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Assessment of the Chemical Quality of Recreational Bathing Water in Argentina by Health Risk Analysis

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ABSTRACT

Del Azul creek (Argentina) is a natural water body used for recreational bathing in which heavy metals and pesticides have been detected. The aim of the study is to estimate the probabilistic non-cancer and cancer risks by recreational bathing, applying U.S. Environmental Protection Agency models for aggregated (exposure through accidental oral intake of water and dermal contact) and cumulative risks (combined exposure to groups of substances) for bathers of 5, 10, 15, and 20 years old. Bathing in Del Azul creek does not generate risks at the concentrations and the exposure scenarios considered. Cypermethrin and arsenic and heptachlor were the riskiest non-cancer and cancer substances, respectively. Our study highlights the importance of considering both routes of exposure because of the great significance of the dermal route and because of the variability of population characteristics, as it has been stated in other studies. These considerations are highly significant for Argentina, where the quality of recreational water for other than microbiological causes is frequently evaluated based only on the harmful oral intake and applied to a hypothetical individual representative of the population.

Key Words: health risk analysis, heavy metals, pesticides, recreational waters.

INTRODUCTION

Bad quality recreational water is associated with pulmonary, sensory organs (eyes, ears), skin, and, particularly, gastrointestinal diseases. Vomiting, diarrhea, and nausea are the most studied ones, associating water quality and the presence of bacteria indicators with such diseases. The impact of water quality on recreational use is mainly described in terms of fecal contamination caused by pathogenic microorganisms (WHO 1998), establishing the general evaluation and monitoring guidelines

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through the *Annapolis Protocol* (WHO 1999). In *Monitoring Bathing Waters—A Practical Guide to the Design and Implementation of Assessments and Monitoring Programmes* (WHO 2000) assessment of the microbiological quality plays a key role when analyzing bathing waters. Regarding chemical contamination of recreational waters, WHO (1998) stated that bathers' health must be ensured, though how to do so is not mentioned.

In Argentina, water quality for recreational use can be evaluated based on the *National Guide Levels for Water Quality for Human Recreation* (NUHR 2007a), although only microbiological parameters are established. The lack of a national regulatory framework with guideline levels for substances potentially toxic in recreational waters makes it difficult to evaluate their bathing aptitude. Thus, when hazardous chemicals are detected in water bodies, the most frequent management procedure to assess their bathing aptitude is to compare them with their safe levels for water for human consumption established in the *National Guidance Levels for Environmental Water Quality for Sources of Water Provision for Human Consumption* (NUHR 2007b), or those of the *Argentinean Food Code* (NCAR 1969, 1971). However, these water management procedures present some drawbacks (Peluso *et al*. 2011). Given the fact that toxic substances have been detected in water bodies that could potentially be or are currently used for bathing or swimming, it is necessary to evaluate the health risk for the people exposed to them without adequate management tools legally recognized.

The health risk based on the U.S. Environmental Protection Agency (USEPA) model is an estimate of the likelihood that a chemical agent of concern will generate cancer or non-cancer effects in exposed people (USEPA 1989). Although many papers can be found in which health risk assessment based on these models were used, the application of them to recreational activities (bathing, swimming) has seldom been reported in the literature (Albering *et al*. 1999; Filipsson *et al*. 2009) for other than microbiological causes. Among the papers in which such models are applied, many have been focused on swimming pools, mainly centered on trihalometanes as hazardous substances (Blando and Cohn 2004; Panyakapo *et al*. 2008; Lee *et al*. 2009; Chen *et al*. 2011) and chlorine by-products generated after water disinfection (Panyakapo *et al*. 2008).

In the absence of specific tools for assessing water quality for recreational bathing, the aim of our study is to evaluate the potential health risk due to the hazardous substances (heavy metals, chlorinated pesticides) founded in the Del Azul creek (Buenos Aires province, Argentina) using USEPA models. The risks were calculated using individual, aggregated (accidental intake and dermal contact) and cumulative probabilistic models, for bathers of 5, 10, 15, and 20 years old.

METHODOLOGY

Study Region

The Buenos Aires province, located in central-eastern Argentina, represents more than 10% of the total surface of the country and it is the province with the largest population (14 million inhabitants, according to the Directorate of Provincial Statistics of Buenos Aires Province 2010). The province is one of Argentina's main

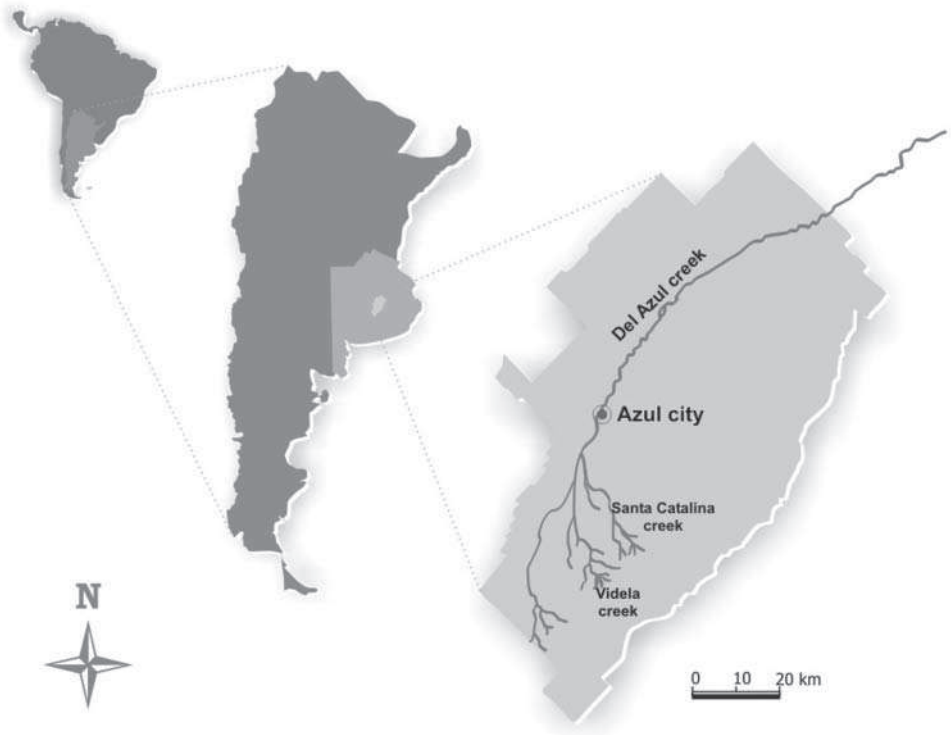

Figure 1. Location of Del Azul creek and tributaries in Azul county, in the center of the Buenos Aires province, Argentina.

agricultural producers, representing 40% of the national production (MLA 2004). The most prominent crops are: soybean (3.7 million Ha sowed); wheat (2.9 million Ha), sunflower (1.1 million Ha), and maize (0.8 million Ha), according to the year 2005/2006 data (Directorate of Provincial Statistics of Buenos Aires Province 2010).

The increase in the use of pesticides (from 73 to 236 million kilograms between 1995 and 2005, according to the Chamber of Agricultural Health and Fertilizer of Argentina 2007) generated environmental impacts like water, soil, and biota pollution. And several studies have revealed the presence of biocides in water compartments of Argentina (Zubillaga *et al*. 1987; Janiot *et al*. 1994; Loewy *et al*. 1999; Menone *et al*. 2000; Rovedatti *et al*. 2001; Miglioranza *et al*. 2004; Jergentz *et al*. 2005; Silva *et al*. 2005; Marino and Ronco 2005; Peruzzo *et al*. 2008).

The Del Azul creek basin (6000 km^2) is located in the centre of the Buenos Aires province (36°00′/37°20′S - 60°12′/58°52′W) (Figures 1 and 2). This creek is a 160 km long natural water body with tributaries (Videla and Santa Catalina creeks). In the middle is located Azul city (60,000 inhabitants), about 300 km distant from the capital city of Argentina. This water body has an enlargement in the urban fringe zone used as a bathing beach during the summer season. However, there are other bathing beaches although they are less frequented. The number of people visiting the Del Azul creek bathing resort during a weekend in summer is about 10,000, many of whom are bathers.

Figure 2. Location and names of sampling stations at Del Azul creek.

The agricultural sector of the basin is located in the topographically upper zone (250 m above sea level), to the south of the city (135 m above sea level), and upstream. Corn, wheat, soy, and sunflower are grown in this area. Due to the potential runoff of pollutants from farm soils adjacent to the stream, the water quality of Del Azul creek is monitored periodically by sampling of pesticides and heavy metals (Peluso *et al*. 2011).

Health Risk Analysis Model

The exposure to the hazardous substances was analyzed in the light of two alternatives of body contact during recreational bathing: accidental water ingestion and skin contact. In both cases, USEPA models were used, applying Eqs. (1) and (2),

respectively (USEPA 1992a, 2004).

$$\text{ADDI} = [\text{Conc} * \text{Ir} * \text{EF} * \text{ED}]/[\text{BW} * \text{AT}] \quad (1)$$

$$\text{ADDS} = [\text{DA}_{\text{event}} * \text{ESA} * \text{EF} * \text{ED} * \text{FC}]/[\text{BW} * \text{AT}] \quad (2)$$

where ADDI = average daily dose by accidental intake (mg kg^{-1} day^{-1}), ADDS = average daily dose by skin contact (mg kg^{-1} day^{-1}), DA_{event} = absorbed dose per event (mg cm^{-2} $event^{-1}$), Conc = concentration of the hazardous substance in water (mg L^{-1}), Ir = daily water intake rate (L day^{-1}), EF = exposure frequency (day $year^{-1}$), ED = exposure duration (year), BW = weight of the exposed individual (kg), AT = correction factors of average time for chronic exposure (ED × 365 days for non-cancer risk estimation, 70 years × 365 days for cancer risk estimation), ESA = exposed skin area (cm^2), and FC = correction factor of surface and volume units (10,000 cm^2 m^{-2} × 0.001 L cm^{-3}). These parameters are further explained in the text.

