2.1E−9	6.8E−8	3.3E−9	1.4E−7	5.4E−9	2.1E−7
Exposure during 13 years as adult	1.1E−10	3.3E−9	3.4E−10	2.0E−8	4.5E−10	2.3E−8
Exposure during 42 years as adult	3.5E−10	1.1E−8	1.1E−9	6.4E−8	1.5E−9	7.4E−8
Exposure during a whole lifetime (1–80 years)	2.5E−9	7.7E−8	4.3E−9	2.0E−7	6.8E−9	2.8E−7

Table 8
Uncertainty intervals in daily intakes of BaP-equivalents on a lifetime basis, evaluated with probability bounds analysis, assuming independence or partial dependencies between exposure factors (mg/kg-day).

Exposure duration	Independence		Partial dependencies	
	Median	95th percentile	Median	95th percentile
Age 1–6 years	[1.1E−11, 1.9E−8]	[1.5E−10, 1.8E−7]	[3.0E−12, 7.2E−8]	[2.1E−11, 5.6E−7]
Age 1–13 years	[1.7E−11, 3.7E−8]	[2.4E−10, 3.5E−7]	[4.3E−12, 1.4E−7]	[3.1E−11, 9.9E−7]
13 years as adult	[5.2E−13, 3.9E−9]	[6.2E−12, 4.8E−8]	[2.2E−13, 3.3E−8]	[1.6E−12, 2.7E−7]
42 years as adult	[1.7E−12, 1.3E−8]	[2.0E−11, 1.5E−7]	[7.2E−13, 1.1E−7]	[5.2E−12, 8.8E−7]

exposure. The concentrations of arsenic in the analyzed sediment samples are far below the current Swedish guideline of 10 mg/kg for contaminated soil [21].

The calculated exposures can also be compared to dietary intakes, a major exposure route for many environmental pollutants. The dietary intakes of chromium and arsenic among Swedish women have been estimated to 20 and 60 μg/day, respectively, or 3.3E−4 and 1.0E−3 mg/kg-day assuming a body weight of 60 kg [32]. The average intake of benzo[a]pyrene alone is 230 ng/day in Sweden, or 3.8E−6 mg/kg-day assuming a body weight of 60 kg [33]. The exposures estimated here were substantially below these figures and the relative importance would be even less if we also considered inhalation.

4. Conclusions

Recreational and outdoor activities are important to the well-being of people in urban areas. We expect that the general outline of this assessment and the exposure factors herein can be reused to assess similar exposure situations elsewhere. For example, this investigation indicates that beach visitors spend less time at the beach and in the water as compared to assumptions in the literature [1–2,5]. However, both climate and cultural differences motivate further studies to obtain realistic estimates.

The oral intake constitutes a major part of the exposure to metals, highlighting the importance of correctly estimating the intake of sediment and water. Recent investigations by the US EPA have added an important empirical database for these calculations [13–15]. In contrast, skin absorption is assumed to account for the major uptake of PAH. This assumption is, however, questionable and further experimental investigations are therefore needed.

The application of probability bounds analysis enabled a full description of uncertainty and variability in the exposure assessment from the data available. The 95 percentiles of the probability boxes covered the RME estimated with a deterministic approach, but it is interesting to note that the upper bounds of the uncertainty are at least twice the point estimates. Here, the probabilistic method is thus better suited to provide a conservative and protective exposure estimate.

This risk assessment focus on bathing activities, but many visitors came to Lake Trekanten also for other reasons than bathing; some were watching their children, reading a book, or just talking to each other. The interviews during the observation study showed that visitors enjoyed the beauty of the lake and appreciated having this oasis inside the city. The contamination and the present investigation have caused some concern, but almost everyone appreciated the effort and was interested in the water quality.

A general conclusion from this study is that currently the contamination in the deeper part of the lake should not cause concern for using the public bathing place. The exposure for polycyclic aromatic hydrocarbons and metals while bathing in Lake Trekanten, at the presently measured levels, cannot be anticipated to cause any significant adverse influence on public health.

However, changes in the deep sediments and a subsequent release of toxic materials could occur if the redox conditions are altered or if the sediments are physically disturbed. A long-term monitoring of the contamination in the lake therefore seems justified. Fishing is another important recreational activity that could contribute to the exposure, but this route is not covered here.

