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Fabio Peluso [a b], Natalia Othax [a c], José Gonzalez Castelain [a] & Sabrina Dubny [a b]

[a] Flatlands Hydrology Institute "Dr. Eduardo Usunoff", Azul, Argentina

[b] Commission of Scientific Research of Buenos Aires Province (CIC), La Plata, Argentina

[c] National Commission for Scientific and Technological Research (CONICET), Ciudad Autónoma de Buenos Aires, Argentina

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Applying Health Risk Analysis to Assess the Chemical Quality of Water for Recreational Bathing: The Case of Tres Arroyos Creek, Buenos Aires, Argentina

Fabio Peluso,[1,2] Natalia Othax,[1,3] José Gonzalez Castelain,[1] and Sabrina Dubny[1,2]
[1]Flatlands Hydrology Institute "Dr. Eduardo Usunoff", Azul, Argentina; [2]Commission of Scientific Research of Buenos Aires Province (CIC), La Plata, Argentina; [3]National Commission for Scientific and Technological Research (CONICET), Ciudad Autónoma de Buenos Aires, Argentina

ABSTRACT

The aim of the study is to assess the probabilistic non-cancer and cancer risks by recreational bathing in Tres Arroyos creeks (southeastern Buenos Aires Province, Argentina). In these waters, hazardous substances (heavy metals, pesticides) have been detected, possibly related to agricultural activities. To assess such risk, USEPA models in aggregated (exposure through accidental oral water intake and dermal contact simultaneously) and cumulative approaches (combined exposure to more than one substance) were applied, performed for bathers of 5, 10, 15, and 20 years old. The results show that chronic bathing in these waters is not harmful at the concentrations and the exposure scenarios considered. Arsenic was the riskiest substance for both non-cancer and cancer effects, affecting mainly the youngest age group, and the accidental water intake during bathing was the most relevant pathway of exposure. On the other hand, the study highlights the key role of the frequency and duration of the bath event. We discuss the results in light of a previous paper of our authorship concluding that the health risk assessment is a valid alternative to analyze recreational water quality, which, unfortunately, is unused by water management agencies in Argentina.

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

INTRODUCTION

Published papers on risk assessment applied to chemicals in waters based on U.S. Environmental Protection Agency (USEPA) models are relatively common, although the most frequent papers are on contaminants in drinking water (Zakharova

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Address correspondence to Fabio Peluso, Instituto de Hidrología de Llanuras "Dr. Eduardo Usunoff," Av. Italia 780, Azul 7300, Argentina. E-mail: fpeluso@faa.unicen.edu.ar

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; Phan *et al.* 2010; Muhammad *et al.* 2010; Legay *et al.* 2011; Muhammad *et al.* 2011; Wang *et al.* 2011). On the other hand, papers on risk assessment based on chemicals in recreational waters are less frequent. However, many can be cited (Hussain *et al.* 1998; Albering *et al.* 1999; Baars 2002; Dor *et al.* 2003; Blando and Cohn 2004; Goldblum *et al.* 2006; Panyakapo *et al.* 2008; Filipsson *et al.* 2009; Lee *et al.* 2009; Kumar and Xagoraraki 2010, Chen *et al.* 2011; Peluso *et al.* 2012). Nevertheless, if we only focus on those in which risk assessment was applied due to the presence of chemicals in outdoor waters, the list would be significantly reduced.

The lack in Argentina of a national regulatory framework with guideline levels for substances potentially toxic in recreational waters makes their bathing aptitude difficult to evaluate (Peluso *et al.* 2012). Frequently, its aptitude is analyzed by comparing the concentrations of the substance with the tolerable chemical levels for drinking water as a surrogate management tool. Due to the absence of specific tools for assessing water quality for recreational bathing, we studied whether the health risk assessment based on the U.S. Environmental Protection Agency (USEPA) models can be used to cover up this management void.

In Tres Arroyos County, in the southeast of Buenos Aires province (Figure 1), three creeks flow through the county's capital city area (Tres Arroyos city, 57,000 inhabitants). In several locations of these creeks at the urban area or in the neighborhoods, children frequently hold recreational bathing activities. These outdoor activities are spontaneous, in locations of the water course that are not formal beaches. As hazardous substances were found in these waters (heavy metals, pesticides), the aim of our study is to evaluate the potential health risk due the chronic exposure to them by children who bathe in these waters. The risks were calculated using probabilistic USEPA models for individual, aggregated (accidental oral intake and dermal contact) and cumulative exposure, for bathers of 5, 10, 15, and 20 years old.

In a recent paper, we estimated the risk for recreational bathing in a creek located in another area of Buenos Aires Province, 200 km distant from the study area, in which pesticides and heavy metals were present because the heavy agricultural activities held in the basin (Peluso *et al.* 2012). This study, in the methodological point of view, is based on that paper. Given the relationship between both studies, we will discuss similarities and differences of the results between them.

METHODOLOGY

Region of Study

The Buenos Aires Province, located in central-eastern Argentina, has the largest population of the country (15.6 million inhabitants; 50% of the population of the country according to the Directorate of Provincial Statistics of Buenos Aires Province (2012)). The agricultural production of this province reaches almost 46% of the entire country (MLA 2012). The most prominent crops are: soybean (5.8 million Ha sowed), wheat (2.3 million Ha), maize (1.2 million Ha), and sunflower (0.9 million

Figure 1. Location of Tres Arroyos County, in the southeast of Buenos Aires province, Argentina.

Ha), according to the 2010/2011 data (Directorate of Provincial Statistics of Buenos Aires Province 2012).

The Tres Arroyos Creeks basin (38°22′46″S - 60°16′38″W at a central point) is located to the south of the Chaco Pampean Region, about 490 km away from the capital city of Argentina. This basin is 3,017 km^2 (Martinez *et al.* 2008), mainly located within Tres Arroyos county, containing three creeks (first, second, and third branches of Tres Arroyos Creek) that, as mentioned above, flows through the city of Tres Arroyos (Figure 2). Downstream from the city, these streams meet, forming a single water course (Claromecó Creek) that runs throughout the county in a North-South direction until it is discharged into the Argentino Sea. These water courses are typical shallow streams of pampean plains: narrow (from 5 to 20 m), shallow (from 0.50 to 1.50 m), and slow current (water flow between 0.6 to 1.6 $m^3 s^{-1}$ during low-waters, according to Martinez *et al.* 2008).

The Tres Arroyos basin had an average annual temperature and precipitation of 15°C and 680 mm, respectively, for the period 2008–2010; the average temperature and precipitation for the summers of this period were 22°C and 85 mm (INTA

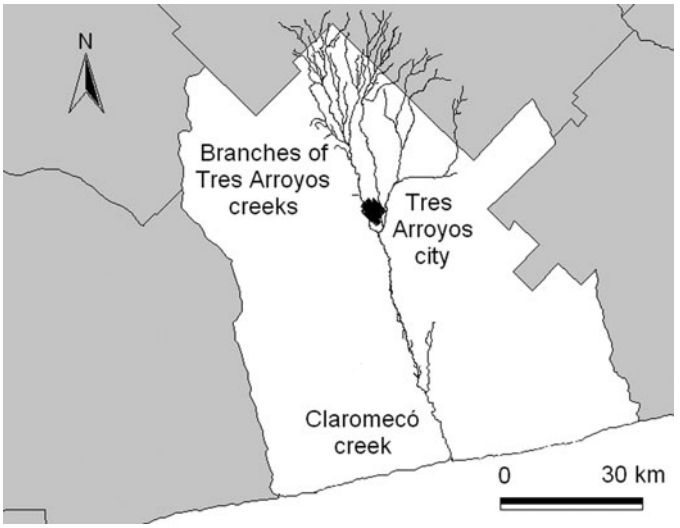

Figure 2. Tres Arroyos County showing the studied creeks.

2012). This basin is mainly an agricultural area in which wheat, soy, sunflower, and corn are the most important crops, with 142,300, 112,700, 61,600, and 9,100 Ha sowed, respectively (Directorate of Provincial Statistics of Buenos Aires Province 2012). Moreover, near the city and downstream, the third branch flows beside a rather small industrial park of agri-foods industries. Due to the potential runoff of pollutants from farm soils or from the industrial effluents adjacent to the streams, the water quality of the mentioned creeks is monitored periodically by sampling of pesticides and heavy metals (Peluso *et al.* 2011).