Non-cancer risk calculation (NCR) was conducted using the risk quotient (R). R is the ratio between the average daily dose (ADD) and the toxicological reference dose (RfD) for the particular chemical involved, specific for the route of exposure (USEPA 1989). This quotient reveals the extent of the potential harm in terms of the proportion that the daily dose of a hazardous substance exceeds its toxicological safety threshold. If the risk scores (R) are less than 1.0, the non-cancer health risk is assumed to be negligible (USEPA 1989).

Cancer risk calculation (CR) was also estimated based on the ADD of each route of exposure. Cancer risk (incremental lifetime cancer risk) was calculated by multiplying ADD by a toxicological reference value, the Slope Factor (SF), which is also specific for each exposure pathway (USEPA 1989). In the ADD for the cancer risk estimation, AT adopts 70 years as lifetime duration. It was assumed as safe criteria 1.0 and $1.0E^{-05}$ for NCR and CR, respectively.

The aggregated and cumulative health risks (risk caused by the simultaneous exposure through different routes of contact to the same hazardous substance, and by the simultaneous exposure when more than one hazardous substance is present, respectively) were calculated using an additive model forming a risk index, which was used by the USEPA for health risk assessments of initial prospecting, or screening model (USEPA 1989, 2007). This methodology is justified in the case when data on toxicology of the mixture, or similar, are unavailable (Mumtaz *et al.* 2007). It was assumed the same safe criteria for aggregated and cumulative risks as those for individual substance by individual route of exposure.

Parameters of the Model

Concentration of the hazardous substances in water (Conc)

The study was based on 12 water samples from eight sites at Del Azul creek between December of 2005 and December of 2007 (bimonthly sampling). The sampling was done in dry periods (without rain in the last 5 days). The location and name of the sampling stations are shown in Figure 2. Water samples for metals and pesticides determination were collected in high-density polyethylene and amber glass bottles with internal teflon tops, respectively. Samples were taken at a subsurface level (30 cm below surface), in the middle of the watercourse. The samples contained

some sediments resuspended by the sample staff's footsteps because, to take such simples, it is necessary to walk on the creek bottom to the sample point. Therefore, the water sample is highly representative of the water that may be ingested during bathing. The samples were refrigerated (4°C) until analysis and the analyses were made on the samples, which were previously shaken.

The substances are summarized in Table 1. To measure the concentrations of heavy metals (As, Cu, Cr, Hg, and Zn) we used a Varian Spectraa 55 Absorption Atomic Spectrophotometer, and VGA-77 Cold Vapor Generation, according to USEPA methods SW 846 M 7061A, SW 846 M 7190 EAA, SW 846 M 7210, SW 846 M 7470A, and SW 846 M 7950, respectively.

For isolation and concentration of organic compounds in aqueous samples, we used the procedures indicated in USEPA's method SW 846 M 3510C Separatory Funnel Liquid-Liquid Extraction. The organochlorine pesticides (α-HCH, β-HCH, γ-HCH, δ-HCH, aldrin, γ-chlordane, endosulfan, endosulfan sulphate, heptachlor), cypermethrin, and chlorpyrifos concentrations were measured with a Hewlett-Packard 5890II gas chromatograph with ECD and NPD detector, according to USEPA methods SW 846 M 8081 CG ECD, M 508-CG ECD, and SW 846 M 8141 CG NPD, respectively. The concentrations of 2,4-D and acetochlor were analyzed with a Waters Wat081110 HPLC-UV detector (USEPA method SW 846 M 8321A HPLC-UV). The glyphosate concentration was determined with a Waters Wat 081110 HPLC and derivatization post-column and fluorescence detector, based on the USEPA M 547 method. The xylenes concentrations were measured with a Hewlett-Packard-Agilent 6890A Plus+ gas chromatograph - flame ionization detector, according to USEPA's SW 846 M 8015 B/C/D CG-FID method.

Probability distributions were fitted to the concentrations of each substance with Crystal Ball 7.1 (Decisioneering 2007a), and the best-fitted probability distribution model and descriptive parameters are presented in Table 1. The software matches the concentration data against each continuous probability distribution (beta, exponential, gamma, normal, lognormal, logistic, Pareto, Weibull, and others), selecting the best fit by using Anderson Darling goodness-of-fit tests, determining the set of parameters for the distribution that best describes the characteristics of the data. These probability distributions were used as the input distributions for the concentration term of the exposure model calculation.

In all the water samples, there were non-detect concentrations. These were replaced by the 95% upper confidence limit (95% UCL) of the arithmetic mean of the group of the detected concentrations of the substance. The 95 percent UCL of a mean is a value that, when calculated repeatedly for randomly drawn subsets of site data equals or exceeds the true mean 95% of the time (USEPA 1992b). The UCL estimation was performed using ProUCL software v.4.1 (USEPA 2010a), which carries out a number of UCL parametric and distribution-free non-parametric methods, and recommends the one most appropriate to use based on the distribution of the data.

Intake rate (Ir), event duration (t_{event}), frequency (EF), and duration (ED) of recreational exposure

The exposed individuals were grouped into age categories of 5, 10, 15, and 20 years old. The assumed intake rate was 0.05 L per hour of the bath event duration

Table 1. Probability distribution model parameters and descriptive statistics of concentrations in Del Azul creek water samples (in mg L^{-1}).

Substance	Fitted Distribution	Min.[a]	Max.[b]	Me.[c]	SD[d]	Med.[e]	95P[f]	Others	
								Mid.[g]	Sc.[h]
2,4 D (2,4 Dichloro phenoxy acetic acid)	T-student	2.35 E^{-03}	2.98 E^{-01}	7.93 E^{-02}	2.91 E^{-02}	7.77 E^{-02}	1.27 E^{-01}	7.70 E^{-02}	2.32 E^{-02}
α–HCH (Hexachloro cyclohexane)	Beta	8.13 E^{-07}	6.60 E^{-05}	3.54 E^{-05}	2.01 E^{-05}	3.63 E^{-05}	6.41 E^{-05}	α 0.71	β 0.52
								Like.[j]	Sc.
β–HCH	Min. Ext.[i]	1.77 E^{-06}	5.00 E^{-06}	2.80 E^{-06}	6.51 E^{-07}	2.72 E^{-06}	3.99 E^{-06}	2.00 E^{-06}	1.00 E^{-06}
								Mid.	Sc.
σ–HCH	T-student	6.64 E^{-08}	1.90 E^{-05}	5.04 E^{-06}	2.44 E^{-06}	4.84 E^{-06}	9.33 E^{-06}	4.74 E^{-06}	2.21 E^{-06}
								Mean	Sc.
γ–HCH	Logistic	7.47 E^{-07}	9.50 E^{-06}	3.96 E^{-06}	8.53 E^{-07}	3.95 E^{-06}	5.36 E^{-06}	3.95 E^{-08}	S4.71 E^{-07}
								Like.	Sc.
Acetochlor	Min. Ext.	5.82 E^{-05}	7.10 E^{-02}	1.94 E^{-02}	1.24 E^{-02}	1.81 E^{-02}	4.18 E^{-02}	1.05 E^{-02}	2.42 E^{-02}
								Mid.	Sc.
Aldrin	T-student	4.04 E^{-07}	1.80 E^{-05}	6.04 E^{-06}	2.15 E^{-06}	5.95 E^{-06}	9.64 E^{-06}	6.00 E^{-06}	2.00 E^{-06}
Arsenic	Beta	1.00 E^{-02}	6.00 E^{-02}	2.84 E^{-02}	1.23 E^{-02}	2.65 E^{-02}	5.17 E^{-02}	α 1.29	β 3.07
								Loc.[k]	
Cypermethrin	Lognormal	4.00 E^{-04}	1.94 E^{+00}	4.74 E^{-01}	2.57 E^{+02}	4.39 E^{-03}	2.98 E^{-01}	3.71 E^{-01}	
								Loc.	Sc.
Chlorpyrifos	Gamma	2.00 E^{-04}	3.10 E^{-02}	4.53 E^{-03}	2.95 E^{-03}	3.93 E^{-03}	1.20 E^{-02}	−3.13 E^{-04}	1.83 E^{-03}
								Loc.	Sc.
Copper	Gamma	4.71 E^{-03}	1.60 E^{-02}	5.78 E^{-03}	2.01 E^{-03}	4.84 E^{-03}	1.04 E^{-02}	4.71 E^{-03}	8.94 E^{-03}
								Like.	Sc.
Chromium	Min. Ext.	1.64 E^{-03}	1.10 E^{-02}	3.97 E^{-03}	1.55 E^{-03}	3.75 E^{-03}	6.81 E^{-03}	2.86 E^{-03}	3.03 E^{-03}
								Like.	Sc.
Endosulfan	Min. Ext.	1.00 E^{-06}	7.50 E^{-06}	2.65 E^{-06}	1.07 E^{-06}	2.51 E^{-06}	4.58 E^{-06}	1.85 E^{-06}	2.11 E^{-06}
								Mid.	Sc.
Endosulfan sulphate	T-student	9.16 E^{-06}	3.70 E^{-05}	2.17 E^{-05}	4.01 E^{-06}	2.17 E^{-05}	2.83 E^{-05}	2.17 E^{-05}	3.54 E^{-06}
									Sc..
γ–Chlordane	Logistic	1.53 E^{-06}	8.90 E^{-06}	2.05 E^{-06}	4.29 E^{-07}	1.95 E^{-06}	2.89 E^{-06}		4.89 E^{-07}
									Sc.
Glyphosate	Logistic	5.01 E^{-03}	1.11 E^{+00}	1.59 E^{-01}	1.22 E^{-01}	1.34 E^{-01}	4.03 E^{-01}		1.00 E^{-01}
									Sc.
Heptachlor	Logistic	1.55 E^{-05}	6.30 E^{-05}	2.04 E^{-05}	4.02 E^{-06}	1.95 E^{-05}	2.84 E^{-05}		3.39 E^{-06}
Mercury	Beta	6.39 E^{-04}	1.60 E^{-03}	1.10 E^{-03}	3.64 E^{-04}	1.08 E^{-03}	1.60 E^{-03}	α 0.30	β 0.30
								Like.	Sc.
Xylenes	Min. Ext.	1.56 E^{-02}	3.40 E^{-01}	1.33 E^{-01}	7.30 E^{-02}	1.25 E^{-01}	2.64 E^{-01}	1.02 E^{-01}	1.28 E^{-01}
								Like.	Sc.
Zinc	Max. Ext.[l]	8.00 E^{-03}	4.80 E^{-02}	1.60 E^{-02}	5.90 E^{-03}	1.48 E^{-02}	2.70 E^{-02}	1.24 E^{-02}	4.92 E^{-03}

[a]Minimum value of concentration of the fitted distribution; [b]maximum value; [c]arithmetic mean; [d]standard deviation; [e]median; [f]95th percentile; [g]midpoint; [h]scale; [i]minimum extreme; [j]likeliest; [k]location; [l]maximum extreme.

for 5, 10, and 15 years old and 0.025 for 20 years old according to the USEPA (1989) and Filipsson *et al*. (2009), and they were the only deterministic input parameters in the exposure model. The remaining parameters were treated probabilistically using the probability distributions as the inputs of the respective parameter in the exposure model's calculation.