In this investigation, we specified the exposure pathways and the exposure model, gathered data on actual behaviour in the exposure situation (by an observation study), compiled supplementary data on general exposure factors, and characterized the uncertainty and the variability in these factors and the final exposure estimates. This assessment methodology can be adapted and used routinely in other situations with more heavily contaminated surface sediments and lake water. It is also possible, by iterative calculations, to estimate clean-up targets for contaminated shores and beaches [34]. Probability bounds analysis is a useful alternative to Monte Carlo-simulation when data are lacking to specify distributions. The additional behavioural data given here are probably applicable in many urban areas.

Acknowledgements

This study was funded by the City of Stockholm and the District Administration of Liljeholmen. We gratefully acknowledge the use of data from studies on water ingestion among swimmers conducted by the US EPA, and the advice provided by Drs. Otis Evans and Alfred P. Dufour. We also like to thank two anonymous reviewers for helpful suggestions to improve the manuscript.

References

[1] B.-J. Baars, The wreckage of the oil tanker 'Erika'—human health risk assessment of beach cleaning, sunbathing and swimming, Toxicol. Lett. 128 (2002) 55–68.
[2] F. Dor, R. Bonnard, C. Gourier-Fréry, A. Cicolella, R. Dujardin, D. Zmirou, Health risk assessment after decontamination of the beaches polluted by the wrecked ERIKA tanker, Risk Anal. 23 (2003) 1199–1208.
[3] M.R. Adams, C.A. Hanna, J.A. Mayernik, W.M. Mendez Jr., Probabilistic health risk assessment for exposures to estuary sediments and biota contaminated with polychlorinated biphenyls, polychlorinated terphenyls and other toxic substances, Risk Anal. 14 (1994) 577–594.
[4] D.K. Goldblum, A. Rak, M.D. Ponnapalli, C.J. Clayton, The Fort Totten mercury pollution risk assessment: a case history, J. Hazard. Mater. A136 (2006) 406–417.
[5] H.J. Albering, J.-P. Rila, E.J.C. Moonen, J.A. Hoogewerff, J.C.S. Kleinjans, Human health risk assessment in relation to environmental pollution of two artificial freshwater lakes in the Netherlands, Environ. Health Perspect. 107 (1999) 27–35.
[6] Test Methods for Evaluating Solid Waste, Physical/Chemical Methods. ASW-846. Method 3540C: Soxhlet Extraction. U.S. Environmental Protection Agency, Washington, DC, 1996.
[7] Test Methods for Evaluating Solid Waste, Physical/Chemical Methods. ASW-846. Method 3520C: Continuous Liquid–Liquid Extraction. U.S. Environmental Protection Agency, Washington, DC, 1996.
[8] I.C.T. Nisbet, P.K. LaGoy, Toxic equivalency factors (TEFs) for polycyclic aromatic hydrocarbons (PAHs), Regul. Toxicol. Pharm. 16 (1992) 290–300.
[9] L. Flowers, H. Carlson-Lynch, M. Gehlhaus, C. Keshava, P. McClure, G. Rice, J. Stickney, J. Strong, L. Teuschler, The relative potency factor (RPF) approach and polycyclic aromatic hydrocarbon (PAH) mixtures. Poster presentation, Society for Risk Analysis Annual Meeting, San Antonio, TX, 2007.
[10] Risk Assessment Guidance for Superfund, Volume 1, Human Health Evaluation Manual (Part A). Report EPA/540/1-89/002. U.S. Environmental Protection Agency, Washington, DC, 1989.
[11] M. Filipsson, B. Bergbäck, T. Öberg, Exposure Factors in Risk Assessment—An Inventory of Data (In Swedish: Exponeringsfaktorer vid riskbedömning—Inventering av dataunderlag). Report 5802. Swedish Environmental Protection Agency, Stockholm, Sweden, 2008.
[12] E.A. Gehan, S.L. George, Estimation of human body surface area from height and weight, Canc. Chemother. Rep. 54 (1970) 225–235.
[13] O. Evans, L.J. Wymer, T.D. Behymer, A.P. Dufour, An Observational Study: Determination of the Volume of Water Ingested During Recreational Swimming Activities. Poster presentation, National Beaches Conference, Niagara Falls, NY, 2006.