The Tres Arroyos basin is located in an area with naturally contaminated groundwater with As and F (Paoloni *et al.* 2002; Del Blanco *et al.* 2006; Varni *et al.* 2006; Nicolli *et al.* 2012), with concentrations ranging from 0.009 to 0.5 mg L^{-1} of As, with an arithmetic mean equal to 0.07 and a standard deviation of 0.06 mg L^{-1} (Varni *et al.* 2006), exceeding 7-fold the safe limit for drinking water established by the World Health Organization (WHO) of 0.01 mg L^{-1} (WHO 2008). The groundwater feeding of the creeks and shallow ponds of the basin would therefore be the cause of the high concentration of As, as well as of F (Varni *et al.* 2006) in the creeks presented in this article.

Health Risk Analysis Model

The health risk based on the USEPA model is an estimate of the likelihood that a chemical will generate cancer or non-cancer effects in exposed people (USEPA 1989). The exposure to a hazardous chemical in water during recreational bathing may be produced by two principal pathways: accidental water ingestion and skin contact. For both cases, USEPA models (Eqs. (1) and (2), USEPA 1992a, 2004) were used.

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

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

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 dermal 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.

The non-cancer risk (NC R) was calculated using the risk quotient, which 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). If the risk scores (R) are less than 1.0, the non-cancer health risk is assumed to be negligible (USEPA 1989).

Cancer risk (CR) was also estimated based on the ADD of each route of exposure. CR (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 is assumed 70 years, which is the statistical lifetime duration. The safe criterion for CR was set as 1.0 E^{-05}, according to Peluso *et al.* (2012).

The aggregated (risk caused by the simultaneous exposure through different routes of contact to one or more hazardous substances) and cumulative risks (caused by the simultaneous exposure to several hazardous substances) were estimated using

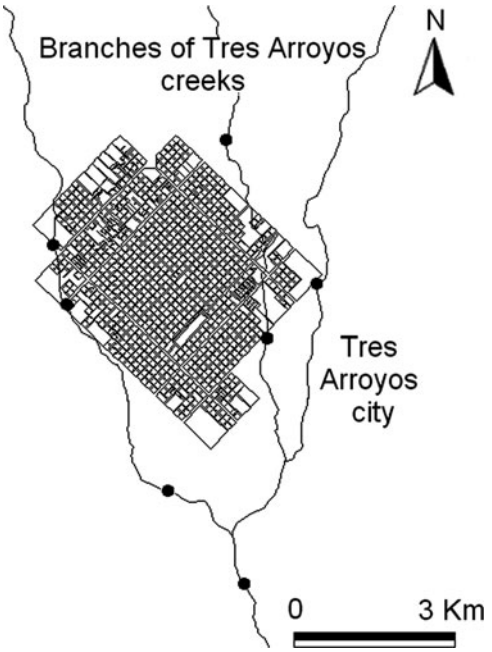

Figure 3. Zoom of the study area at urban zone, showing the three branches of Tres Arroyos Creeks. Dots indicate some of the water sampling stations.

an additive model forming a Risk Index, used by USEPA for screening health risk assessment (USEPA 1989, 2007) and justified in cases when data on toxicology of the mixture, or similar, are unavailable (Mumtaz *et al.* 2007). The same safe criteria for aggregated and cumulative risks as those for individual substances by individual routes of exposure were assumed.

Parameters of the Model

Concentration of the hazardous substances in water (conc)

The study was based on 90 water samples from 10 sites at the three branches of Tres Arroyos and Claromecó Creeks (Figures 2 and 3) between June of 2008 and December of 2010 (quarterly sampling, conducted in dry periods, without rain in the last 5 days). These sites were chosen for being the most frequently used for bathing activities. Water samples for metals and pesticides determination were collected in high-density polyethylene and amber glass bottles with internal teflon tops, respectively. Samples were obtained in the middle of the watercourse at a subsurface level (30 cm below surface). The samples contained some sediments resuspended by the sample staff's footsteps during the sample collection; therefore the water is highly representative of the water that may be in contact with the bathers. The samples were refrigerated (4°C) until analysis and the analyses were made on the samples after shaking them.

The substances are summarized in Table 1. To measure the concentrations of heavy metals (Al, Cu, and Zn) and As, a Varian Spectraa55 Absorption Atomic Spectrophotometer was used with GTA110 Graphite Furnace and VGA-77 Cold Vapor Generation, according to USEPA methods SW 846 M 3020 A – M 7010 and M 7061 EAA, respectively.

For isolation and concentration of organic compounds in aqueous samples, we used the procedures indicated in USEPA method SW 846 M 3510 C Separatory Funnel Liquid-Liquid Extraction. The organochlorine pesticides (α-Hexachlorocyclohexane –HCH–, γ-HCH, δ-HCH, aldrin, γ-chlordane, dichloro diphenyldichloroethane –DDD–, dieldrin, endosulfan, endosulfan sulphate, heptachlor), and cypermethrin 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 toluene concentrations were measured with a Hewlett-Packard-Agilent 6890A Plus+ gas chromatograph–flame ionization detector, according to USEPA method SW 846 M 8015 B/C/D CG-FID.

Probability distributions based on all the concentration sampled of each substance were used as input for the concentration term of the exposure model calculation. In other words, the concentration term is a synthetic integration of the 10 sites monitored. The model was fitted with Crystal Ball 7.1 (Decisioneering 2007a); 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, pointing to the set of parameters for the distribution that best describes the characteristics of the data. The best-fitted probability distribution model and descriptive

Table 1. Probability distribution model parameters and descriptive statistics of the concentrations and percentage of the concentrations below the detection limit of the hazardous substances in the water samples of Tres Arroyos and Claromecó Creeks (in mg L^{-1}).