The duration of the bath event (relevant for the absorbed dose per event calculation ($-DA_{event}$ in Eq. (2)) and its frequency during the year for each age group was estimated based on a questionnaire administered by the authors of this article conducted at the Del Azul bath resort during the summer of 2010–2011. This unpublished survey asked 1000 randomly selected visitors about personal information (gender, age, family composition, bodyweight) as well as information about their bath patterns as well as those of their families. Some of the questions were about the following: the number of visits to the resort during the 2010–2011 summer; whether the participants are bathers or not; if bathers, how long the bath event is, and so on. The number of surveys was 250 for each age group, on randomly selected visitors.

The duration of the bath event and the annual frequency of bath days by age were probabilistically established based on the survey responses. The best fitted probability distribution model of these parameters by age was conducted with Crystal Ball 7.1 (Decisioneering 2007b) and results are presented in Table 2.

Although there were responses in the survey that explained that people bathed "every day of the summer" (meaning 90 days), the maximum number of bath days was estimated by computing the number of days with temperatures more than 27°C and without rains in the whole day, according to BDH (2011). The maximum number of bath days was established to be 54. The duration of exposure was probabilistically treated, assuming a triangular probability distribution with the lower and upper limits of 1 and 30 years, respectively, and a mode value of 15 years, which was common to the four age groups.

Bodyweight (BW), body skin area (BSA), and exposed skin area (ESA)

The probability distribution model and its descriptive parameters for the BW for the four age groups are shown in Table 2, and it is based on Lejarraga and Orfila's (1987) anthropometric data for Argentina's population. The body skin area was estimated by applying the DuBois and DuBois (1916) model, which is given in Eq. (3).

$$BSA = H^{0.725} \times BW^{0.425} \times 0.007184 \tag{3}$$

where BSA = body skin area, H = body height (cm), based on Lejarraga and Orfila (1987); BW = bodyweight (kg).

If it is assumed that when a bather is in full contact with water, BSA should be equal to ESA in Eq. (2). However, it is not necessarily accurate in reality to assume that the entire bath event duration involves a full-body exposure (*i.e.*, the body totally submerged). To calculate the body surface area exposed for use in the dermal dose estimation, a correction factor called Bath Pattern was applied. This was an attempt to estimate what percentage of the skin surface of a bather's body was in contact with the water during the bath event. That means that the skin area in contact with

Table 2. Probability distribution model and descriptive statistics of the parameters of the dose of exposure by age group: duration of the bath event, frequency of bathing days, bodyweight, height, body surface, bath pattern and exposed skin area.

Age Group	Param.	Fitted Distributions	Min.	Max.	Me.	SD	Med.	95P	Others	
										Sc.
5	t_{event} [a]	Logistic	0.00	30.00 E^{+01}	13.42 E^{+01}	74.10	12.88 E^{+01}	26.24 E^{+01}		61.95
									Loc.	Sc.
	EF [b]	Gamma	0.00	54.00	15.58	14.48	10.81	45.75	−0.11	43.22
	BW [c]	Normal	13.50	25.50	19.50	2.50	19.50	19.54		
	BSA [d]	Normal	76.19 E^{+02}	80.70 E^{+02}	76.19 E^{+02}	16.84 E^{+01}	76.16 E^{+02}	78.93 E^{+02}		
									Mo [g]	
	BP [e]	Triangular	0.07	1.00	0.49	0.19	0.47	0.83	0.41	
									α	β
	ESA [f]	Beta	6.27 E^{+02}	76.86 E^{+02}	37.17 E^{+02}	14.37 E^{+02}	36.38 E^{+02}	63.92 E^{+02}	2.27	3.49
									Like	Sc.
10	t_{event}	Min.Ext.	0.00	30.00 E^{+01}	18.81 E^{+01}	25.38	19.54 E^{+01}	27.77 E^{+01}	21.82 E^{+01}	62.24
									Loc.	Sc.
	EF	Gamma	0.00	54.00	19.91	14.34	15.75	47.59	−0.11	34.58
	BW	Normal	23.50	44.50	33.56	5.00	33.56	41.72		
	BSA	Normal	91.79 E^{+02}	13.34 E^{+03}	11.26 E^{+03}	72.59 E^{+01}	11.26 E^{+03}	12.40 E^{+03}		
									Mo	
	BP	Triangular	0.07	1.00	0.49	0.19	0.47	0.83	0.40	
									α	β
	ESA	Beta	83.84 E^{+01}	12.01 E^{+03}	55.26 E^{+02}	22.17 E^{+02}	53.40 E^{+02}	94.35 E^{+03}	2.36	3.95
									Loc.	Sc.
15	t_{event}	Gamma	0.00	30.00 E^{+01}	15.03 E^{+01}	23.87	15.12 E^{+01}	27.38 E^{+01}	25.52	4.37
									Loc.	Sc.
	EF	Gamma	0.00	54.00	17.94	14.83	13.91	47.11	−0.11	41.31
	BW	Normal	34.00	74.00	50.30	7.33	50.30	62.10		
	BSA	Normal	12.25 E^{+03}	18.11 E^{+03}	15.00 E^{+03}	95.92 E^{+01}	15.00 E^{+03}	16.59 E^{+03}		

	BP	Triangular	0.07	1.00	0.50	0.19	0.49	0.84	Mo. 0.44	
	ESA	Beta	11.39 E^{+02}	16.06 E^{+03}	75.03 E^{+02}	28.73 E^{+02}	74.16 E^{+02}	12.67 E^{+03}	α 2.94	β 4.24
20	t_{event}	Beta	0.00	24.00 E^{+01}	46.63	49.78	29.30	15.17 E^{+01}	α 0.42	β 3.25
	EF	Beta	0.00	54.00	11.59	13.26	5.96	41.93	Loc. −0.11	Sc. 57.72
	BW	Normal	50.00	85.00	65.50	7.60	65.50	78.47		
	BSA	Normal	15.24 E^{+03}	20.70 E^{+03}	17.82 E^{+03}	98.36 E^{+01}	17.84 E^{+03}	19.47 E^{+03}		
	BP	Triangular	0.07	1.00	0.51	0.19	0.49	0.85	Mo 0.45	
	ESA	Beta	12.96 E^{+02}	19.13 E^{+03}	90.25 E^{+02}	34.54 E^{+02}	86.92 E^{+02}	14.86 E^{+03}	α 2.94	β 4.17

[a]Duration of the bath event (minutes); [b]annual frequency of bathing days (days); [c]bodyweight (kg); [d]body surface area (cm^2); [e]bath pattern (dimensionless); [f]exposed skin area (cm^2); [g]mode.

water is a function of the event bath duration, as can be viewed in Eq. (4).

$$\mathrm{ESA} = \mathrm{BSA} \times \mathrm{BP} \tag{4}$$

where ESA = exposed skin area (cm^2), BP = bath pattern (dimensionless).

Based on direct observation of bathers' behavior in the Del Azul bath resort, it can be said that during half of the bath event, only their legs and hands were inside the water. Only during brief moments, were the bathers fully submerged (swimming) or only with their feet in the water (wading). BP was calculated by age group as the percentage of the underwater body during the bath event based on the recommended values for surface area of body parts from the USEPA (2004). A triangular probability distribution was assumed, by age group, in which the minimum value corresponds to the percentage of the feet in respect to the entire body surface area, the mode to the feet+legs+hands, and the maximum when the entire body was totally submerged. Shown in Table 2 are the model's parameters used for ESA, BSA, and BP calculations.

Dermal dose absorbed

The absorbed dose per event (DA_{event}) was estimated based on a steady-state approach from the USEPA (2004). For inorganic substances, DA_{event} was calculated by Eq. (5).

$$DA_{event} = t_{event} \times K_p \times \mathrm{Conc} \tag{5}$$

where t_{event} = event duration (hr $event^{-1}$), K_p = stratum corneum permeability coefficient of the substance (cm hr^{-1}), estimated based on molecular weight (Mw, in gr) and the coefficient of octanol-water partition (Kow, dimensionless), as shown in Eq. (6) (USEPA 2004).

$$\mathrm{Log}\, K_p = -2.80 + 0.66 \log \mathrm{Kow} - 0.0061\mathrm{Mw} \tag{6}$$

where Conc = concentration of the hazardous substance in water (mg mL^{-1})

For organic substances, DA_{event} was estimated by Eq. (7) or by Eq. (8). If the duration of event (t_{event}) was less than the time to reach steady state for the absorption of the substance through the skin (t^*), Eq. (7) was applied (short exposure duration); if the event took longer (longer exposure duration) Eq. (8) was used (USEPA 2004). For short exposure duration, DA_{event} is proportional to the K_p, but if the exposure is long, we need to consider the contribution of the permeability of the viable epidermis (USEPA 2004). B is the dimensionless ratio of the permeability coefficient of a compound through the stratum corneum relative to its permeability coefficient across the viable epidermis, and is calculated by using Eq. (9) (USEPA 2004).

$$DA_{event} = 2 \times FA \times K_p \times \mathrm{Conc} \times (6 \times \tau \times t_{event})/\pi)^{-0.5} \tag{7}$$

$$DA_{event} = FA \times K_p \times \mathrm{Conc} \times [t_{event}/(1+B) + 2\tau((1+3B+3B^2/(1+B)^2)] \tag{8}$$

$$B = K_p(MW^{0.5}/2.6) \tag{9}$$

where DA_{event} = absorbed dose per event (mg cm^{-2} $event^{-1}$), FA = fraction absorbed (dimensionless), Conc = concentration of the hazardous substance in water (mg L^{-1}), t_{event} = event duration (hr $event^{-1}$), τ = lag time per event (hr $event^{-1}$), calculated based on the thickness and the effective diffusion coefficient for chemical

transfer through the stratum corneum of the skin (USEPA 2004), as it can be viewed in Eq. (10)

$$\tau = l_{sc}^2/6 \times D_{sc} \tag{10}$$

where l_{sc}: apparent thickness of stratum corneum (cm), assumed $1.0E^{-03}$ cm, D_{sc}: Effective diffusion coefficient for chemical transfer through the stratum corneum (cm^2/h), estimated based on Eq. (11) (USEPA 2004).

$$D_{sc} = l_{sc} \times 10^{(-2.8-0.0056\,Mw)} \tag{11}$$

The time to reach a steady state for the skin absorption (t^*) is 2.4 times the lag time per event (τ) given that $B \leq 0.6$ (USEPA 2004). The absorbed dose per event and other terms of Eqs. (5)–(11) are presented in Table 3.

