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# HEALTH RISK ASSESSMENT CASE STUDY OF TRACE METALS IN COLLECTED RAINWATER FOR DOMESTIC USES

Guillaume Busset<sup>1</sup>, Claire Vialle<sup>2,3,\*</sup>, Mireille Montréjaud-Vignoles<sup>2,3</sup>,  
MarieChristine Huau<sup>4</sup>, Séverine Jacob<sup>5</sup> and Caroline Sablayrolles<sup>2,3</sup>

<sup>1</sup>CATAR-CRITT Agro-ressources ; 4 Allée Emile Monso, 31030 Toulouse, France

<sup>2</sup>Université de Toulouse; INP; LCA (Laboratoire de Chimie Agro-Industrielle); ENSIACET, 4 Allée Emile Monso, 31030 Toulouse, France

<sup>3</sup>INRA; LCA (Laboratoire de Chimie Agro-Industrielle), 31029 Toulouse, France

<sup>4</sup>Veolia Eau ; Direction Technique ; Immeuble Giovanni Battista B, 1, rue Giovanni Battista Pirelli, 94410 Saint Maurice, France

<sup>5</sup>Veolia Eau ; 36-38 avenue Kleber, 75016 Paris, France

\* Corresponding author

Claire Vialle - Laboratoire de Chimie Agro-Industrielle , 4 Allée Emile Monso, BP 44362, 31030 Toulouse, France - Phone: +33 5 34 32 35 51 - Fax: +3) 5 34 32 35 97 - E-mail: claire.vialle@ensiacet.fr

## ABSTRACT

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A health risk assessment methodology based on modelling with a Monte Carlo simulation was applied to harvested rainwater that could be used for domestic use in southern France. Firstly a pilot roof run-off collection system was installed and trace metals analysis in collected rainwater were developed. Next, a model of exposure was built based only on chronic ingestion of rainwater during domestic activities. For exposure scenarios, three type of population were investigated: infants, children, and adults. A fourth scenario was used to determine an average risk for an entire lifetime. A Monte Carlo simulation was carried out to assess uncertainty propagation. The preponderant trace metals in reused rainwater were zinc, iron, aluminium and copper, with average concentrations equal to  $4.40 \times 10^{-1} \text{ mg.L}^{-1}$ ,  $5.43 \times 10^{-2} \text{ mg.L}^{-1}$ ,  $5.27 \times 10^{-2} \text{ mg.L}^{-1}$  and  $2.06 \times 10^{-2} \text{ mg.L}^{-1}$ , respectively. When maximum concentration values were used in calculations, hazard quotients did not exceed  $6.4 \times 10^{-2}$ , i.e., about 10% of the risk limit. The highest hazard quotients observed for a person during his or her entire lifespan were found for zinc ( $6.4 \times 10^{-2}$ ) and lead ( $4.5 \times 10^{-2}$ ). These health risk assessment results showed that no risk of non-carcinogenic effects from trace metals could be expected from the harvest and reuse of rainwater for domestic activities.

**KEYWORDS:** heavy metals, ingestion, monte-carlo simulation, reuse, risk assessment , roof run-off

## 1. INTRODUCTION

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A potential lack of fresh water is one of the major issues facing mankind today. This predicament can be attributed to several complex causes, including world population increases, urbanisation, land use transformations, and pollution [1]. Hence, harmful consequences could occur, including both health problems and social conflicts. Among existing solutions, a major interest seems to be given to the use of roof-collected rainwater. Consequently, it would be necessary to measure the amount of available rainwater and its quality. This interest has been shown in numerous recent studies in countries such as Jordan, Canada, Australia, Brazil, and Greece [2-7]. As in many countries, France must conserve its water resources and thus has taken an interest in harvesting rainwater for domestic activities for many years. However, even though there are investigations about rainwater reuse at a large scale, French law currently forbids the use of this water for drinking and for washing the body or clothes [8]. Indeed, it is essential to be certain that there is no health risk before using water for these activities, especially when the collected rainwater is to be drunk. Now, some data are available on this subject. Although some metals, such as iron (Fe), copper (Cu), manganese (Mn), and zinc (Zn), are essential for living organisms at specific concentrations, toxic effects are observed when these concentrations increase. Ingestion of drinking water containing significant amounts of metals may result in adverse health effects, from a shortness of breath to several types of cancer. Currently, agricultural runoff [9] and corrosion of household plumbing systems are important sources of trace metals.