			Parameters						
Substances	CAS[a]	P. Curve	Min[b]	Max[c]	AM[d]	SD[e]	Others	UCL[f]	Perc. below DL
Aluminium	7429-90-5	Lognormal	$2.50\,E^{-02}$	$4.52\,E^{+00}$	$9.09\,E^{-01}$	$8.43\,E^{-01}$	Loc[g] $2.00\,E^{-02}$	$9.57\,E^{-01}$	4.45
Arsenic	7440-38-2	Logistic	$3.00\,E^{-03}$	$1.73\,E^{-01}$	$7.28\,E^{-02}$	$3.00\,E^{-02}$	Sc[h] $2.00\,E^{-02}$	$6.20\,E^{-02}$	3.33
Copper	7440-50-8	Lognormal	$5.00\,E^{-03}$	$6.37\,E^{+00}$	$8.67\,E^{-01}$	$1.21\,E^{+00}$	Loc $1.50\,E^{-03}$	$7.37\,E^{-01}$	21.73
Zinc	7440-66-6	Lognormal	$7.00\,E^{-03}$	$9.84\,E^{-02}$	$2.45\,E^{-02}$	$1.45\,E^{-02}$	Loc $1.50\,E^{-02}$	$2.63\,E^{-02}$	15.22
α - HCH[i]	319-84-6	Max. Extreme[j]	$6.00\,E^{-07}$	$2.78\,E^{-05}$	$8.06\,E^{-06}$	$4.83\,E^{-06}$	Like[k] $5.83\,E^{-06}$ Sc $4.00\,E^{-06}$	$8.16\,E^{-06}$	36.95
δ - HCH	319-86-8	Min. Extreme[l]	$4.00\,E^{-08}$	$6.23\,E^{-07}$	$2.15\,E^{-07}$	$1.26\,E^{-07}$	Like $2.04\,E^{-07}$ Sc $1.90\,E^{-07}$	$1.20\,E^{-07}$	70.43
γ - HCH	58-89-9	Pareto	$5.00\,E^{-07}$	$2.79\,E^{-06}$	$1.29\,E^{-06}$	$4.12\,E^{-07}$	Loc $8.91\,E^{-07}$ Sh[m] 2.50	$1.31\,E^{-06}$	60.86
Aldrin	309-00-2	Lognormal	$2.00\,E^{-07}$	$4.94\,E^{-06}$	$1.55\,E^{-06}$	$7.38\,E^{-07}$	Loc $1.66\,E^{-07}$	$1.32\,E^{-06}$	65.22
DDD[n]	72-54-8	Min. Extreme	$1.00\,E^{-07}$	$4.52\,E^{-07}$	$1.72\,E^{-07}$	$9.12\,E^{-08}$	Like $1.86\,E^{-07}$ Sc $1.19\,E^{-07}$	$1.37\,E^{-07}$	72.60
Dieldrin	60-57-1	Beta	$1.00\,E^{-06}$	$1.60\,E^{-06}$	$9.96\,E^{-07}$	$3.56\,E^{-07}$	Like $1.10\,E^{-06}$	$1.09\,E^{-06}$	71.73
Endosulfan	115-29-7	Min. Extreme	$1.00\,E^{-07}$	$1.47\,E^{-06}$	$4.85\,E^{-07}$	$2.81\,E^{-07}$	Like $4.40\,E^{-07}$ Sc $4.42\,E^{-07}$	$2.58\,E^{-07}$	70.43
Endosulfan Sulphate	1031-07-8	Min. Extreme	$2.50\,E^{-06}$	$1.16\,E^{-05}$	$5.84\,E^{-06}$	$2.50\,E^{-06}$	Like $6.66\,E^{-06}$ Sc $2.62\,E^{-06}$	$4.80\,E^{-06}$	69.57
γ - Chlordane	5566-34-7	Min. Extreme	$4.00\,E^{-07}$	$2.55\,E^{-06}$	$9.74\,E^{-07}$	$5.41\,E^{-07}$	Like $9.71\,E^{-07}$ Sc $7.72\,E^{-07}$	$6.53\,E^{-07}$	66.09
Heptachlor	76-44-8	Min. Extreme	$1.17\,E^{-05}$	$2.81\,E^{-05}$	$1.37\,E^{-05}$	$5.46\,E^{-06}$	Like $1.57\,E^{-05}$ Sc $5.43\,E^{-06}$	$1.35\,E^{-05}$	63.08
Cypermethrin	97955-44-7	Min. Extreme	$3.00\,E^{-03}$	$6.94\,E^{-02}$	$2.75\,E^{-02}$	$1.50\,E^{-02}$	Like $2.78\,E^{-02}$ Sc $2.10\,E^{-02}$	$1.53\,E^{-02}$	25.30
Toluene	108-88-3	Min. Extreme	$1.00\,E^{-02}$	$8.15\,E^{-03}$	$8.09\,E^{-03}$	$3.14\,E^{-05}$	Like $8.11\,E^{-03}$ Sc $2.47\,E^{-05}$	$8.10\,E^{-03}$	43.47

[a]CAS number, [b]minimum value of concentration of the fitted distribution, [c]maximum value, [d]arithmetic mean, [e]standard deviation, [f]UCL value, [g]location, [h]scale, [i]hexachlorocyclohexane, [j]maximum extreme, [k]likeliest, [l]minimum extreme, [m]shape, [n]dichlorodiphenyldichloroethane.

parameters of each substance are presented in Table 1. The non-detect concentrations of each substance in the water samples were replaced by the 95% upper confidence limit (95% UCL) of the arithmetic mean of the group of the detected concentrations. The 95% 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). The probability distribution of each substance was truncated at the limit of detection and at the maximum concentration sampled, as left and right tails, respectively.

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

The exposed individuals were grouped into age categories of 5, 10, 15, and 20 years old, according to Peluso *et al.* (2012). The assumed intake rate was 0.05 L per hour of the bath event duration 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 in the calculation of the exposure model were treated probabilistically.

The duration of the bath event and the annual frequency of bath days by age were probabilistically established based on a questionnaire conducted at the Del Azul bath resort during the summer of 2010–2011, as it was applied in Peluso *et al.* (2012). The best fitted probability distribution model of these parameters by age was estimated with Crystal Ball 7.1 (Decisioneering 2007b) and results are presented in Table 2.

Although the survey was performed in another town, the distance (200 km) is short, meaning that summer climatic conditions relevant for bathing activities (temperature, wind, rain, *etc.*) are not so different from those at the Del Azul bath resort. The maximum number of bath days (days with temperatures more than 27°C and without rains in the whole day, as was established in Peluso *et al.* 2012), during summers of 2008–2009, 2009–2010, and 2010–2011 were 64, 46, and 57 days of these characteristics (based on INTA 2012), not so different to the maximum number of bath days elected (54) based on Peluso *et al.* (2012).

On the other hand, although there are no formal bath resorts at the Tres Arroyos and Claromecó Creeks, the demographic and socioeconomic characteristics of the population of Tres Arroyos city are not so different from those of Azul city.

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)

In Table 2 are presented the probability distribution model and its descriptive parameters for the BW for the four age groups, based on Lejarraga and Orfila (1987). The body skin area was calculated applying the DuBois and DuBois (1916)

Table 2. Probability distribution models and descriptive statistics of the parameters of the Dose of Exposure by age group: duration of the bath event, frequency of bathing days, bodyweight, body surface, bath pattern and exposed skin area, based on Peluso *et al.* (2012).

Age group	Param.	Fitted distributions	Min.	Max.	AM	SD	95P[a]	Others	
5	t_{event}[b]	Logistic	0.00	3.00 E^{+02}	1.34 E^{+02}	74.10	2.62 E^{+02}	Sc 61.95	
	EF[c]	Gamma	0.00	54.00	15.58	14.48	45.75	Loc –0.11	Sc. 43.22
	BW[d]	Normal	13.50	25.50	19.50	2.50	19.54		
	BSA[e]	Normal	7.62 E^{+03}	8.07 E^{+03}	7.62 E^{+03}	1.68 E^{+02}	7.89 E^{+03}		
	BP[f]	Triangular	0.07	1.00	0.49	0.19	0.83	Mo[g] 0.41	
	ESA[g]	Beta	6.27 E^{+02}	7.69 E^{+03}	3.72 E^{+03}	1.44 E^{+03}	6.39 E^{+03}	α 2.27	β 3.49
10	t_{event}	Min. Ext.	0.00	3.00 E^{+02}	1.88 E^{+02}	25.38	2.78 E^{+02}	Like 2.18 E^{+02}	Sc 62.24
	EF	Gamma	0.00	54.00	19.91	14.34	47.59	Loc. –0.11	Sc. 34.58
	BW	Normal	23.50	44.50	33.56	5.00	41.72		
	BSA	Normal	9.18 E^{+03}	1.33 E^{+04}	1.13 E^{+04}	7.26 E^{+02}	1.24 E^{+04}		
	BP	Triangular	0.07	1.00	0.49	0.19	0.83	Mo 0.40	
	ESA	Beta	8.38 E^{+02}	1.20 E^{+04}	5.53 E^{+03}	2.22 E^{+03}	9.44 E^{+04}	α2.36	β3.95
15	t_{event}	Gamma	0.00	3.00 E^{+02}	1.50 E^{+02}	23.87	2.74 E^{+02}	Loc 25.52	Sc 4.37
	EF	Gamma	0.00	54.00	17.94	14.83	47.11	Loc. –0.11	Sc. 41.31
	BW	Normal	34.00	74.00	50.30	7.33	62.10		
	BSA	Normal	1.23 E^{+04}	1.81 E^{+04}	1.50 E^{+04}	9.59 E^{+02}	1.66 E^{+04}		
	BP	Triangular	0.07	1.00	0.50	0.19	0.84	Mo 0.44	
	ESA	Beta	1.14 E^{+03}	1.61 E^{+04}	7.50 E^{+03}	2.87 E^{+03}	1.27 E^{+04}	α 2.94	β 4.24
20	t_{event}	Beta	0.00	2.40 E^{+02}	46.63	49.78	1.52 E^{+02}	α 0.42	β 3.25
	EF	Beta	0.00	54.00	11.59	13.26	41.93	Loc –0.11	Sc 57.72
	BW	Normal	50.00	85.00	65.50	7.60	78.47		
	BSA	Normal	1.52 E^{+04}	2.07 E^{+04}	1.78 E^{+04}	9.84 E^{+02}	1.95 E^{+04}		
	BP	Triangular	0.07	1.00	0.51	0.19	0.85	Mo 0.45	
	ESA	Beta	1.30 E^{+03}	1.91 E^{+04}	9.03 E^{+03}	3.45 E^{+03}	1.49 E^{+04}	α 2.94	β 4.17