Probabilistic Risk Calculation

The non-cancer R and cancer R of individual substances were calculated first for both contact pathways (dermal and ingestion) individually and then for the aggregated exposure. Subsequently, groups of substances were formed assuming identical toxicological effect. The cumulative aggregated health risk was calculated by applying an additive model for both types of effects. The groups were: heavy metals (As, Cu, Cr, Hg, Zn) and chlorinated pesticides (α, β, δ and γ–hexachlorociclohexane (HCH), aldrin, endosulfan, endosulfan sulphate, γ–chlordane, heptachlor). Moreover, the complete set of substances was considered as a group (all the substances), representing a hypothetical worst case scenario.

The risk calculation was made with Crystal Ball 7.1 software (Decisioneering 2007a), applying Monte Carlo for simple random sampling for 5,000 trials based on the probability distribution of each variable. During a single trial, one value was randomly selected based on the defined probability distribution for each variable, and then the output of the model was calculated. Monte Carlo output is a new distribution of probabilities that includes the variability and uncertainty in the inputs of the parameters. For each R distribution obtained, the arithmetic mean, the standard deviation, the maximum value, and the 95th percentile were estimated as risk indicators. The analysis of the risk results was made based on the 95th percentile of the distribution as high-end risk indicator (semi-conservative approach), as it can be viewed in Figure 3.

Shown in Table 3 are the toxicological reference values of the substances by oral intake and skin contact, for both non-cancer and cancer effects, respectively. The RfD and SF used for accidental water intake risk calculation were obtained from USEPA's IRIS database (2010b). The RfD and SD for dermal risk calculation were estimated based on USEPA (2004), following Eqs. (12) and (13), because chemical-specific dermal toxicity factors are not available.

$$RfD_{derm} = RfD_{in} \times ABS_{GI} \tag{12}$$

$$SD_{derm} = SD_{in}/ABS_{GI} \tag{13}$$

where RfD_{derm} and SD_{derm} = dermal reference dose (mg kg^{-1} day^{-1}) and dermal slope factor (kg day mg^{-1}), RfD_{in} and SD_{in} = intake reference dose (mg kg^{-1} day^{-1}) and intake slope factor (kg day mg^{-1}), ABS_{GI} = fraction of contaminant absorbed in gastrointestinal tract (dimensionless) in the critical toxicity study.

Table 3. Parameters used in the absorbed dose per event calculation by age group and toxicological reference values of the substances for oral and dermal contact exposure pathways.

								DA_{event} [h]				RfD [i]		SF [j]		
Substances	MW[a]	Log KOW [b]	Kp[c]	B[d]	τ[e]	T*[f]	FA[g]	5	10	15	20	Int.[l]	Derm. [m]	Int.	Derm.	ABS_{GI} [k]
2,4 D (2,4 Dichloro phenoxy acetic acid)	2.21 E^{+02}	2.62	4.94 E^{-03}	2.83 E^{-02}	1.85	4.44	1.00	2.33 E^{-07}	2.68 E^{-07}	2.44 E^{-07}	1.16 E^{-07}	1.00 E^{-02}	9.00 E^{-03}	N.A.	N.A.	9.00 E^{-01}
α–HCH (Hexachloro cyclohexane)	2.91 E^{+02}	4.26	2.48 E^{-02}	1.62 E^{-01}	4.56	10.94	0.90	9.52 E^{-09}	1.10 E^{-08}	1.00 E^{-08}	5.55 E^{-09}	3.00 E^{-04}	3.00 E^{-04}	6.30 E^{+00}	6.30 E^{+00}	1.00 E^{+00}
β–HCH	2.91 E^{+02}	4.26	2.48 E^{-02}	1.62 E^{-01}	4.56	10.94	0.90	8.61 E^{-10}	9.55 E^{-10}	9.08 E^{-10}	5.99 E^{-10}	3.00 E^{-04}	3.00 E^{-04}	1.80 E^{+00}	1.80 E^{+00}	1.00 E^{+00}
σ–HCH	2.91 E^{+02}	4.26	2.48 E^{-02}	1.62 E^{-01}	4.56	10.94	0.90	8.08 E^{-10}	9.87 E^{-10}	8.93 E^{-10}	4.16 E^{-10}	3.00 E^{-04}	3.00 E^{-04}	1.30 E^{+00}	1.30 E^{+00}	1.00 E^{+00}
γ–HCH	2.91 E+02	4.26	2.48 E^{-02}	1.62 E^{-01}	4.56	10.94	1.00	8.15 E^{-10}	9.23 E^{-10}	8.68 E^{-10}	5.12 E^{-10}	3.00 E^{-04}	3.00 E^{-04}	1.30 E^{+00}	1.30 E^{+00}	1.00 E^{+00}
Acetochlor	2.70 E^{+02}	3.37	8.33 E^{-03}	5.26 E^{-02}	3.47	8.34	1.00	2.83 E^{-06}	3.27 E^{-06}	3.04 E^{-06}	1.93 E^{-06}	2.00 E^{-02}	2.00 E^{-02}	N.A.	N.A.	1.00 E^{+00}
Aldrin	3.65 E^{+02}	3.01	1.40 E^{-03}	1.03 E^{-02}	11.88	28.51	0.70	6.15 E^{-11}	7.65 E^{-11}	6.48 E^{-11}	3.44 E^{-11}	3.00 E^{-05}	3.00 E^{-05}	1.70 E^{+01}	1.70 E^{+01}	1.00 E^{+00}
Arsenic			1.00 E^{-03}					1.59 E^{-07}	1.87 E^{-07}	1.69 E^{-07}	8.00 E^{-08}	3.00 E^{-04}	3.00 E^{-04}	1.50 E^{+00}	1.50 E^{+00}	1.00 E^{+00}
Cypermethrin	4.16 E^{+02}	6.38	1.26 E^{-01}	9.85 E^{-01}	23.08	88.98	0.60	3.17 E^{-04}	3.84 E^{-04}	3.32 E^{-04}	1.31 E^{-04}	1.00 E^{-02}	1.00 E^{-02}	N.A.	N.A.	1.00 E^{+00}
Chlorpyrifos	3.51 E^{+02}	4.66	2.11 E^{-02}	1.52 E^{-01}	9.87	23.69	0.90	1.36 E^{-07}	1.60 E^{-07}	1.43 E^{-07}	7.58 E^{-08}	3.00 E^{-03}	3.00 E^{-03}	N.A.	N.A.	1.00 E^{+00}
Copper			1.00 E^{-03}					3.19 E^{-08}	3.95 E^{-08}	3.57 E^{-08}	1.63 E^{-08}	4.00 E^{-02}	4.00 E^{-02}	N.A.	N.A.	1.00 E^{+00}
Chromium			1.00 E^{-03}					1.94 E^{-08}	2.34 E^{-08}	2.12 E^{-08}	1.02 E^{-08}	1.50 E^{+00}	1.95 E^{-02}	N.A.	N.A.	1.30 E^{-02}
Endosulfan	4.07 E^{+02}	3.5	1.73 E^{-03}	1.34 E^{-02}	20.44	49.05	0.90	1.31 E^{-10}	1.48 E^{-10}	1.37 E^{-10}	8.48 E^{-11}	6.00 E^{-03}	6.00 E^{-03}	N.A.	N.A.	1.00 E^{+00}
Endosulfan sulphate	4.23 E^{+02}	3.64	1.74 E^{-03}	1.38 E^{-02}	25.13	60.32	0.90	1.00 E^{-09}	1.11 E^{-09}	1.06 E^{-09}	6.60 E^{-10}	6.00 E^{-03}	6.00 E^{-03}	N.A.	N.A.	1.00 E^{+00}
γ–Chlordane	4.10 E^{+02}	5.54	3.78 E^{-02}	2.94 E^{-01}	21.21	50.90	0.60	9.32 E^{-10}	1.07 E^{-09}	9.72 E^{-10}	5.80 E^{-10}	5.00 E^{-04}	4.00 E^{-04}	2.50 E^{-04}	4.38 E^{-01}	8.00 E^{-01}
Glyphosate	1.69 E^{+02}	−4.47	1.89 E^{-07}	9.43 E^{-07}	0.95	2.27	1.00	3.24 E^{-10}	3.96 E^{-10}	3.42 E^{-10}	1.97 E^{-10}	1.00 E^{-01}	1.00 E^{-01}	N.A.	N.A.	1.00 E^{+00}
Heptachlor	3.73 E^{+02}	5.86	9.87 E^{-02}	7.34 E^{-01}	13.24	51.19	0.60	3.78 E^{-08}	4.32 E^{-08}	4.00 E^{-08}	2.48 E^{-08}	5.00 E^{-04}	5.00 E^{-04}	4.50 E^{+00}	4.50 E^{+00}	1.00 E^{+00}
Mercury			1.00 E^{-03}					5.69 E^{-09}	6.57 E^{-09}	6.07 E^{-09}	2.84 E^{-09}	3.00 E^{-04}	3.00 E^{-04}	N.A.	N.A.	1.00 E^{+00}
Xylenes	1.06 E^{+02}	3.09	4.49 E^{-02}	1.78 E^{-01}	0.42	1.01	1.00	4.07 E^{-05}	4.66 E^{-05}	4.36 E^{-05}	2.37 E^{-05}	2.00 E^{-01}	2.00 E^{-01}	N.A.	N.A.	1.00 E^{+00}
Zinc			6.00 E^{-04}					4.88 E^{-08}	5.93 E^{-08}	5.29 E^{-08}	2.38 E^{-08}	3.00 E^{-01}	3.00 E^{-01}	N.A.	N.A.	1.00 E^{+00}