Our study is designed to meet the specific need to assess the possible human health risk linked to the reuse of roof-collected water. Thus, we present here a complete system of harvesting and reuse of rainwater in a house in southern France, and we assess the health risks incurred by a typical family by examining the trace metals that are measured. An important aspect of this health risk assessment lies in the modelling of water ingestion, whose uncertainty is taken into account with a Monte Carlo simulation.

## 2. MATERIAL AND METHODS

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### 2.1. Study Area

The pilot house is located in Rabastens in southwestern France. The village is situated in country at an average altitude of 174 m. The climate is oceanic with a warm summer. Every year, approximately 760 mm of rain falls in this region,

and the average temperature spread is from 7.9 °C to 18.3 °C. The house under study is occupied by a family consisting of two parents and two children. The family uses tap water from the public network for all of their activities that necessitate water (flushing, watering, and drinking). Wastewater is sent to a septic tank for individual treatment. A commercially available rainwater collection system (Sotralentz Habitat) was used in this study. Rainwater is first collected from the tiled roof (204 m<sup>2</sup>). This water is then channelled via open zinc gutters and downpipes to a mesh wire filter before entering an underground PEHD storage tank (5 m<sup>3</sup> capacity) through a calm inlet. Any overflow is fed into a nearby canal. The pumping system used a submerged intake with an inlet filter attached to a float, which pumped the water inside the house through a treatment process composed of a 25-µm filter and an active carbon filter. When insufficient water was available in the tank, a probe activates a valve to allow pumping from a backup drinking water tank. The rainwater collected is available for toilet flushing and can supply two WCs and an outside tap. Figure 1 shows the general system scheme.

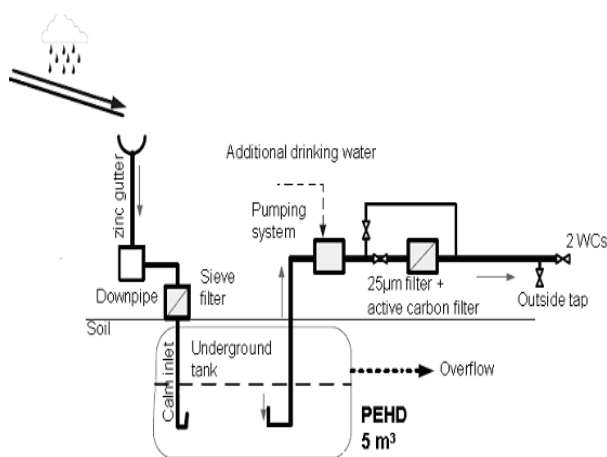


FIGURE 1 - System of rainwater harvesting

## 2.2. Sampling and analysis

Sampling was carried out from January 2009 to January 2010. Water samples were collected monthly from the outside tap, which results in twelve samples. Samples were taken after water had been running into the waste stream for at least one minute. All samples were placed in 120 mL polyethylene bottles that had been previously washed with 10 % nitric acid and rinsed with de-ionised water. Samples were acidified on site with 1 mL of nitric acid and then transported to the laboratory in a chilled cold-box. Metals under investigation were Al, Cd, Cr, Cu, Fe, Ni, Zn, Pb, and Hg. Elemental analyses were performed by inductively coupled plasma mass spectrometry. The analysis method was based on ISO 17294.