[a]95th percentile, [b]duration of the bath event (minutes), [c]annual frequency of bathing days (days), [d]bodyweight (kg), [e]body surface area (cm^2), [f]bath pattern (dimensionless), [g]mode, [h]exposed skin area (cm^2).

model (Eq. (3)).

$$BSA = H^{0.725} \times BW^{0.425} \times 0.007184 \quad (3)$$

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

During the bath event, the full-body is not necessarily involved all the time. On the contrary, the body will be only occasionally totally submerged. For that, if we only assume that the bather is in full contact with the water, BSA (Eq. (3)) is equal to ESA (Eq. (2)). However, it is not accurate in reality. To calculate the exposed body surface area for its use in the dermal dose estimation, a correction factor called Bath Pattern (BP) was applied, as can be viewed in Eq. (4). This estimates what percentage of the skin surface of a bather's body was in contact with the water during the bath event.

$$ESA = BSA \times BP \quad (4)$$

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

Based on direct observation of bathers' behavior in the Del Azul bath resort, only during brief moments were the bathers fully submerged in the water. BP was calculated by age group as the percentage of the underwater body parts during the bath event in respect to the total surface area of the whole body. To calculate these percentage, a triangular probability distribution by age group was assumed, in which the minimum value corresponds to the percentage of the feet only in respect to the entire body surface area, the mode to the feet + legs + hands surfaces, and the maximum when the entire body was totally submerged. BP was calculated based on USEPA (2004). The parameters of the model used for ESA, BSA, and BP calculations are given in Table 2.

Dermal Dose Absorbed

The absorbed dermal dose per event (DA_{event}) was estimated based on Peluso *et al.* (2012), using the steady-state approach developed by USEPA (2004). For inorganic substances, DA_{event} was calculated by using Eq. (5).

$$DA_{event} = t_{event} \times Conc \times K_p \quad (5)$$

where t_{event} = e duration (hr $event^{-1}$), Conc = concentration of the hazardous substance in water (mg L^{-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).

$$\text{Log } K_p = -2.80 + 0.66 \log Kow - 0.0061\ Mw \quad (6)$$

For organic substances, DA_{event} was estimated based on Eq. (7) or on Eq. (8). If the duration of event (t_{event}) was less than the time to reach steady state for 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).

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

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

where DA_{event} = absorbed dermal 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 $event^{-1}$).

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 this is calculated by using Eq. (9) (USEPA 2004).

$$B = k_p\ (MW^{0.5}/2.6) \quad (9)$$

The lag time per event was 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).

$$\tau = l_{sc}^2/6 \times D_{sc} \quad (10)$$

where l_{sc} = apparent thickness of stratum corneum (cm), assumed 1.0 E^{-03} cm, D_{sc} = Effective diffusion coefficient for chemical transfer through the stratum corneum (cm^2 hr^{-1}), estimated based on Eq. (11) (USEPA 2004).

$$Dsc = l_{sc} \times\ 10^{(-2.8-0.0056\ Mw)} \quad (11)$$

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 dermal dose per event and other terms of Eqs. (5) through (11) are presented in Table 3.

Non-Cancer and Cancer Risk Calculation for Single, Aggregated, and Cumulative Exposure

The NC and CR 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 (Al, As [included although it is a metalloid], Cu, Zn) and Chlorinated Pesticides (α, δ and γ–hexachlorociclohexane (HCH), aldrin, γ-chlordane, dichlorodiphenyldichloroethane –DDD–, endosulfan, endosulfan sulphate and 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 by use of 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 every 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

Table 3. Parameters used for the estimation of the Absorbed Dermal Dose per Event, and toxicological reference values of the hazardous substances for accidental water intake and dermal contact exposure routes.

								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]
Aluminium	N.A.	N.A.	$1.00\,E^{-03}$	N.A.	N.A.	N.A.	N.A.	$6.82\,E^{-06}$	$9.21\,E^{-06}$	$7.71\,E^{-06}$	$2.99\,E^{-06}$	$1.00\,E^{+00}$	$1.00\,E^{+00}$	N.A.	N.A.	$1.00\,E^{+00}$
Arsenic	N.A.	N.A.	$1.00\,E^{-03}$	N.A.	N.A.	N.A.	N.A.	$3.84\,E^{-07}$	$4.57\,E^{-07}$	$4.19\,E^{-07}$	$2.02\,E^{-07}$	$3.00\,E^{-04}$	$3.00\,E^{-04}$	$1.50\,E^{+00}$	$1.50\,E^{+00}$	$1.00\,E^{+00}$
Copper	N.A.	N.A.	$1.00\,E^{-03}$	N.A.	N.A.	N.A.	N.A.	$8.74\,E^{-06}$	$1.14\,E^{-05}$	$9.87\,E^{-06}$	$3.05\,E^{-06}$	$4.00\,E^{-02}$	$4.00\,E^{-02}$	N.A.	N.A.	$1.00\,E^{+00}$
Zinc	N.A.	N.A.	$6.00\,E^{-04}$	N.A.	N.A.	N.A.	N.A.	$8.63\,E^{-08}$	$1.13\,E^{-07}$	$9.61\,E^{-08}$	$4.11\,E^{-08}$	$3.00\,E^{-01}$	$3.00\,E^{-01}$	N.A.	N.A.	$1.00\,E^{+00}$
α - HCH	$2.91\,E^{+02}$	4.26	$2.48\,E^{-02}$	$1.62\,E^{-01}$	4.56	10.94	0.9	$3.55\,E^{-09}$	$4.14\,E^{-09}$	$3.75\,E^{-09}$	$2.25\,E^{-09}$	$3.00\,E^{-04}$	$3.00\,E^{-04}$	$6.30\,E^{+00}$	$6.30\,E^{+00}$	$1.00\,E^{+00}$
δ - HCH	$2.91\,E^{+02}$	4.26	$2.48\,E^{-02}$	$1.62\,E^{-01}$	4.56	10.94	0.9	$9.23\,E^{-11}$	$1.04\,E^{-10}$	$9.70\,E^{-11}$	$6.03\,E^{-11}$	$3.00\,E^{-04}$	$3.00\,E^{-04}$	$1.30\,E^{+00}$	$1.30\,E^{+00}$	$1.00\,E^{+00}$
γ - HCH	$2.91\,E^{+02}$	4.26	$2.48\,E^{-02}$	$1.62\,E^{-01}$	4.56	10.94	0.9	$4.69\,E^{-10}$	$5.26\,E^{-10}$	$4.86\,E^{-10}$	$2.92\,E^{-10}$	$3.00\,E^{-04}$	$3.00\,E^{-04}$	$1.30\,E^{+00}$	$1.30\,E^{+00}$	$1.00\,E^{+00}$
Aldrin	$3.65\,E^{+02}$	3.01	$1.40\,E^{-03}$	$1.03\,E^{-02}$	11.88	28.51	1	$6.33\,E^{-11}$	$7.13\,E^{-11}$	$6.47\,E^{-11}$	$4.00\,E^{-11}$	$3.00\,E^{-05}$	$3.00\,E^{-05}$	$1.70\,E^{+01}$	$1.70\,E^{+01}$	$1.00\,E^{+00}$
DDD	$3.20\,E^{+02}$	5.8	$1.79\,E^{-01}$	$1.23\,E^{+00}$	6.65	26.00	0.8	$5.48\,E^{-10}$	$6.14\,E^{-10}$	$5.71\,E^{-10}$	$3.54\,E^{-10}$	$2.00\,E^{-03}$	$2.00\,E^{-03}$	$2.40\,E^{-01}$	$2.40\,E^{-01}$	$1.00\,E^{+00}$
Dieldrin	$3.81\,E^{+02}$	4.56	$1.22\,E^{-02}$	$9.19\,E^{-02}$	14.61	35.05	0.8	$2.73\,E^{-10}$	$3.01\,E^{-10}$	$2.84\,E^{-10}$	$1.87\,E^{-10}$	$5.00\,E^{-05}$	$5.00\,E^{-05}$	$1.60\,E^{+01}$	$1.60\,E^{+01}$	$1.00\,E^{+00}$
Endosulfan	$4.07\,E^{+02}$	3.5	$1.73\,E^{-03}$	$1.34\,E^{-02}$	20.44	49.05	0.9	$3.06\,E^{-11}$	$3.49\,E^{-11}$	$3.19\,E^{-11}$	$2.00\,E^{-11}$	$6.00\,E^{-03}$	$6.00\,E^{-03}$	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	0.9	$3.56\,E^{-10}$	$3.99\,E^{-10}$	$3.67\,E^{-10}$	$2.45\,E^{-10}$	$6.00\,E^{-03}$	$6.00\,E^{-03}$	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	0.7	$1.04\,E^{-09}$	$1.18\,E^{-09}$	$1.09\,E^{-09}$	$6.84\,E^{-10}$	$5.00\,E^{-04}$	$4.00\,E^{-04}$	$3.50\,E^{-01}$	$4.38\,E^{-01}$	$8.00\,E^{-01}$
Heptachlor	$3.73\,E^{+02}$	5.86	$9.87\,E^{-02}$	$7.34\,E^{-01}$	13.24	51.19	0.8	$2.94\,E^{-08}$	$3.27\,E^{-08}$	$3.08\,E^{-08}$	$2.02\,E^{-08}$	$5.00\,E^{-04}$	$5.00\,E^{-04}$	$4.50\,E^{+00}$	$4.50\,E^{+00}$	$1.00\,E^{+00}$
Cypermethrin	$4.16\,E^{+02}$	6.38	$1.26\,E^{-01}$	$9.85\,E^{-01}$	23.08	88.98	0.6	$8.59\,E^{-05}$	$9.69\,E^{-05}$	$8.93\,E^{-05}$	$5.72\,E^{-05}$	$1.00\,E^{-02}$	$1.00\,E^{-02}$	N.A.	N.A.	$1.00\,E^{+00}$
Toluene	$9.21\,E^{+01}$	2.73	$3.11\,E^{-02}$	$1.15\,E^{-01}$	0.35	0.84	1	$1.18\,E^{-06}$	$1.25\,E^{-06}$	$1.22\,E^{-06}$	$7.77\,E^{-07}$	$8.00\,E^{-02}$	$8.00\,E^{-02}$	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 (hr), [g]fraction absorbed (dimensionless), based on USEPA (2004), [h]95th percentile of the absorbed dermal dose per event (mg cm^{-2} $event^{-1}$) by age group, [i]reference dose (mg kg^{-1} d^{-1}), [j]slope factor (kg d mg ^{1}), [k]contaminant fraction absorbed in gastrointestinal tract (dimensionless), based on USEPA (2004), [l]oral intake, [m]dermal. N.A.: not applicable.