[a]Molecular weight (gr); [b]log of the coefficient of octanol-water partition (dimensionless); [c]dermal permeability coefficient of the compound in water (cm hr^{-1}); [d]ratio of the permeability of a compound through the stratum corneum relative to the viable epidermis; [e]lag time per event (hr $event^{-1}$); [f]time to reach steady state of the skin absorption (h); [g]fraction absorbed (dimensionless), based on USEPA (2004); [h]95th percentile of the absorbed dose per event (mg cm^{-2} $event^{-1}$) by age group; [i]reference dose (mg kg^{-1} day^{-1}); [j]slope factor (kg day mg^{-1}); [k]contaminant fraction absorbed in gastrointestinal tract (dimensionless), based on USEPA (2004); [l]oral intake; [m]dermal.

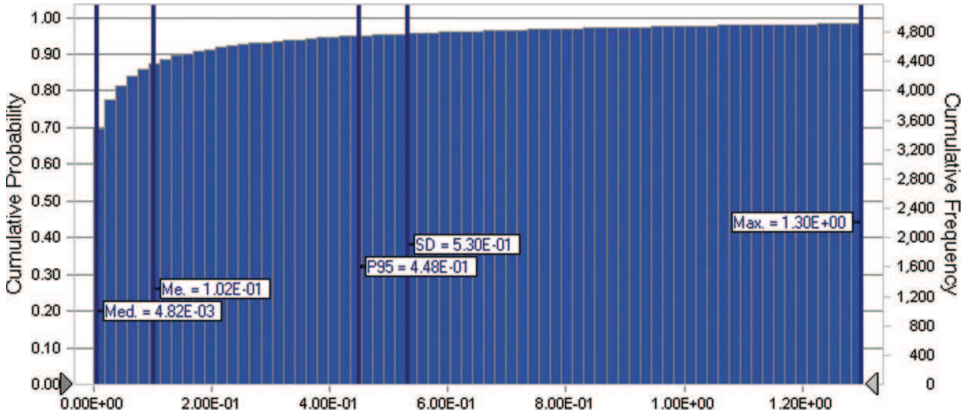

Figure 3. Cumulative probability distribution of risk showing the arithmetic mean (Me), the standard deviation (SD), the median (Med), the 95th percentile (P95), and the maximum value (Max). (Color figure available online.)

Statistical Analysis

All mathematical calculations such as means, standard deviations, medians, 95th percentiles, percentages, minimums, and maximums were calculated using Excel 2007 (Microsoft Office). The Kruskal-Wallis test was used to determine if the risks scores differed between age subgroups, testing the difference between the medians of three or more independent non-normal subgroups (non-parametric test). The null hypothesis is that the mean ranks of the k groups will not substantially differ. In our study, p-values $< .05$ were considered to indicate a significant difference between the compared subgroups. This analysis was conducted using Statistica 7.0 (Statsoft 2004).

Sensitivity Analysis

To know the contribution of each exposure parameter to the R a sensitivity analysis was made with Crystal Ball 7.1. (Decisioneering 2007a). This calculates sensitivity based on the rank correlation coefficients between every parameter of the model and the model's results while the simulation is running (Decisioneering 2007b).

RESULTS

Non-Carcinogenic Risk

From Table 4 it can be observed that the R from the aggregated exposure (accidental water intake + dermal contact) of Del Azul Creek water for non-cancer substances never reached the assumed acceptability criteria of 1.0. The riskiest substance for all of the age groups was cypermethrin, followed by arsenic.

Studying the non-cancer R of the different substance groups, the highest cumulative risks obviously arise from the All the Substances group, at almost one order of magnitude less than the safe criteria. All age groups had a gap between the R

Table 4. Non-carcinogenic risks by age, through aggregated exposure by individual and by groups of substances and descriptive statistics of the dose of exposure (in mg kg^{-1} d^{-1}) for intake and dermal routes for the age group with greatest non-cancer cumulative risk.

		10 Years Old												
			Derm.					Int.						
Substances	5 Years Old[a]	Aggr. [b]	Me.	SD	Max.	Med.	P95	Me.	SD	Max.	Med.	P95	15 Years Old	20 Years Old
2,4 D	1.35 E^{-02}	1.31 E^{-02}	2.25 E^{-05}	2.39 E^{-05}	2.00 E^{-04}	1.48 E^{-05}	7.11 E^{-05}	1.96 E^{-05}	1.97 E^{-05}	1.82 E^{-04}	1.34 E^{-05}	5.91 E^{-05}	9.39 E^{-03}	3.30 E^{-03}
α–HCH	8.44 E^{-04}	9.35 E^{-04}	6.98 E^{-08}	8.50 E^{-08}	6.60 E^{-07}	3.86 E^{-08}	2.48 E^{-07}	8.59 E^{-09}	9.96 E^{-09}	6.53 E^{-08}	4.91 E^{-09}	2.99 E^{-08}	7.05 E^{-04}	2.68 E^{-04}
β–HCH	5.74 E^{-05}	6.10 E^{-05}	5.66 E^{-09}	5.68 E^{-09}	4.38 E^{-08}	3.86 E^{-09}	1.73 E^{-08}	6.99 E^{-10}	6.59 E^{-10}	4.16 E^{-09}	5.02 E^{-10}	2.02 E^{-09}	4.84 E^{-05}	1.93 E^{-05}
δ–HCH	1.07 E^{-04}	1.19 E^{-04}	1.03 E^{-08}	1.22 E^{-08}	1.26 E^{-07}	6.06 E^{-09}	3.44 E^{-08}	1.26 E^{-09}	1.39 E^{-09}	1.51 E^{-08}	7.76 E^{-10}	4.06 E^{-09}	9.73 E^{-05}	3.56 E^{-05}
γ–HCH	8.80 E^{-05}	9.70 E^{-05}	8.80 E^{-09}	8.69 E^{-09}	6.02 E^{-08}	6.13 E^{-09}	2.63 E^{-08}	9.80 E^{-10}	9.13 E^{-10}	5.31 E^{-09}	7.06 E^{-10}	2.87 E^{-09}	7.79 E^{-05}	3.12 E^{-05}
Acetochlor	2.88 E^{-03}	3.06 E^{-03}	1.24 E^{-05}	1.65 E^{-05}	1.53 E^{-04}	6.29 E^{-06}	4.47 E^{-05}	4.68 E^{-06}	5.91 E^{-06}	5.32 E^{-05}	2.40 E^{-06}	1.68 E^{-05}	2.17 E^{-03}	7.58 E^{-04}
Aldrin	2.47 E^{-04}	2.33 E^{-04}	8.56 E^{-10}	9.08 E^{-10}	9.22 E^{-09}	5.58 E^{-10}	2.66 E^{-09}	1.49 E^{-09}	1.50 E^{-09}	1.16 E^{-08}	1.01 E^{-09}	4.55 E^{-09}	1.53 E^{-04}	4.80 E^{-05}
Arsenic	9.27 E^{-02}	8.25 E^{-02}	7.77 E^{-07}	9.33 E^{-07}	8.61 E^{-06}	4.57 E^{-07}	2.70 E^{-06}	7.03 E^{-06}	7.44 E^{-06}	5.34 E^{-05}	4.55 E^{-06}	2.30 E^{-05}	4.89 E^{-02}	1.08 E^{-02}
Cypermethrin	2.88 E^{-01}	3.95 E^{-01}	9.38 E^{-04}	4.12 E^{-03}	8.76 E^{-02}	4.64 E^{-05}	4.16 E^{-03}	1.46 E^{-05}	6.17 E^{-05}	1.30 E^{-03}	7.68 E^{-07}	6.38 E^{-05}	3.13 E^{-01}	7.74 E^{-02}
Chlorpyrifos	1.34 E^{-02}	1.38 E^{-02}	1.13 E^{-05}	1.50 E^{-05}	1.60 E^{-04}	6.10 E^{-06}	4.03 E^{-05}	1.11 E^{-06}	1.37 E^{-06}	1.22 E^{-05}	6.24 E^{-07}	3.85 E^{-06}	1.10 E^{-02}	4.19 E^{-03}
Copper	1.33 E^{-04}	1.17 E^{-04}	1.59 E^{-07}	1.78 E^{-07}	1.47 E^{-06}	1.02 E^{-07}	5.07 E^{-07}	1.45 E^{-06}	1.46 E^{-06}	1.58 E^{-05}	1.01 E^{-06}	4.19 E^{-06}	7.06 E^{-05}	1.71 E^{-05}
Chromium	1.69 E^{-05}	2.07 E^{-05}	1.08 E^{-07}	1.26 E^{-07}	1.27 E^{-06}	6.56 E^{-08}	3.59 E^{-07}	9.85 E^{-07}	1.02 E^{-06}	7.90 E^{-06}	6.48 E^{-07}	3.03 E^{-06}	1.49 E^{-05}	4.08 E^{-06}
Endosulfan	7.45 E^{-07}	7.38 E^{-07}	7.75 E^{-10}	8.58 E^{-10}	7.10 E^{-09}	4.98 E^{-10}	2.50 E^{-09}	6.49 E^{-10}	6.76 E^{-10}	5.01 E^{-09}	4.35 E^{-10}	2.04 E^{-09}	5.36 E^{-07}	1.78 E^{-07}
Endosulfan sulphate	6.01 E^{-06}	5.79 E^{-06}	7.20 E^{-09}	7.14 E^{-09}	6.07 E^{-08}	4.98 E^{-09}	2.19 E^{-08}	5.39 E^{-09}	5.05 E^{-09}	4.08 E^{-08}	3.88 E^{-09}	1.55 E^{-08}	4.29 E^{-06}	1.54 E^{-06}
γ–Chlordane	6.28 E^{-05}	6.79 E^{-05}	8.97 E^{-09}	8.93 E^{-09}	7.53 E^{-08}	6.19 E^{-09}	2.68 E^{-08}	5.05 E^{-10}	4.71 E^{-10}	3.68 E^{-09}	3.64 E^{-10}	1.47 E^{-09}	5.43 E^{-05}	2.22 E^{-05}
Glyphosate	1.55 E^{-03}	1.43 E^{-03}	1.32 E^{-09}	1.91 E^{-09}	3.33 E^{-08}	6.55 E^{-10}	4.88 E^{-09}	3.98 E^{-05}	5.48 E^{-05}	7.42 E^{-04}	2.05 E^{-05}	1.43 E^{-04}	7.86 E^{-04}	1.57 E^{-04}
Heptachlor	1.01 E^{-03}	1.13 E^{-03}	1.87 E^{-07}	1.85 E^{-07}	1.87 E^{-06}	1.29 E^{-07}	5.66 E^{-07}	5.09 E^{-09}	4.76 E^{-09}	5.91 E^{-08}	3.70 E^{-09}	1.46 E^{-08}	9.19 E^{-04}	3.68 E^{-04}
Mercury	3.48 E^{-03}	3.02 E^{-03}	3.01 E^{-08}	3.38 E^{-08}	2.96 E^{-07}	1.81 E^{-08}	1.00 E^{-07}	2.73 E^{-07}	2.74 E^{-07}	2.00 E^{-06}	1.83 E^{-07}	8.52 E^{-07}	1.81 E^{-03}	4.17 E^{-04}
Xylenes	3.16 E^{-03}	3.84 E^{-03}	1.90 E^{-04}	2.36 E^{-04}	2.31 E^{-03}	1.05 E^{-04}	6.75 E^{-04}	3.27 E^{-05}	3.80 E^{-05}	2.50 E^{-04}	1.89 E^{-05}	1.14 E^{-04}	2.81 E^{-03}	9.54 E^{-04}
Zinc	4.84 E^{-05}	4.16 E^{-05}	2.61 E^{-07}	2.94 E^{-07}	2.60 E^{-06}	1.63 E^{-07}	8.52 E^{-07}	3.94 E^{-06}	3.95 E^{-06}	3.56 E^{-05}	2.73 E^{-06}	1.18 E^{-05}	2.51 E^{-05}	5.97 E^{-06}
Metals	9.61 E^{-02}	8.49 E^{-02}											5.08 E^{-02}	1.12 E^{-02}
Chlorinated pesticides	2.30 E^{-03}	2.46 E^{-03}											1.92 E^{-03}	7.75 E^{-04}
All the sub.	3.63 E^{-01}	4.59 E^{-01}											3.49 E^{-01}	8.79 E^{-02}
Arsenic/ all the sub.	2.55 E^{+01}	1.80 E^{+01}											1.40 E^{+01}	1.23 E^{+01}
Cypermethrin/ all the sub.	7.94 E^{+01}	8.62 E^{+01}											8.96 E^{+01}	8.82 E^{+01}
Arsenic/ heavy metals	9.65 E^{+01}	9.72 E^{+01}											9.64 E^{+01}	9.65 E^{+01}
Cypermethrin/ arsenic	3.11	4.79											6.40	7.16