## 2.3. Health risk assessment methodology

Health risk assessment (HRA) is a decision-support tool developed by several international organisations, including the World Health Organization (WHO) and the United Nations Environment Programme. This standard is applied by different national organisations, such as the Environment Protection Agency (EPA) and the Agency for Toxic Substances and Disease Registry (ATSDR) in the United States, the National Institute of Risks and the Industrial Environment in France (INERIS), and the National Institute for Public Health and the Environment in Netherlands (RIVM). HRA results can be used to reduce and prevent risks to human health. Indeed, this methodology enables one to determine the risk incurred by human beings if they are exposed to different toxic agents [10]. The different types of toxic effects are divided into two categories, carcinogenic and non-carcinogenic. Non-carcinogenic effects occur from a threshold of concentration of an agent, whereas carcinogenic effects manifest themselves with a certain probability as soon as they are in contact [11]. These effects can appear with oral or dermal exposure or by the inhalation of an agent. Exposure can be acute when it lasts for few hours or few days or chronic if it occurs over several years. Although no international standardisation has yet been established, HRA is mostly carried out in the same way [12]. Hence, we follow this method, which is usually realised in four steps: hazard characterisation, choice of toxicological reference values, exposure scenarios determination, and, finally, risk assessment [10].

*Hazard characterisation:* The first step of an HRA aims to identify the hazardous agents, taking into account their adverse effects on human health. In this study, the substances subject to the risk assessment are the trace metals that were measured during the monitoring campaign, namely aluminium, cadmium, chromium, copper, iron, lead, mercury, nickel and zinc. Moreover, as low concentrations and exposure times affect the entire lifespan, only effects due to chronic exposure are considered. Furthermore, only oral exposure is taken into account, as dermal exposure is underdeveloped in HRA, and the experimental difficulty does not allow us to quantify the number of aerosols that are inhaled. No scientific study on this matter has yet been realised. For these reasons, the adverse effects taken into account in our considerations come from the oral route for a chronic exposure.

*Acceptable Daily Intake selection:* For all of the targeted substances, a dose below which no risk will occur is the Acceptable Daily Intake (ADI). It is expressed in milligrams of agent per kilogram of body weight per day. Generally, this reference dose is established by the EPA, RIVM, ATSDR or WHO and based on several scientific studies on animals or on epidemiologic human data. The ADI only concerns non-carcinogenic effects and are derived from the No-Observed-Adverse-Effect Level (NOAEL) or from the Benchmark Dose (BMD). The NOAEL is defined as the highest observed dose of a substance without adverse effect occurrence. As for the BMD, it is the dose that produces the critical effects with an increase above control levels [12]. Then, ADI is estimated by applying different uncertainty factors, which accounts for the weakest populations or simply allows extrapolation of results on animals to humans [12]. Finally, different ADI can be determined for a substance per type and route of exposure. In accordance to the organisation, the ADI is called a Minimum Reference Dose by the US EPA, Minimal Risk Levels by the US ATSDR and Tolerable Daily Intake by WHO and RIVM. Most ADIs are gathered in the ITER table available on the website of the US National Medicine Library [13]. In this study, the lowest values of ADI have been selected in order to maximise risk evaluation. Table 1 gives these ADI values.

*Exposure:* Exposure scenarios assessment is probably the most important step to assess the overall health risk. Theoretically, it depends on four parameters: route of exposure, activity, type of exposure, and type of population. As decided previously, the route of exposure and type of exposure are respectively limited to ingestion and to chronic exposure. As for activities, six, encompassed by five more general activities, are considered here. Thus, reused rainwater is supposed to be potentially ingested during watering garden, consumption of crude vegetables from garden, floor washing and cross-connection to water supplies. Finally, the type of population is usually divided into three categories by age: infants, children, and adults, being defined by birth to 3 years old, from 4 to 14 years old and from 15 to 79 years, respectively [14]. For infants and children, corresponding average weights have been estimated from individual growth curves (resp. 5 kg and 25 kg). According to the French National Institute of Statistics and Economic Studies, adults are supposed to weigh 70 kg [15]. Exposure is then quantified through the Average Daily Dose (ADD) given by equation 1 (Table 2) [11]. It is then convenient to aggregate all the results for one type of population. The  $ADD_{ing,p,chronic}$  is given by the equation 2 (Table 2). In order to calculate an average risk for the entire life of a person, it is possible to evaluate an average ADD because exposure is chronic [11]. This value is directly calculated with the weighted mean shown in equation 3 (Table 2).