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 a high-end risk indicator (semi-conservative approach).

The RfD and SF used for accidental water intake risk calculations were obtained from USEPA's IRIS database (2010b). The RfD and SD for dermal risk calculations were estimated based on USEPA (2004), following Eq. (12) and (13).

$$RfD_{derm} = RfD_{in} \times ABS_{GI} \quad (12)$$

$$SD_{derm} = SD_{in} / ABS_{GI} \quad (13)$$

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.

In Table 3 are presented the toxicological reference values of the substances by oral intake and skin contact, for both NC and C effects, respectively, as well as ABS_{GI} values.

Statistical Analysis

All simple mathematical calculations such as means, standard deviations, 95th percentiles (other than in MC calculations), 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 arithmetic means 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 the exposure parameters to the risk results, a sensitivity analysis was performed with Crystal Ball 7.1. (Decisioneering 2007a). This estimates sensitivity based on the rank correlation coefficients between every parameter of the model and the results while the simulation is running (Decisioneering 2007b).

RESULTS

Non-Cancer Aggregated Risk by Age

The R from the aggregated exposure for NC substances (All the Substances group for accidental water intake + dermal contact) of Tres Arroyos Creeks, never reached the acceptability criteria of 1.0 (R between 0.38 to 0.062 according to the age group, following the sequence 5 YO > 10 YO > 15 YO > 20 YO), as shown in Table 4. The Kruskal-Wallis test on the All the Substances cumulative and aggregated NC R of the Monte Carlo trials showed a significant difference between the four age groups ($p < .05$).

Table 4. Aggregated non-cancer and cancer risks by individual and by groups of substances.

	NC				C			
Substances	5 YO[a]	10 YO	15 YO	20 YO	5 YO	10 YO	15 YO	20 YO
Aluminium	$1.04\,E^{-03}$	$9.62\,E^{-04}$	$5.15\,E^{-04}$	$5.79\,E^{-05}$	—	—	—	—
Arsenic	$2.49\,E^{-01}$	$2.11\,E^{-01}$	$1.24\,E^{-01}$	$1.65\,E^{-02}$	$2.59\,E^{-05}$	$2.32\,E^{-05}$	$1.32\,E^{-05}$	$2.77\,E^{-06}$
Copper	$2.52\,E^{-02}$	$2.58\,E^{-02}$	$1.51\,E^{-02}$	$1.40\,E^{-03}$	—	—	—	—
Zinc	$8.34\,E^{-05}$	$7.05\,E^{-05}$	$4.07\,E^{-05}$	$4.97\,E^{-06}$	—	—	—	—
Metals	$2.72\,E^{-01}$	$2.28\,E^{-01}$	$1.35\,E^{-01}$	$1.80\,E^{-02}$	$2.59\,E^{-05}$	$2.32\,E^{-05}$	$1.32\,E^{-05}$	$2.77\,E^{-06}$
α - HCH	$1.77\,E^{-04}$	$1.97\,E^{-04}$	$1.68\,E^{-04}$	$5.66\,E^{-05}$	$8.05\,E^{-08}$	$8.68\,E^{-08}$	$7.18\,E^{-08}$	$2.42\,E^{-08}$
δ - HCH	$4.95\,E^{-06}$	$5.67\,E^{-06}$	$4.39\,E^{-06}$	$1.49\,E^{-06}$	$4.42\,E^{-10}$	$5.03\,E^{-10}$	$3.95\,E^{-10}$	$1.30\,E^{-10}$
γ - HCH	$2.72\,E^{-05}$	$2.90\,E^{-05}$	$2.33\,E^{-05}$	$8.67\,E^{-06}$	$2.57\,E^{-09}$	$2.72\,E^{-09}$	$2.14\,E^{-09}$	$7.78\,E^{-10}$
Aldrin	$7.50\,E^{-05}$	$7.00\,E^{-05}$	$4.99\,E^{-05}$	$1.36\,E^{-05}$	$8.94\,E^{-09}$	$8.64\,E^{-09}$	$5.90\,E^{-09}$	$1.82\,E^{-09}$
γ - Chlordane	$4.16\,E^{-05}$	$4.43\,E^{-05}$	$3.57\,E^{-05}$	$1.28\,E^{-05}$	$1.68\,E^{-09}$	$1.80\,E^{-09}$	$1.48\,E^{-09}$	$4.99\,E^{-10}$
DDD	$4.37\,E^{-06}$	$4.56\,E^{-06}$	$3.91\,E^{-06}$	$1.45\,E^{-06}$	$4.76\,E^{-10}$	$5.01\,E^{-10}$	$4.45\,E^{-10}$	$1.54\,E^{-10}$
Dieldrin	$1.04\,E^{-04}$	$1.10\,E^{-04}$	$8.93\,E^{-05}$	$3.21\,E^{-05}$	$1.96\,E^{-08}$	$2.08\,E^{-08}$	$1.66\,E^{-08}$	$5.70\,E^{-09}$
Endosulfan	$1.48\,E^{-07}$	$1.48\,E^{-07}$	$1.02\,E^{-07}$	$2.89\,E^{-08}$	—	—	—	—
Endosulfan Sulphate	$1.87\,E^{-06}$	$1.78\,E^{-06}$	$1.27\,E^{-06}$	$3.75\,E^{-07}$	—	—	—	—
Heptachlor	$1.00\,E^{-03}$	$1.10\,E^{-03}$	$8.66\,E^{-04}$	$3.55\,E^{-04}$	$5.26\,E^{-07}$	$5.89\,E^{-07}$	$4.53\,E^{-07}$	$1.75\,E^{-07}$
Chlorinated Pesticides	$1.38\,E^{-03}$	$1.48\,E^{-03}$	$1.19\,E^{-03}$	$4.70\,E^{-04}$	$6.38\,E^{-07}$	$7.02\,E^{-07}$	$5.44\,E^{-07}$	$2.14\,E^{-07}$
Cypermethrin	$1.28\,E^{-01}$	$1.48\,E^{-01}$	$1.18\,E^{-01}$	$4.40\,E^{-02}$	—	—	—	—
Toluene	$3.35\,E^{-04}$	$3.48\,E^{-04}$	$2.84\,E^{-04}$	$8.71\,E^{-05}$	—	—	—	—
All the Substances	$3.79\,E^{-01}$	$3.48\,E^{-01}$	$2.33\,E^{-01}$	$6.19\,E^{-02}$	$2.66\,E^{-05}$	$2.37\,E^{-05}$	$1.37\,E^{-05}$	$2.94\,E^{-06}$
Metals/All the Substances	$7.17\,E^{+01}$	$6.56\,E^{+01}$	$5.77\,E^{+01}$	$2.91\,E^{+01}$	$9.74\,E^{+01}$	$9.77\,E^{+01}$	$9.64\,E^{+01}$	$9.41\,E^{+01}$
Chlorinated Pesticides/All the Substances	$3.63\,E^{-01}$	$4.24\,E^{-01}$	$5.09\,E^{-01}$	$7.59\,E^{-01}$	$2.40\,E^{+00}$	$2.96\,E^{+00}$	$3.98\,E^{+00}$	$7.27\,E^{+00}$
Arsenic/All the Substances	$6.58\,E^{+01}$	$6.07\,E^{+01}$	$5.33\,E^{+01}$	$2.67\,E^{+01}$	$9.74\,E^{+01}$	$9.77\,E^{+01}$	$9.64\,E^{+01}$	$9.41\,E^{+01}$
Second Risky Substance/All the Substances	$3.38\,E^{+01}$	$4.25\,E^{+01}$	$5.07\,E^{+01}$	$7.11\,E^{+01}$	$1.98\,E^{+00}$	$2.48\,E^{+00}$	$3.32\,E^{+00}$	$5.95\,E^{+00}$
Arsenic/Heavy Metals	$9.18\,E^{+01}$	$9.26\,E^{+01}$	$9.24\,E^{+01}$	$9.18\,E^{+01}$	$1.00\,E^{+02}$	$1.00\,E^{+02}$	$1.00\,E^{+02}$	$1.00\,E^{+02}$
Arsenic/Second Risky Substance	$1.95\,E^{+02}$	$1.43\,E^{+02}$	$1.05\,E^{+02}$	$3.76\,E^{+01}$	$4.92\,E^{+03}$	$3.93\,E^{+03}$	$2.91\,E^{+03}$	$1.58\,E^{+03}$