[a]Years old; [b]aggregated exposure.

from heavy metals and chlorinated pesticides (at one order of magnitude, approximately). From the two riskiest substances before mentioned, one belongs to the metal group (arsenic), and the other is not a chlorinated pesticide. As shown in the table, cypermethrin aggregated risk represented the 80% of the All the Substances risk, and arsenic contributed with the remaining 20%, which was greater than 95% of the aggregated risk of the metals group.

Running the Kruskal-Wallis test on the all the substances cumulative and aggregated R of the Monte Carlo trials showed a significant difference between the four age groups ($p < .05$). The order of importance of the All the Substances aggregated non-cancer R by age group was 10 > 5 > 15 > 20 years old. Cypermethrin generated the highest risk for the 10-years-old group and arsenic for the 5-years-old group. The non-cancer R of the first substance was from 3 to 7 times greater than the second (from the 5 to the 20 years old age groups). Although the cypermethrin/all the substances risk ratio increased according to age, the arsenic/all the substances ratio decreased.

Analyzing the percentage ratios of the differences between the non-cancer R of all the age groups, Del Azul water quality was almost 70% riskiest for the youngest children. The 10-years-old group was at greater risk than the other age groups: the all the substances R for this age group was almost 25–30% greater than the 5- and 15-years-old groups, and 90% greater than the 20-years-old group.

Shown in Figure 4 are the arsenic, cypermethrin, and cumulative all the substances group non-cancer risk scores separately for both pathways of exposure, by age group. It was evident that the relative importance of the dermal contact to aggregated non-cancer and cumulative risk, whose values were never less than 80%

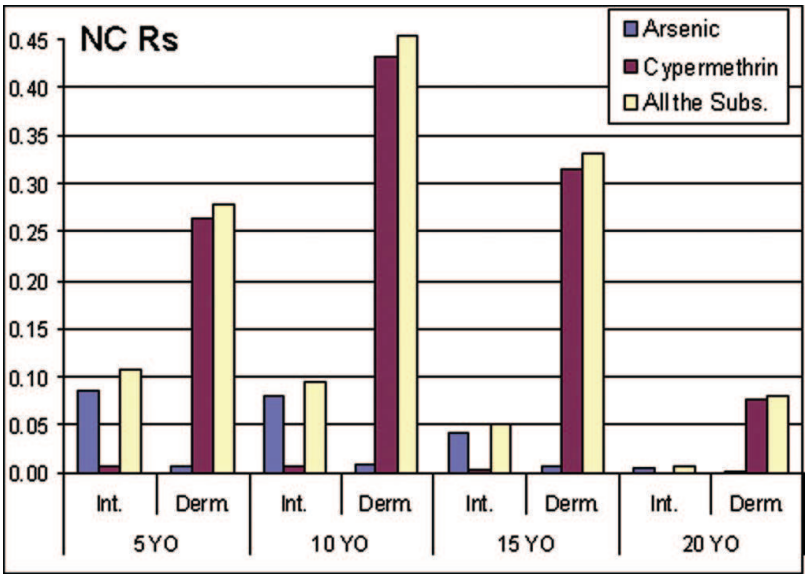

Figure 4. Non-cancer risk scores (NC Rs) for arsenic, cypermethrin, and cumulative all the substances group for intake and dermal contact, by age group. (Color figure available online.)

of the aggregated exposure of each age group, but had increased almost 90% in the older groups. On the other hand, intake decreased the relative importance from 20% to 10%. Obviously, if the absolute values of the risk scores were considered, the figure would clearly show the importance of the cypermethrin on the dermal and the arsenic on the intake route. Although cypermethrin is 667 times less toxic for the dermal route and arsenic 10 times for the oral route than the most toxic substance found in Del Azul water samples (aldrin for both routes, as can be estimated based on RfDs on Table 3), the dose for these substances was greater if compared with other substances. This can be appreciated in Table 4, where the dermal and intake dose of exposure of each substance for the 10-years-old age group are displayed. Cypermethrin had the greatest dermal and oral dose, but arsenic had the 5th greatest intake dose (cypermethrin > glyphosate > xylenes > 2,4-D > arsenic).

To determine the importance of each parameter of the exposure estimation in the risk values for each age group, it was necessary to appreciate the results of the sensitivity analysis. Shown in Table 5 is the percentage of the contribution to the variance of the non-cancer risk by age from the exposure parameters for aggregated, and for oral and dermal pathways for all the substances cumulative group.

For aggregated exposure, the concentration of the key dangerous substances (cypermethrin) explained almost 20% of the entire variance (5- and 20-years-old group, even lower). The frequency of the exposure generated the biggest contribution to the variances (more than 60% for all of the age groups). From the three variables related to the behavior of bathers (frequency of the exposure (*EF*), event duration (t_{event}), and bath pattern), only *EF* and t_{event} had relevant contributions: *EF* + t_{event} explained more than 70% of the variance for the four age groups, greater in the 5- and 20-years-old groups. Analyzing the results by exposure pathway, *EF* and t_{event} by intake were greater than by the dermal route for the four age groups. The Bath Pattern was only present for the dermal route, but with minor importance.

On the other hand, it was clear that cypermethrin was relevant for the dermal route and arsenic, when it appears in sensitivity analysis, for the intake route, as seen in Figure 4. Other substances were absent from the sensitivity analysis. The age group with major variance contribution for these substances was the 10-years-old group. Given the fact that *EF* and t_{event} for the 10-years-old group had the greatest descriptive statistic values (mean, median, 95th percentile, and maximum, in Table 2) as well as the biggest variance contribution for the cypermethrin concentrations (Table 5), this could explain why the 10-years-old group had the greatest aggregated non-cancer and cumulative R (Table 4).

Carcinogenic Risk

Because the model and data for the calculation of the dose between non-cancer and cancer R were common, the patterns of the results from the cancer risk calculation were obviously similar to the non-cancer risk in many aspects.

The cancer R scores through aggregated exposure are provided in Table 6. It was observed that no substance by itself reached the assumed criterion health concern risk level of $1.0E^{-05}$. Only all the substances cumulative cancer R for the 5-years-old age group reached the health concern risk level, although only slightly above. Arsenic, a carcinogenic metal, was described as the riskiest substance, generating between 80 and 90% of all the substances cancer R. However, the chlorinated

Table 5. Contributions to the variance of the non-cancer R by age of the exposure parameters for aggregated, intake and dermal contact pathways for all the substances cumulative group.