As exposure parameters are very uncertain because of their dependence on behaviour and statistical hypotheses, we built a model based on average and extreme values of volume ingested and frequency of exposure. Then, a triangular density is used to describe each sensitive parameter in order to apply a Monte Carlo simulation.

*Risk assessment:* The last step of HRA is the risk calculation with the hazard quotient (HQ). It has been observed that carcinogenic effects have not yet been shown or quantified for the trace metals under investigation, so the health risk is not calculated for this type of effects. Non-carcinogenic health risks for the ingestion of trace metals are given by the equation 4 (Table 2). Similar to ADD, it is possible to calculate the average hazard quotient ( $\overline{HQ}_{ing,chronic}$ ) for a person during his or her entire life (equation 5 (Table 2)). A hazard quotient equal or above unity ( $\geq 1$ ) means that population is exposed to a risk, whereas if this quotient is below unity, no risks are likely to occur. Calculation of hazard quotients is attained through use of a Monte Carlo simulation. Despite uncertainty on inputs, this tool permits one to obtain a model that represents a pertinent and realistic system profile. Algorithms generate random values that are then applied to the system. From this methodology, a statistical profile of the system is obtained that gives average (most probable value), minimum, and maximum values. For these calculations, we used the software “Simulación 4.0,” de-veloped by José Ricardo Varela (Universidad del CEMA, Argentine)

TABLE 1 - Acceptable daily intakes selected [13].

	ADI (mg.kg <sup>-1</sup> body weight .day <sup>-1</sup> )	Origin (year of determination)
<b>Aluminium</b>	1.0	ATSDR (2008)
<b>Cadmium</b>	1.0x10 <sup>-4</sup>	ATSDR (2008)
<b>Chromium</b>	1.0x10 <sup>-3</sup>	ATSDR (2008)
<b>Copper</b>	1.4x10 <sup>-1</sup>	RIVM (2000)
<b>Iron</b>	8.0x10 <sup>-1</sup>	WHO (1996)
<b>Lead</b>	3.5x10 <sup>-3</sup>	WHO (1993) - RIVM (1999)
<b>Mercury (methyl mercury)</b>	1.0x10 <sup>-4</sup>	RIVM (2000) - US EPA (2001)
<b>Nickel</b>	8.0x10 <sup>-3</sup>	ITER (1999)
<b>Zinc</b>	3.0x10 <sup>-1</sup>	US EPA - ATSDR (2005)

TABLE 2 - HRA equations.

**Equation 1**

$$ADD_{ing,p,a,chronic} = \frac{C_{ing,p,a} \times Q_{ing,p,a} \times F_{ing,p,a}}{W_p}$$

Where  $ADD_{ing,p,a,chronic}$  is the Average Daily Dose for an agent ingested (*ing*) by the population  $p$ , who realises the activity  $a$ , during the entire life (*chronic*),  $C_{ing,p,a,chronic}$  is the concentration of the substance in water,  $Q_{ing,p,a}$  is the quantity of ingested water,  $F_{ing,p,a}$  is the daily frequency and  $W_p$  is the body weight of the population  $p$ .