Heavy metals was the riskiest substances group for all the considered age categories, explaining between 70 and 60% of the All the Substances NC R of the youngest exposed people, following a 5 YO > 10 YO > 15 YO age sequence. The R from Chlorinated Pesticides was less than 1% of the R from All the Substances in all the age groups.

Analyzing the NC R of the different substances, the riskiest substance for 5, 10, and 15 YO age groups was As (included in Heavy metal group), that contributed more than 50% for the All the Substances aggregated risk for these age groups, and almost 90% of the Metals group non-cancer R for all the age categories. The second riskiest substance was cypermethrin (not a Chlorinated Pesticides), explaining from 30 to 70% of the NC R, according to the age classes (5 YO < 10 YO < 15 YO < 20 YO). For the 20 YO age category, cypermethrin was riskier than As. The As/cypermethrin relationship decreased according to the age group, from 2 (5 YO) to 0.40 times (20 YO) in the R of As in relation with the R of cypermethrin. The Kruskal-Wallis test on As and cypermethrin aggregated NC R of the Monte Carlo trials evidenced a significant difference between the four age groups ($p < .05$).

Cancer Aggregated Risk by Age

The CR scores can be appreciated as shown in Table 4. It was observed that All the Substances cumulative cancer R for the three younger age groups of exposed people reached the assumed criterion health concern risk level of 1.0 E^{-05} (5 YO > 10 YO > 15 YO > 20 YO). Arsenic, the only carcinogenic metal of the substances considered, was the riskiest C substance, generating above 90% of the All the Substances CR. The Chlorinated Pesticides CR was much lower, being the remaining of the percentage generated by As since cypermethrin and toluene were NC toxic substances. The riskiest Chlorinated Pesticide was heptachlor (the second riskiest cancer substance), almost two orders of magnitude lesser than the cancer safe risk level, following the R sequence according to age (10 YO > 5 YO > 15 YO > 20 YO). Comparing the R of the As with the R of the heptachlor, the R of the first substance was from 5 (5 YO) to 1.5 (20 YO) times the R of the second one.

Non-Cancer and Cancer Risks by Route of Exposure

In Table 5 are presented the NC and CR scores for each substance and of groups of substances by pathways of exposure, for the age classes with the widest gap: 5 and 20 YO. The results for NC and C were quite similar, though some differences could be spotted.

All the substances group is more toxic by NC effects by intake for the younger exposed age class than for the adults (being 67 to 21% the rate of the NC R by intake over the aggregated NC R), indicating the relative increasing of the dermal route importance according to the age. Metals were toxic mainly due to the exposure by accidental intake (from 90 to 70% of the aggregated NC risk according to 5 and 20 YO age group) while the other category of substances are toxic mainly through the dermal route. The chlorinated pesticides group was remarkably risky by dermal route (with the exception of aldrin, endosulfan, and endosulfan sulphate), also cypermethrin. Cypermethrin, being the second riskiest substance, had an important role in the great decrease in the All the Substances NC R by intake with respect to

Table 5. Non-carcinogenic and carcinogenic risks for 5 and for 20 YO by route of exposure, by individual and by groups of substances indicating the importance of the oral pathway as percent of the aggregated exposure.