[14] A.P. Dufour, O. Evans, T.D. Behymer, R. Cantú, Water ingestion during swimming activities in a pool: a pilot study, J. Water Health 4 (2006) 425–430.
[15] O. Evans, R. Cantú, T.D. Behymer, D.D. Kryak, A.P. Dufour, A Pilot Study to Determine the Water Volume Ingested by Recreational Swimmers. Presentation, Society for Risk Analysis Annual Meeting, Seattle, WA, 2001.
[16] M.B. Shoaf, J.H. Shirai, G. Kedan, J. Schaum, J.C. Kissel, Adult dermal sediment loads following clam digging in tide flats, Soil Sediment. Contam. 14 (2005) 463–470.
[17] M.B. Shoaf, J.H. Shirai, G. Kedan, J. Schaum, J.C. Kissel, Child dermal sediment loads following play in a tide flat, J. Expos. Anal. Environ. Epidemiol. 15 (2005) 407–412.
[18] Exposure Factors Handbook. Report EPA/600/P-95/002F. U.S. Environmental Protection Agency, Washington, DC, 1997.
[19] Child-Specific Exposure Factors Handbook. Report EPA-600-P-00-002B. U.S. Environmental Protection Agency, Washington, DC, 2002.
[20] Risk Assessment Guidance for Superfund, Volume 1, Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment). Report EPA/540/R/99/005. U.S. Environmental Protection Agency, Washington, DC, 2004.
[21] Guideline Values for Contaminated Soil—Model Description and Guidance. Draft for Comment 2007-10-19 (In Swedish: Riktvärden för förorenad mark—Modellbeskrivning och vägledning, Remissversion). Swedish Environmental Protection Agency, Stockholm, Sweden, 2007.
[22] J.J. Yang, T.A. Roy, A.J. Krueger, W. Neil, C.R. Mackerer, In vitro and in vivo percutaneous absorption of benzo(a)pyrene from petroleum crude-fortified soil in the rat, Bull. Environ. Contam. Toxicol. 43 (1989) 207–214.
[23] T.A. Roy, R. Singh, Effect of soil loading and soil sequestration on dermal bioavailability of polynuclear aromatic hydrocarbons, Bull. Environ. Contam. Toxicol. 67 (2001) (2001) 324–331.
[24] J. Kao, The influence of metabolism on percutaneous absorption, in: R.I. Bronaugh, H.I. Maibach (Eds.), Percutaneous Absorption, Marcel Dekker, New York, NY, 1989, pp. 259–282.
[25] P. Sartorelli, L. Montomoli, A.G. Sisinni, R. Bussani, D. Cavallo, V. Foà, Dermal exposure assessment of polycyclic aromatic hydrocarbons: in vitro percutaneous penetration from coal dust, Toxicol. Ind. Health 17 (2001) 17–21.
[26] M.S. Abdel-Rahman, G.A. Skowronski, R.M. Turkall, Assessment of the dermal bioavailability of soil-aged benzo(a)pyrene, Hum. Ecol. Risk Assess. 8 (2002) 429–441.
[27] B. Efron, R. Tibshirani, An Introduction to the Bootstrap, Chapman and Hall, New York, NY, 1993.
[28] R.R. Lester, L.C. Green, I. Linkov, Site-specific applications of probabilistic health risk assessment: review of the literature since 2000, Risk Anal. 27 (2007) 635–658.
[29] S. Ferson, RAMAS Risk Calc 4. 0 Software: Risk Assessment with Uncertain Numbers, Lewis Publishers, Boca Raton, FL, 2002.
[30] P. Sander, B. Bergbäck, T. Öberg, Uncertain numbers and uncertainty in the selection of input distributions—consequences for a probabilistic risk assessment of contaminated land, Risk Anal. 26 (2006) 1363–1375.
[31] S. Ferson, L.R. Ginzburg, Different methods are needed to propagate ignorance and variability, Reliab. Eng. Syst. Safe. 54 (1996) 133–144.
[32] L. Jorhem, W. Becker, S. Slorach, Intake of 17 elements by Swedish women, determined by a 24-h duplicate portion study, J. Food Comp. Anal. 11 (1998) 32–46.
[33] Scientific opinion of the panel of contaminants in the food chain on request from the European Commission on polycyclic aromatic hydrocarbons in food, EFSA J. 724 (2008) 1–114.
[34] Risk Assessment Guidance for Superfund: Volume III—Part A, Process for Conducting Probabilistic Risk Assessment. Report EPA/540/R/02/002. U.S. Environmental Protection Agency, Washington, DC, 2001.