**Equation 2**

$$ADD_{ing,p,chronic} = \sum_a ADD_{ing,p,a,chronic}$$

$ADD_{ing,p,a,chronic}$  is the Average Daily Dose for an agent ingested (*ing*) by the population  $p$  during the entire life (*chronic*)

**Equation 3**

$$\overline{ADD}_{ing,chronic} = \frac{\sum_p ADD_{ing,p,chronic} \times \frac{T_{p,chronic}}{T_{\mu,p,chronic}}}{\sum_p \frac{T_{p,chronic}}{T_{\mu,p,chronic}}}$$

Where  $\overline{ADD}_{ing,chronic}$  is the average ADD for a person during its entire life,  $ADD_{ing,p,chronic}$  is the ADD for a type of population,  $T_{p,chronic}$  the period of exposure of the population  $p$  for a chronic exposure,  $T_{\mu,p,chronic}$ , the period on which ADI are calculated (in general, the entire life of a person, i.e. 79 years (Afssa, 2009)).

**Equation 4**

$$HQ_{ing,p,chronic} = \frac{ADD_{ing,p,chronic}}{ADI_{chronic}}$$

Where  $HQ_{ing,p,chronic}$  is the quotient risk for an agent ingested (*ing*) by the population  $p$ , during the entire life (*chronic*),  $ADD_{ing,p,chronic}$  the corresponding average daily dose and  $ADI_{ing,p,chronic}$  the corresponding acceptable daily intake.

**Equation 5**

$$\overline{HQ}_{ing,chronic} = \frac{\overline{ADD}_{ing,chronic}}{ADI_{chronic}}$$

Where  $HQ_{ing,p,chronic}$  is the quotient risk for an agent ingested (*ing*) by the population  $p$ , during the entire life (*chronic*),  $ADD_{ing,p,chronic}$  the corresponding average daily dose and  $ADI_{ing,p,chronic}$  the corresponding acceptable daily intake.

### 3. RESULTS AND DISCUSSION

#### 3.1. Trace metal concentrations

After collecting data from one year of exposure to trace metals, a representative profile of the real concentrations of metals under investigation was obtained. Table 3 shows the minimum, average and maximum measured concentrations and compares them to the normal values seen in French drinking water [16]. Limits are exceeded for the maximum concentration measure of iron and aluminium. Moreover, the highest concentrations of lead and zinc are only slightly under the limit. These preliminary results have confirmed that we can analyse the risk incurred by people who would be exposed to these concentrations of metals. Mercury minimum, maximum and average concentrations are the same because as the amount of this metal in the water was under the detection limit, the value chosen was the half of the limit of quantification. With these experimental values, a triangular distribution of concentration is created by the Monte Carlo simulation.

TABLE 3 - Concentrations of trace metals in reused rainwater (mg.L<sup>-1</sup>)

Substance	Number of samples	C <sub>ing,p,a</sub> ( mg.L <sup>-1</sup> )			
		French norm	Average	Min	Max
Aluminium	13	2.0x10 <sup>-1</sup>	5.27x10 <sup>-2</sup>	7.79x10 <sup>-3</sup>	2.26x10 <sup>-1</sup>
Cadmium	13	5.0x10 <sup>-3</sup>	2.40x10 <sup>-5</sup>	5.00x10 <sup>-6</sup>	5.00x10 <sup>-5</sup>
Chromium	13	5.0x10 <sup>-2</sup>	1.70x10 <sup>-4</sup>	3.00 10 <sup>-5</sup>	3.90x10 <sup>-4</sup>
Copper	13	1.0x10 <sup>0</sup>	2.06x10 <sup>-2</sup>	4.76x10 <sup>-3</sup>	4.73x10 <sup>-2</sup>
Iron	13	2.0x10 <sup>-1</sup>	5.43x10 <sup>-2</sup>	4.99x10 <sup>-3</sup>	2.52x10 <sup>-1</sup>
Nickel	13	2.0x10 <sup>-2</sup>	1.45x10 <sup>-3</sup>	3.10x10 <sup>-4</sup>	4.98x10 <sup>-3</sup>
Zinc	13	5.0x10 <sup>0</sup>	4.40x10 <sup>-1</sup>	1.85x10 <sup>-2</sup>	8.61x10 <sup>-1</sup>
Lead	13	1.0x10 <sup>-2</sup>	2.83x10 <sup>-3</sup>	5.50x10 <sup>-4</sup>	6.97x10 <sup>-3</sup>
Mercury	10	1.0x10 <sup>-6</sup>	5.00x10 <sup>-5</sup>	5.00x10 <sup>-5</sup>	5.00x10 <sup>-5</sup>