	NC						C					
	5 YO			20 YO			5 YO			20 YO		
Substances	Int	Derm	Int/Aggr[a]	Int	Derm	Int/Aggr	Int	Derm	Int/Aggr	Int	Derm	Int/Aggr
Aluminium	$9.74\,E^{-04}$	$7.37\,E^{-05}$	93.30	$4.33\,E^{-05}$	$1.56\,E^{-05}$	74.70	—	—	—	—	—	—
Arsenic	$2.32\,E^{-01}$	$1.79\,E^{-02}$	93.20	$1.19\,E^{-02}$	$4.30\,E^{-03}$	72.11	$2.39\,E^{-05}$	$1.86\,E^{-06}$	92.11	$2.36\,E^{-06}$	$4.27\,E^{-07}$	85.27
Copper	$2.36\,E^{-02}$	$1.80\,E^{-03}$	93.66	$1.02\,E^{-03}$	$3.74\,E^{-04}$	73.04	—	—	—	—	—	—
Zinc	$7.98\,E^{-05}$	$3.84\,E^{-06}$	95.75	$4.07\,E^{-06}$	$8.81\,E^{-07}$	81.99	—	—	—	—	—	—
Metals	$2.51\,E^{-01}$	$1.93\,E^{-02}$	92.45	$1.30\,E^{-02}$	$4.65\,E^{-03}$	72.03	$2.39\,E^{-05}$	$1.86\,E^{-06}$	92.11	$2.36\,E^{-06}$	$4.27\,E^{-07}$	85.27
α - HCH	$2.54\,E^{-05}$	$1.53\,E^{-04}$	14.32	$1.29\,E^{-06}$	$5.55\,E^{-05}$	2.28	$1.15\,E^{-08}$	$6.89\,E^{-08}$	14.30	$1.05\,E^{-09}$	$2.32\,E^{-08}$	4.35
δ - HCH	$6.75\,E^{-07}$	$4.25\,E^{-06}$	13.64	$3.46\,E^{-08}$	$1.46\,E^{-06}$	2.32	$6.35\,E^{-11}$	$3.75\,E^{-10}$	14.34	$5.90\,E^{-12}$	$1.24\,E^{-10}$	4.54
γ - HCH	$3.87\,E^{-06}$	$2.37\,E^{-05}$	14.25	$2.12\,E^{-07}$	$8.45\,E^{-06}$	2.45	$3.57\,E^{-10}$	$2.24\,E^{-09}$	13.88	$3.56\,E^{-11}$	$7.41\,E^{-10}$	4.58
Aldrin	$4.86\,E^{-05}$	$2.96\,E^{-05}$	64.88	$2.58\,E^{-06}$	$1.12\,E^{-05}$	18.96	$5.75\,E^{-09}$	$3.48\,E^{-09}$	64.36	$5.71\,E^{-10}$	$1.23\,E^{-09}$	31.40
γ - Chlordane	$1.99\,E^{-06}$	$3.98\,E^{-05}$	4.79	$9.44\,E^{-08}$	$1.27\,E^{-05}$	0.73	$8.03\,E^{-11}$	$1.60\,E^{-09}$	4.78	$6.92\,E^{-12}$	$4.95\,E^{-10}$	1.39
DDD	$8.78\,E^{-08}$	$4.27\,E^{-06}$	2.01	$4.21\,E^{-09}$	$1.44\,E^{-06}$	0.29	$9.41\,E^{-12}$	$4.66\,E^{-10}$	1.98	$9.19\,E^{-13}$	$1.52\,E^{-10}$	0.60
Dieldrin	$1.87\,E^{-05}$	$8.84\,E^{-05}$	17.91	$9.55\,E^{-07}$	$3.13\,E^{-05}$	2.97	$3.50\,E^{-09}$	$1.64\,E^{-08}$	17.87	$3.32\,E^{-10}$	$5.37\,E^{-09}$	5.83
Endosulfan	$7.80\,E^{-08}$	$7.16\,E^{-08}$	52.52	$3.96\,E^{-09}$	$2.48\,E^{-08}$	13.68	—	—	—	—	—	—
Endosulfan Sulphate	$9.38\,E^{-07}$	$9.60\,E^{-07}$	50.25	$4.69\,E^{-08}$	$3.36\,E^{-07}$	12.51	—	—	—	—	—	—
Heptachlor	$2.63\,E^{-05}$	$9.78\,E^{-04}$	2.62	$1.35\,E^{-06}$	$3.54\,E^{-04}$	0.38	$1.41\,E^{-08}$	$5.12\,E^{-07}$	2.68	$1.36\,E^{-09}$	$1.74\,E^{-07}$	0.77
Chlorinated Pesticides	$1.20\,E^{-04}$	$1.27\,E^{-03}$	8.75	$6.83\,E^{-06}$	$4.65\,E^{-04}$	1.45	$3.39\,E^{-08}$	$6.09\,E^{-07}$	5.32	$3.40\,E^{-09}$	$2.12\,E^{-07}$	1.59
Cypermethrin	$2.76\,E^{-03}$	$1.26\,E^{-01}$	2.15	$1.37\,E^{-04}$	$4.39\,E^{-02}$	0.31	—	—	—	—	—	—
Toluene	$9.03\,E^{-05}$	$2.59\,E^{-04}$	26.99	$4.95\,E^{-06}$	$8.32\,E^{-05}$	5.68	—	—	—	—	—	—
All the Substances	$2.54\,E^{-01}$	$1.42\,E^{-01}$	66.90	$1.31\,E^{-02}$	$4.88\,E^{-02}$	21.21	$2.39\,E^{-05}$	$2.45\,E^{-06}$	89.90	$2.36\,E^{-06}$	$6.28\,E^{-07}$	80.29

[a]Aggregated exposure.

Table 6. Contributions to the variance of the non-cancer and cancer risk scores for 5 and 20 YO of the exposure parameters for aggregated, intake and dermal contact pathways for All the Substances cumulative group.

	NCR						CR					
	5 YO			20 YO			5 YO			20 YO		
All the Substances	Aggr	Int	Derm	Aggr	Int	Derm	Aggr	Int	Derm	Aggr	Int	Derm
EF	77.4	71.2	72.1	73.2	57	73	66.7	65.9	68.5	58.7	54.9	65.2
t_{event}	15.3	20.3	7.2	18.9	38.6	14.4	18.2	18.6	13.7	33.4	36.6	21.3
Cypermethrin Conc.	2.6		14.3	5		8.5						
Arsenic Conc.	3	6.7			3.2		7.0	7.4	3.6	2.9	3.9	2.4
Bath Pattern			5.5	1.9		3.2			5.5			3.9
ED							7.0	6.9	7.6	3.9	3.6	5.5
Other	1.7	1.8	0.9	1	1.2	0.9	1.1	1.2	1.1	1.1	1.0	1.7

dermal route from 5 to 20 YO. This is explained in Table 4, where it is shown that the increase in the importance of the NC risk generated by cypermethrin according to the age, rising to almost 70% of the aggregated and cumulative risk in the adult group.

The CR for All the Substances group by intake for the 5 YO age group was the 90% of the aggregated CR, decreasing only to 80% for the adult group. Taking into account that the CR of the riskiest substance (As, dangerous mainly for ingestion route) was between 5 to 1.5 times the CR of the second risk substance (heptachlor, dangerous mainly by dermal pathway), the rates of the importance of the ingestion route with relation to the dermal one remained at high values (90–80%, as can be viewed in Table 5), unlike the NC R All the substances group case. The tendency of the results of the C substances according to the age is quite similar to the NC substances: in both cases, it occurred because of the relative increasing of the dermal route importance with relation to the intake. However, this increment is less important in CR due the As R dominance, as previously stated.

The results of the sensitivity analysis indicated the importance of each parameter of the exposure estimation in the risk values for each age group. Shown in Table 6 are the percentage of the contribution to the variance of the exposure parameters for aggregated and for exposure pathways of All the Substances cumulative NC and C risk, for the 5 and 20 YO age groups. Two variables related to the time of the bath had the biggest contribution to the variances: the Frequency of the Exposure (*EF*, from 60 to 80% of the total variance for the aggregated risk of both age groups, greater in 5 YO NC R) and the Event Duration (t_{event}, from 15 to 30% of the total variance, not relevant differences between NC or C risk, but greater for 20 YO).

The concentration of the key dangerous substances (As and cypermethrin), even together explained less than 10% of the entire variance. Arsenic presented a slightly greater contribution than cypermethrin, except in 20 YO aggregated NC risk.

Analyzing the results by exposure pathway, the contribution of *EF* was slightly smaller and t_{event} slightly greater by intake than by the dermal route for both types of risk analysis and age groups considered. Arsenic contributed mainly by intake and

cypermethrin by dermal route, but, in this case, only for NC risk. The Bath Pattern contributed only for the dermal route.

DISCUSSION

The NC and C risks for the recreational bathing at Tres Arroyos and Claromecó Creeks were negligible, although the aggregated and cumulative cancer risks exceed the cancer risk criterion. Three arguments support that judgment. First, as it was commented in Peluso *et al.* (2012), several cancer health risk limits that are very different in the degree of conservatism can be found in the literature: between 1.0 E^{-06} (*e.g.*, USEPA) and 1.0 E^{-04} (Baars 2002; Harper and Harris 2008; Phan *et al.* 2010), unlike the non-cancer safe level that is always equal to 1. Second, given that there were not set specific safe criteria for recreational water use, it is frequent to apply the USEPA drinking water limit of 1.0 E^{-06} as surrogate, which is a very conservative approach to be applied on recreational accidental water intake. Third, the USEPA stated about the cumulative risk assessment 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).