	5 Years Old			10 Years Old			15 Years Old			20 Years Old		
All the Substances	Aggr.	Int.	Derm.	Aggr.	Int.	Derm.	Aggr	Int.	Derm.	Aggr.	Int.	Derm.
EF	70.64	73.71	57.61	64.35	74.24	47.85	61.80	70.58	51.67	64.70	58.69	62.50
t_{event}	11.59	19.97		6.17	13.26	6.22	11.04	20.85	6.09	15.60	37.58	9.97
Cypermethrin conc.	13.40		30.13	23.24	9.70	41.23	22.24		35.21	16.50		22.76
Arsenic conc.		5.00						6.55				
Bath pattern			5.63						5.37			
Other	4.38	1.31	6.62	6.22	2.79	4.69	4.90	2.02	1.74	3.20	3.71	4.75

Table 6. Carcinogenic risks by age through aggregated exposure by individual and by groups of substances and descriptive statistics of the dose of exposure (in mg kg^{-1} d^{-1}) for intake and dermal routes for the age group with greatest C cumulative risk.

	5 Years Old													
		Derm.					Int.							
Substances	Aggr.	Me.	SD	Max.	Med.	P95	Me.	SD	Max.	Med.	P95	10 Years Old	15 Years Old	20 Years Old
α–HCH	3.51 E^{-07}	1.18 E^{-08}	1.85 E^{-08}	1.71 E^{-07}	4.45 E^{-09}	4.79 E^{-08}	1.84 E^{-09}	2.92 E^{-09}	3.55 E^{-08}	6.61 E^{-10}	7.67 E^{-09}	4.01 E^{-07}	3.12 E^{-07}	1.05 E^{-07}
β–HCH	7.18 E^{-09}	9.51 E^{-10}	1.28 E^{-09}	1.45 E^{-08}	4.68 E^{-10}	3.64 E^{-09}	1.49 E^{-10}	2.06 E^{-10}	2.02 E^{-09}	6.94 E^{-11}	5.63 E^{-10}	7.90 E^{-09}	6.23 E^{-09}	2.25 E^{-09}
δ–HCH	9.71 E^{-09}	1.70 E^{-09}	2.56 E^{-09}	3.71 E^{-08}	7.51 E^{-10}	6.64 E^{-09}	2.64 E^{-10}	3.99 E^{-10}	5.20 E^{-09}	1.12 E^{-10}	1.06 E^{-09}	1.13 E^{-08}	8.20 E^{-09}	3.05 E^{-09}
γ–HCH	7.85 E^{-09}	1.49 E^{-09}	2.01 E^{-09}	2.13 E^{-08}	7.30 E^{-10}	5.63 E^{-09}	2.09 E^{-10}	2.87 E^{-10}	2.66 E^{-09}	9.80 E^{-11}	7.97 E^{-10}	8.79 E^{-09}	6.87 E^{-09}	2.44 E^{-09}
Aldrin	2.92 E^{-08}	1.43 E^{-10}	2.03 E^{-10}	2.12 E^{-09}	6.72 E^{-11}	5.53 E^{-10}	3.13 E^{-10}	4.45 E^{-10}	4.40 E^{-09}	1.39 E^{-10}	1.20 E^{-09}	2.88 E^{-08}	1.82 E^{-08}	5.01 E^{-09}
Arsenic	9.42 E^{-06}	1.15 E^{-07}	1.94 E^{-07}	3.16 E^{-06}	4.30 E^{-08}	4.78 E^{-07}	1.52 E^{-06}	2.31 E^{-06}	2.64 E^{-05}	6.41 E^{-07}	6.01 E^{-06}	8.74 E^{-06}	4.97 E^{-06}	1.01 E^{-06}
γ–Chlordane	2.57 E^{-09}	1.52 E^{-09}	2.03 E^{-09}	2.00 E^{-08}	7.51 E^{-10}	5.54 E^{-09}	1.08 E^{-10}	1.47 E^{-10}	1.17 E^{-09}	5.08 E^{-11}	4.10 E^{-10}	2.92 E^{-09}	2.27 E^{-09}	8.36 E^{-10}
Heptachlor	5.23 E^{-07}	3.15 E^{-08}	4.21 E^{-08}	3.67 E^{-07}	1.57 E^{-08}	1.16 E^{-07}	1.09 E^{-09}	1.49 E^{-09}	1.59 E^{-08}	5.07 E^{-10}	3.98 E^{-09}	5.97 E^{-07}	4.82 E^{-07}	1.76 E^{-07}
Metals	9.42 E^{-06}											8.74 E^{-06}	4.97 E^{-06}	1.01 E^{-06}
Chlorinated pesticides	9.00 E^{-07}											1.00 E^{-06}	8.17 E^{-07}	2.85 E^{-07}
All the sub.	1.03 E^{-05}											9.52 E^{-06}	5.70 E^{-06}	1.29 E^{-06}
Metals (As)/ all the sub.	9.13 E^{+01}											9.18 E^{+01}	8.72 E^{+01}	7.81 E^{+01}
Chlorinated Pesticides/ all the sub.	8.73 E^{+00}											1.05 E^{+01}	1.43 E^{+01}	2.21 E^{+01}
Heptachlor/ all the sub.	5.07 E^{+00}											6.27 E^{+00}	8.46 E^{+00}	1.37 E^{+01}

pesticides cancer R was less, from 10 to 20%. The riskiest chlorinated pesticide was heptachlor.

The Kruskal-Wallis test showed that the differences of the cancer R between age groups were significant ($p < .05$). Meanwhile, for the 5- and 10-years-old age groups heavy metals (arsenic) generated almost 95% of the C cumulative all the substances risk. For 20 years old, it caused only 83%. The chlorinated pesticides were responsible for almost 5 to 20% of the all the substances cancer R.

However, unlike non-cancer aggregated risk, the youngest age group had the major cancer R, followed by those of the 10-years old age group. The order of importance of the all the substances aggregated cancer R by age group was 5 > 10 > 15 > 20 years old. Despite the increase of the chlorinated pesticides R for the 10-years-old age group, it was not enough to offset the decline of all the substances R, given by heavy metals (arsenic) R decrease. This can be clearly viewed in Figure 5, described below.

As it occurred in non-cancer risk, the younger children were at greater risk than the older groups, and the range of the all the substances R between the greatest and the lowest value was about 90%. Analyzing separately the R generated by pathways of exposure (Figure 5), it was clear that the relative importance of the intake pathway with respect to the dermal route was inverse to the non-cancer R. Although arsenic was the major contributor to the all the substances cancer risk, as previously stated, this substance had lesser SF than the majority of the chlorinated pesticides. In other words, arsenic is less toxic in terms of C effects than the others (heptachlor is three times more toxic, as it is shown in Table 3). Analyzing the dose of exposure (Table 6), the dose of arsenic was 1500 times greater that for heptachlor.

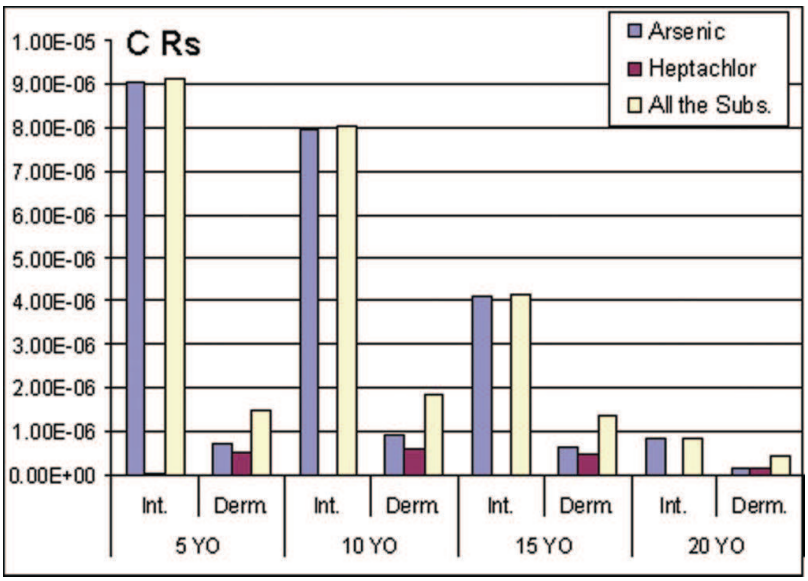

Figure 5. Cancer risk scores (C Rs) for arsenic, heptachlor, and cumulative all the substances group for intake and dermal contact, by age group. (Color figure available online.)

Table 7. Contributions to the variance of the cancer R by age of the exposure parameters for aggregated, intake and dermal pathways of contact for all the substances cumulative group.

	5 Years Old			10 Years Old			15 Years Old			20 Years Old		
All the Substances	Aggr.	Int.	Derm.	Aggr.	Int.	Derm.	Aggr.	Int.	Derm.	Aggr.	Int.	Derm.
EF	67.90	65.30	73.20	67.60	65.50	66.70	67.90	64.40	69.90	64.20	55.40	71.40
t_{event}	17.90	19.50	10.20	10.10	11.00	5.90	14.50	16.50	8.70	29.10	37.60	18.90
Arsenic conc.	6.40	7.80		8.10	10.30		6.40	8.80			2.40	
ED	6.50	6.30	6.90	11.10	10.70	11.10	9.00	8.30	9.80	4.00	3.40	4.50
Bath pattern			6.70			12.10			7.60			3.30
Other	1.20			3.00	2.50	4.20	2.30	2.00	3.90	2.70		1.90

The results of the sensitivity analysis of the cancer R, presented in Table 7, showed similar variance contributions of the variables to those of non-cancer. However, some differences may be mentioned. Although $EF + t_{event}$ were still the main variance contributions, the values were a little greater than the non-cancer R, and t_{event} was increased. Arsenic was the substance with the biggest variance contribution, with greater values than that for the non-cancer risk. Unlike the case of non-cancer R, the duration of the exposure (*ED*) for CR contributed to the variance, although the values are low.