#### 3.2. Exposure modelling

Exposure modelling is based on our own suppositions about the plausible lifestyle of a French family under these conditions. Nevertheless, uncertainties from this hypothesis are compensated by the choice of a realistic and very large range of values.

*Garden watering:* Infants are not involved in watering the garden, whereas children and adults are supposed to participate in this activity 1 day out of 4 (respectively 1 day of 2) during the 6 month dry period, i.e., 45 (respectively 90) days a year. The uncertainty in these data is represented by a variation of the duration of the activity from 4 months to 8 months.

Next, we defined two ways for rainwater ingestion during watering, namely usual and accidental ingestion. One corresponds to ingesting a few drops and the other corresponds to ingesting many drops, equivalent to a sip.

In order to estimate the amount of ingested water, it was necessary to calculate the standard volume of a drop. We considered that a drop of water is the same as one of rain, whose diameter is known as 2 millimetres [17]. Considering a spherical form, volume of a drop is approximately  $33 \text{ mm}^3$ . We supposed that drops are absorbed during usual ingestion every day of watering, whereas accidental ingestion occurs from one to ten times a year. For a triangular distribution, the number of drops was estimated to spread from 1 to 20 with an average value of 5 for usual ingestion and from 100 to 1000 with an average value of 500 for accidental ingestion.

*Crude vegetables consumption:* Exposure from the consumption of crude vegetables concerns children and adults only. We assume that 100 g of salad retains 10.8 mL of reused rainwater and other vegetables (broccoli and cabbages) retain approximately 4.7 mL [18]. According to investigations on French eating behaviour, an adult eats 142 portions of 40 g of salad per year and 48 portions of 250 g of vegetables per year [19]. As for children, they ingest 51 portions of 40 g of salad and 31 portions of 250 g of vegetables [19]. An uncertainty of 50% was applied.

*Floor washing:* Accidental ingestion during floor washing is assumed to occur in the case of splashing and only concerns adults. We assumed one cleaning per week with an accident frequency from 0 to 52 times a year, with the average value being 5 times a year. The volume of drops is again taken as  $33 \text{ mm}^3$ , and the number of ingested drops is 5, 50, and 100 for the minimum, average and maximum values, respectively. Risks due to detergents mixed with water are not taken into account, even though they are probably the most important consideration in this activity.

*Cross-connection to water supply:* Cross-connection with the public network can cause the blend of drinking water with rainwater. Few studies have been led, with the exception of Australia, where some hypotheses have been made. Thus, when this phenomenon occurs, it is supposed that 50% of drinking water comes from the network and 50% from reused rainwater. However, only one home in thousand would be hit by this accident [18]. Given that such a problem can lead to the ingestion of a large amount of rainwater, extreme scenarios have been established: either there is no mix between different waters, or all the water comes from recuperation system.

Table 4 contains the uncertainty parameters used for exposure calculations. Exposure results for all metals appear in Table 5.