The results from this study have shown many issues to highlight, both similarities and differences, with relation to Peluso *et al.* (2012). The present study has shown that the risk scores were different according to the age of the exposed individual as Peluso *et al.* (2012), which is logical because of the similarity between both studies with respect to the exposure parameters values adopted. The 95th percentiles of the NC risk scores of both studies were quite similar: the aggregated and cumulative R of Tres Arroyos water was 3.79 E^{-01} versus 3.63 E^{-01} of the Del Azul Creek. The riskiest NC substance of each study had comparable values (2.49 E^{-01} in this study *vs.* 2.88 E^{-01} in the Del Azul study). However, the substance involved is not the same: in this study, it was As but in the other study, it was cypermethrin. For Del Azul water health risk, cypermethrin generated an important NC risk score through the dermal route (three orders of magnitude higher than As, the second in importance in aggregated NC risk). Due to the role of certain parameters of the dermal exposure (cypermethrin concentration, t_{event} and bath pattern), the 10-years-old group resulted in a higher NC risk than the 5 age group. On the other hand, the order of importance of the As and cypermethrin NC risk values from Del Azul analysis was reversed in the present study. This generated a decrease in the importance of the dermal route on the aggregated NC risk values for the most endangered age group (from 80% in the other paper to 30% in this study). However, the importance of the dermal pathway of exposure increases according to the age, reaching 80% in the adult age group. As in the other study, several substances, other than cypermethrin, have a potential key role on the dermal route, as it is presented in Table 5. If the concentrations of these substances are increased, the risk scores of the dermal routes will accompany this increment. As it was indicated in Peluso *et al.* (2012), in many papers it was considered that organic substances are key contributors to the dermal risk in the context of recreational bathing (Dor *et al.* 2003; Blando and Cohn 2004; Papanyako *et al.* 2008; Filipsson *et al.* 2009). Although in this study the significance of the dermal route was lower than in Peluso *et al.* (2012), the dermal

pathway remains relevant. Because in Argentina the quality of recreational waters is assessed using the concentration limits of the contaminant only for intake (Peluso *et al.* 2011), studies in which the potential harm of substances by dermal routes can be highlighted are extremely important.

In relation to the C risk, this study showed higher values of aggregated and cumulative R than in Del Azul assessment (2.66 E^{-05} *vs.* 1.03 E^{-05}), because of the key role of As concentration, which generates an individual C aggregated risk value that is 75% higher than in the other work. In this study, the major role of the intake route for the most endangered age group was clear. Because of that, as was indicated in Peluso *et al.* (2012), 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 risk values.

Although health risk assessment applied to As in water-related environments based on USEPA models are frequent in the literature, in food (Liang *et al.* 2001; Liu *et al.* 2006; Lee *et al.* 2008; Lin and Liao 2008; Chen *et al.* 2010) or in drinking water (Lim *et al.* 2008; Kavcar *et al.* 2009; Nguyen *et al.* 2009; Man *et al.* 2010; Muhammad *et al.* 2010; Phan *et al.* 2010), studies on the risk by As in recreational waters are much less frequent considering, as it was stated above, there is a small universe of health risk assessment papers based on toxic chemicals in outdoor waters. Although the findings from this study suggest that the risk in general and for As in particular, for recreational water use is low, it must be taken into account that the groundwater in the study area had high As levels. While the Tres Arroyos city population is supplied by a public drinking water distribution system, active domestic wells can remain used for occasional water intake (or frequent—this information is unknown), or for irrigation of vegetables of domestic orchards. The chronic exposure to As by bathing in the creeks could contribute with almost a quarter of the NC risk limit and the entire C risk limit, if we consider the USEPA cancer drinking water limit of 1.0 E^{-06}. Nevertheless, despite these conditions, at present there is no local record of diseases attributable to As exposure.

Several sources of uncertainty remain in our study that may be highlighted, many of which are outlined in Peluso *et al.* (2012). Three of them are: (a) the water sample scarcity without taking into account the sediments beyond those contained in each water sample; (b) using a surrogate population to characterize local exposed residents (bodyweight, skin surface area, exposure duration, lifetime, and intake rate) and the bath patterns (time and frequency of exposure); (c) not considering other sources of exposure as food or domestic water ingestion; in particular, not to consider F as a dangerous substance in bathing activities.

Sediments, cited in the literature as relevant sources of contaminants (Albering *et al.* 1999; Goldblum *et al.* 2006; Filipsson *et al.* 2009); As particularly (Smedley and Kinniburgh 2002), were not considered in our study, beyond those present in water samples. Further work should be conducted to study the sediments in the basin to verify if they are acting as contaminant reservoirs.

The use of a surrogate population to characterize local exposed people is another source of uncertainty, mainly for the variables directly or indirectly related to the bath pattern (time event duration, frequency of exposure, exposed skin surface area). However, in the absence of local information, the Del Azul survey was considered by us a good surrogate source of information, better than other than could be

found in the available literature. Further surveys should be conducted to adjust these exposure variables to the local conditions.

Fish ingestion, another source of risk pointed out in the literature due to the possibility of As bioaccumulation (Phillips 1990; Culioli *et al.* 2009; Shah *et al.* 2009), is not applicable in this case because eating what has been fished is an infrequent practice in this basin. Moreover, data on As bioaccumulation on local fish species are inexistent.

This approach does not consider other potential sources of exposure such as food (*e.g.*, domestic orchard products) or well water ingestion, as was expressed above. This is a limitation of the article's results.

Fluoride, a relevant contaminant of groundwater in the study area, was not included as a toxic substance, which is also a limitation of the study. Although there is no local record of diseases attributable to F exposure, further study will be made considering the integrated risk between exposure by recreational bathing and by domestic well water intake and food. However, at this moment, the information about this integration is scarce, leaving this subject as uncertain.

Beyond these conditions, the health risk assessment is a valid alternative for analysis of recreational water quality, which, unfortunately, is unused by water management agencies in Argentina despite, as it was stated in Peluso *et al.* (2011), the health risk assessment has operational advantages over the traditional management procedure of quality of water analysis.

CONCLUSIONS

The non-cancer (NC) and cancer (C) aggregated and cumulative risks (caused by the simultaneous exposure through different routes of contact to several hazardous substances) for the recreational bathing at Tres Arroyos and Claromecó Creeks were negligible. The simultaneous presence of heavy metals (Al, As, Cu, and Zn), pesticides (α, δ and γ–HCH, aldrin, γ–chlordane, cypermethrin, DDD, dieldrin, endosulfan, endosulfan sulphate and heptachlor), and toluene do not represent a harmful risk for recreational bathers, not even considering aggregated exposure through accidental intake and skin contact to all the substances simultaneously. The age subgroup at greatest health risk was the youngest (5 years old) which had an aggregated and cumulative NC and C health risk scores (Rs) of 3.79 E^{-01} and 2.66 E^{-05}, respectively. The riskiest substance was As (NC and CRs were 2.49 E^{-01} and 2.59 E^{-05}, respectively), followed by cypermethrin and by heptachlor for NC and C health effects, in that order.

The All the Substances NC risk was generated mainly by the intake pathway (almost 70% in the youngest age group), but the importance of this route of exposure decreases according to the age, reaching only 20% in the adult age group, which means that the dermal exposure pathway explains almost 80% of the NC aggregated cumulative risk. For C risk, the role of the intake pathway is higher (almost 90% for the 5-year-old group), decreasing in lesser proportion according to the age.

In both types of risk, the key role of certain parameters related to the bathers' behavior must be pointed out: frequency of the exposure and the bath event

duration. These parameters were major contributors to the variance of the R rather than the substance concentrations.

As had been stated in previous studies of our authorship, considering the variability in the population's characteristics was important to estimate the risk because the exposure parameters related to the bath patterns (the daily bath duration and the annual frequency) had a greater importance than that of the concentration of the hazardous substances in the constitution of the NC and C risk scores. In addition, in this study, it can be appreciated that the key role of the dermal route of exposure that, although lower than in the previous work for the most endangered age group, explains from 30 to 80% of the aggregated NC risk according to the age. This is highly important to highlight due to the practice in Argentina of evaluating the quality of recreational waters using the concentration limits of the contaminant only for the intake pathway of exposure with no distinction between ages of the exposed people. 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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