DISCUSSION

Many scientific papers can be found in which health risk assessment based on USEPA models are applied based on the exposure to individual substances or to several in a cumulative way by individual pathways of contact or by aggregated exposure. The most frequent papers are on contaminants in water used for intake (Zakharova *et al*. 2002; Wang *et al*. 2007; Hamidin *et al*. 2008; Zabin *et al*. 2008; Brown and Foos 2009; Cunningham *et al*. 2009; Fan *et al*. 2009; Kavcar *et al*. 2009; Nguyen *et al*. 2009; Kumar and Xagoraraki 2010; Phang *et al*. 2010; Muhammad *et al*. 2010; Legay *et al*. 2011; Muhammad *et al*. 2011; Wang *et al*. 2011), in food (Huang *et al*. 2006; Liu *et al*. 2007; Lim *et al*. 2008; Khan *et al*. 2008; Sipter *et al*. 2008; Sofuoglu and Kavcar 2008; Sanderson *et al*. 2009; Urban *et al*. 2009; Liang *et al*. 2011), in air (Guo *et al*. 2004; Liao and Chiang 2006; Hu *et al*. 2007; Kao *et al*. 2007; Lonati *et al*. 2007; Li *et al*. 2008; Chowdhury and Champagne 2009; Li *et al* 2009; Durmusoglu *et al*. 2010; Zheng *et al*. 2010; Shi *et al*. 2011; Zhou *et al*. 2011), in soils (Korre *et al*. 2002; Ferreyra-Baptista *et al*. 2005; Gay and Korre 2006; Lim *et al*. 2008; Zhang *et al*. 2009; Yang *et al*. 2009; Man *et al*. 2010; Guney *et al*. 2010), or in multimedia (Xenidis *et al*. 2003; Morra *et al*. 2006; Uyak 2006; Hang *et al*. 2009; Zeng *et al*. 2009; Davoli *et al*. 2010; Fan *et al*. 2010).

The application of risk assessment based on chemicals in outdoor recreational waters is infrequent in published literature, although some can be cited: Albering *et al*. (1999); Filipsson *et al*. (2009). All the studies found by us were based on USEPA models and were focused on heavy metals (Goldblum *et al*. 2006), organic chemicals (Hussain *et al*. 1998; Baars 2002; Dor *et al*. 2003; Blando and Cohn 2004; Kumar and Xagoraraki 2010), or both (Albering *et al*. 1999; Filipsson *et al*. 2009) as contaminants in water. No papers were found regarding risk assessment for pesticides in recreational waters.

The non-cancer and cancer R showed that the recreational use of Del Azul water did not result in a threat to human health. Although the aggregated and cumulative cancer R reached the cancer health risk level criterion, we consider the health of bathers was not at significant risk. In literature, safe limits can be found for C risk on recreational bathing between $1.0E^{-06}$ (*e.g.*, USEPA) and $1.0E^{-04}$ (Baars 2002). The most restrictive recreational safe criteria were based on the USEPA drinking water limit of $1.0E^{-06}$, which is a very conservative approach to be applied on recreational accidental water intake. On the other hand, the USEPA stated that the cumulative risk assessment is a conservative overestimate of the risk generated by individual substances, and that "exceeding the acceptable levels does not imply the

expectation of toxic effects but only that a more detailed risk assessment is needed" (USEPA 2007, pp 5–30), as it is suggested below as a recommendation.

Our study has shown that the risk scores were different according to the age of the exposed individual; therefore, the importance of considering the variability of a population's characteristics in risk assessment has been highlighted in many papers. Thus, the variability was considered according to the age in studies focused on the ingestion of drinking water (Zhao and Kaluarachchi 2002; Brown and Foos 2010), on accidental intake during recreational bathing (Albering *et al*. 1999; Filipsson *et al*. 2009), on inhalation dosimetry (Liao *et al*. 2008; Shi *et al*. 2011), or on ingestion of food (Moon *et al*. 2009). However, the main variation considered was child versus adult. Only in the case of drinking water (Brown and Foos 2010) or diet (Moon *et al*. 2009), were age ranges considered. In Filipsson *et al*. (2009), for the three age groups of childhood (5, 10, and 15 years old) 0.054 L h^{-1} was used, indicating that the methodological decision was based on Dufour *et al*. (2006). These values are very close to the accidental intake, which is 0.05 L h^{-1}, used in our study. The common practice of USEPA is to use 50 mL of water ingested per hour of recreational activity (USEPA 1989) as the deterministic parameter and without discriminating between children and adults.

Taking into account three ranges of age for children allowed discriminating that the 10-years-old non-cancer aggregated R was greater than for the youngest children's group. Given that the accidental water intake was the main pathway of exposure to metals (O'Rourke *et al*. 1999; Filipsson *et al*. 2009), and of more importance than dermal exposure (Goldblum *et al*. 2006), it might be necessary to conduct a study of the true intake rate by age group rather than using a deterministic common value for more accurate metals' risk.

On the other hand, organics as major contributors of risk through dermal pathway are cited in many papers where recreational bathing was considered (Dor *et al*. 2003; Blando and Cohn 2004; Papanyako *et al*. 2008; Filipsson *et al*. 2009). Our study showed the key importance of the dermal route on the risk for the bathers and it is very important to highlight due to the practice to evaluate in Argentina the quality of recreational waters using the concentration limits of the contaminant only for intake (Peluso *et al*. 2011).

Moreover, the key role of certain parameters of exposure must be highlighted (frequency of the exposure, event duration, and bath pattern) in the non-cancer and cancer R from our study. The questionnaire conducted to adjust these variables to reality and to put them in a probabilistic approach is a step forward to avoid falling into deterministic approaches of health risk assessment that could promote risk overestimation.

It is a common practice in risk assessment to consider a hypothetical maximally exposed individual as representative of the exposed population and estimate the dose of exposure based on deterministic values, frequently applying conservative approaches. To perform a true population risk assessment, the variability of the population characteristics (bodyweight, skin surface area, exposure duration, lifetime, and intake rate) should be considered because they can vary between different individuals of the exposed population (Zhao and Kaluarachchi 2002).

Our research tried to consider this situation by improving the assessment by applying probabilistic models and using variables probabilistically distributed.

However, considering a bigger spectrum of exposure scenarios is only a partial reduction in the uncertainty because there will always be remaining residual sources of uncertainty that must be pointed out. It must be highlighted that there exists the scarcity of data of concentrations of hazardous substances. Our analysis was based on few samples, and the need is evident to increase water monitoring, especially according to the season of the year (considering the time of pesticides application in the creek basin). A greater amount of data could change the fitted probability distributions affecting the Monte Carlo results.

Many of the cited papers point to sediments as relevant sources of risk (Albering *et al*. 1999; Goldblum *et al*. 2006; Filipsson *et al*. 2009). Sediments were not considered in our study, although they might have been present in the water samples. Further work should be conducted to study the sediments in the basin to verify if they are acting as contaminant reservoirs, though, in several parts of the creek, they are dragged every year for flood control. Fish ingestion, another source of risk pointed out in the literature, is not applicable in this case because eating what has been fished is an infrequent practice in this basin.

Although data in our research were gathered on bathing behavior by an observational study *in situ*, the surveyed population may be small, providing limited data on relevant exposure variables. More intensive surveys during the next summer will be conducted to improve the knowledge about the potentially exposed population.

The health risk assessment is an alternative to analyze water quality, which, unfortunately, is unused by water management agencies in Argentina. The management procedure is to compare the concentrations found in water with risk-acceptable levels for human consumption of water. However, this method has several drawbacks. Some of them are: the regulatory toxicological safe limits are scarce (*e.g.*, endosulfan and endosulfan sulphate do not have limit values); the analysis is based on single substances only for tap water intake pathway of exposure; the regulatory safe limits are always deterministic values, and so on.

As it was stated by Peluso (2011), health risk assessment has operational advantages over the management procedure cited above: it allows conducting a more exhaustive and realistic study of all exposure processes in order to consider several routes of exposure (digestive, respiratory, skin), scenarios (recreational, residential, work, *etc*.), exposed individuals (children, adults), and even applies to several pathways of exposure or substances, as in this article. On the other hand, the use of a probabilistic technique allows the analysis of the uncertainty and/or variability in the model.

Although this approach is a screening analysis, it could be a complementary study of the microbiological characteristics of the water. The health risk assessment applied to recreational waters is a useful substitute management strategy to ensure the protection of the health of the bathers.

CONCLUSIONS

Pesticides (2,4-D, α, β, δ and γ–hexachlorociclohexane, acetochlor, aldrin, cypermethrin, chlorpyrifos, endosulfan, endosulfan sulphate, γ–chlordane, glyphosate, heptachlor) and heavy metals (As, Cu, Cr, Hg, Zn) in waters from the Del Azul creek

do not pose health risk for recreational bathing, not even considering aggregated exposure through accidental intake and skin contact to all substances simultaneously for children bath population.

The aggregated and cumulative non-cancer (NC) health risk score (R) was 0.46 ($R < 1$) on the age subgroup at greatest health risk (10 years old). Cypermethrin and arsenic were the most dangerous non-cancer substances for the bather through dermal and oral intake, respectively. Cypermethrin aggregated risk represented 80% of the all the substances NC risk. This was mainly due to the magnitude of the dose because of the high value of its concentration in the water, whereas the relevance of arsenic was considered because of its toxicity. The dermal pathway of exposure represented 80% of the total of the aggregated exposure for NC risk, mainly caused by cypermethrin. Moreover, while the relative importance of the intake route decreased with the age of exposed people, the dermal route increased.

The aggregated and cumulative cancer (C) R was $1.03E^{-05}$, reaching the assumed safe limit ($1.0E^{-05}$). This condition was only set for the 5-years-old age group. Arsenic was the most hazardous C substance, generating between 80 and 90% of all the substance cancer R, mainly due to its major dose of exposure through the oral intake pathway.

In both types of risk, the key role of certain parameters related to the bathers' behavior must be highlighted: frequency of the exposure and bath event duration. These parameters were major contributors to the variance of the R than the substance concentrations.

Our research revealed that considering the variability of population characteristics was important to estimate the risk, as it has been stated in other studies. On the other hand, not taking into account the dermal route of exposure for organic substances can produce a great underestimation of the risk. This is very important to highlight due to the practice to evaluate in Argentina the quality of recreational waters using the concentration limits of the contaminant only for the intake pathway of exposure, which is applied to a hypothetical representative of the population. The health risk assessment applied to recreational waters is a useful substitute management strategy to ensure the protection of the bathers' health.

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