**TABLE 4 - Parameters used for exposure calculation: water volume ( $Q_{\text{ing,p,a}}$ ), frequency ( $F_{\text{ing,p,a}}$ ).**

Type of population	Activities	$Q_{\text{ing,p,a}}$ (in L)			$F_{\text{ing,p,a}}$		
		Average	Min	Max	Average	Min	Max
<b>Infants</b>	Accidental network - drinking water	$3.8 \times 10^{-1}$	$0.0 \times 10^0$	$7.5 \times 10^{-1}$	$1.0 \times 10^{-3}$	$0.0 \times 10^0$	$1.0 \times 10^0$
<b>Children</b>	Accidental network - drinking water	$5.0 \times 10^{-1}$	$0.0 \times 10^0$	$1.0 \times 10^0$	$1.0 \times 10^{-3}$	$0.0 \times 10^0$	$1.0 \times 10^0$
	Ingestion via crude vegetables	$1.0 \times 10^{-3}$	$5.0 \times 10^{-4}$	$1.5 \times 10^{-3}$	$8.0 \times 10^{-2}$	$4.0 \times 10^{-2}$	$1.2 \times 10^{-1}$
	Ingestion via crude salad	$5.0 \times 10^{-3}$	$2.5 \times 10^{-3}$	$7.5 \times 10^{-3}$	$1.3 \times 10^{-1}$	$6.5 \times 10^{-2}$	$2.0 \times 10^{-1}$
	Accidental ingestion from garden watering	$1.7 \times 10^{-2}$	$3.4 \times 10^{-3}$	$3.4 \times 10^{-2}$	$5.5 \times 10^{-3}$	$1.4 \times 10^{-3}$	$1.4 \times 10^{-2}$
	Usual ingestion - from garden watering	$1.7 \times 10^{-4}$	$3.4 \times 10^{-5}$	$6.7 \times 10^{-4}$	$1.2 \times 10^{-1}$	$8.2 \times 10^{-2}$	$1.6 \times 10^{-1}$
<b>Adults</b>	Accidental network - drinking water	$1.0 \times 10^0$	$0.0 \times 10^0$	$2.0 \times 10^0$	$1.0 \times 10^{-3}$	$0.0 \times 10^0$	$1.0 \times 10^0$
	Ingestion via crude vegetables	$1.0 \times 10^{-3}$	$5.0 \times 10^{-4}$	$1.5 \times 10^{-3}$	$1.3 \times 10^{-1}$	$6.5 \times 10^{-2}$	$2.0 \times 10^{-1}$
	Ingestion via crude salad	$5.0 \times 10^{-3}$	$2.5 \times 10^{-3}$	$7.5 \times 10^{-3}$	$3.8 \times 10^{-1}$	$1.9 \times 10^{-1}$	$5.7 \times 10^{-1}$
	Accidental ingestion from soil cleaning	$1.7 \times 10^{-3}$	$1.7 \times 10^{-4}$	$3.4 \times 10^{-3}$	$1.4 \times 10^{-2}$	$0.0 \times 10^0$	$1.4 \times 10^{-1}$
	Accidental ingestion from garden watering	$1.7 \times 10^{-2}$	$3.4 \times 10^{-3}$	$3.4 \times 10^{-2}$	$5.5 \times 10^{-3}$	$2.7 \times 10^{-3}$	$8.2 \times 10^{-3}$
	Usual ingestion - from garden watering	$1.7 \times 10^{-4}$	$3.4 \times 10^{-5}$	$6.7 \times 10^{-4}$	$2.5 \times 10^{-1}$	$1.6 \times 10^{-1}$	$3.3 \times 10^{-1}$

TABLE 5 - Exposure (ADD) and health risk (HQ) results with the Monte Carlo simulation applied to our modelling

	Trace metal	Average	Min	Max	Standard deviation
$\overline{ADD}_{ing,chronic}$ ( $mg \cdot kg^{-1} \cdot d^{-1}$ )	Aluminium	$5.5 \times 10^{-4}$	$1.0 \times 10^{-5}$	$4.4 \times 10^{-3}$	$4.6 \times 10^{-4}$
	Cadmium	$1.5 \times 10^{-7}$	$7.6 \times 10^{-10}$	$9.5 \times 10^{-7}$	$1.1 \times 10^{-7}$
	Chromium	$1.1 \times 10^{-6}$	$2.0 \times 10^{-8}$	$7.7 \times 10^{-6}$	$8.6 \times 10^{-7}$
	Copper	$1.4 \times 10^{-4}$	$1.9 \times 10^{-6}$	$9.9 \times 10^{-4}$	$1.0 \times 10^{-4}$
	Iron	$6.0 \times 10^{-4}$	$4.9 \times 10^{-6}$	$4.4 \times 10^{-3}$	$5.2 \times 10^{-4}$
	Nickel	$1.3 \times 10^{-5}$	$1.1 \times 10^{-7}$	$8.2 \times 10^{-5}$	$1.0 \times 10^{-5}$
	Zinc	$2.5 \times 10^{-3}$	$2.8 \times 10^{-5}$	$1.5 \times 10^{-2}$	$1.9 \times 10^{-3}$
	Lead	$2.0 \times 10^{-5}$	$3.2 \times 10^{-7}$	$1.4 \times 10^{-4}$	$1.5 \times 10^{-5}$
	Mercury	$2.9 \times 10^{-7}$	$5.2 \times 10^{-9}$	$1.2 \times 10^{-6}$	$1.8 \times 10^{-7}$
$\overline{HQ}_{ing,chronic}$	Aluminium	$5.5 \times 10^{-4}$	$1.0 \times 10^{-5}$	$4.4 \times 10^{-3}$	$4.6 \times 10^{-4}$
	Cadmium	$1.5 \times 10^{-3}$	$7.6 \times 10^{-6}$	$9.5 \times 10^{-3}$	$1.1 \times 10^{-3}$
	Chromium	$1.1 \times 10^{-3}$	$2.0 \times 10^{-5}$	$7.7 \times 10^{-3}$	$8.6 \times 10^{-4}$
	Copper	$1.0 \times 10^{-3}$	$1.4 \times 10^{-5}$	$7.1 \times 10^{-3}$	$7.4 \times 10^{-4}$
	Iron	$7.5 \times 10^{-4}$	$6.1 \times 10^{-6}$	$5.5 \times 10^{-3}$	$6.4 \times 10^{-4}$
	Nickel	$1.6 \times 10^{-3}$	$1.4 \times 10^{-5}$	$1.0 \times 10^{-2}$	$1.3 \times 10^{-3}$
	Zinc	$8.5 \times 10^{-3}$	$9.2 \times 10^{-5}$	$5.0 \times 10^{-2}$	$6.4 \times 10^{-3}$
	Lead	$5.7 \times 10^{-3}$	$9.1 \times 10^{-5}$	$4.1 \times 10^{-2}$	$4.4 \times 10^{-3}$
	Mercury	$2.9 \times 10^{-3}$	$5.2 \times 10^{-5}$	$1.2 \times 10^{-2}$	$1.8 \times 10^{-3}$

### 3.3. Risk characterisation

Table 5 also presents the results of the health risk assessment of reused rainwater for trace metals in the case of chronic ingestion. Figure 2 illustrates these results for a person during his or her entire life. Error bars come from the Monte Carlo simulations. Under the meteorological conditions of the Rabastens site, the maximum risks were found for zinc and lead,  $5.0 \times 10^{-2}$  and  $4.1 \times 10^{-2}$ , respectively. Furthermore, the risks for iron and zinc, whose maximum concentrations were found to be higher than those seen in normal French drinking water (table 3), are still lower than the risk limit. The average risk for the maximum concentrations of metals does not exceed 1.1 % of the risk threshold.

The possibility of cross-contamination between collected rainwater and normal water greatly influences the maximum daily exposure dose because the volume and frequency of exposure are higher than those seen for other activities (Table 4). The extreme scenario, which consists of a complete connection to the potable network, was therefore tested. Such a scenario would consider that people only drink collected rainwater for their entire lifespan. Even with these rough hypotheses, we find that hazard quotients remain under the risk limit, with the highest equal to  $5.0 \times 10^{-2}$  for zinc.

From these results, inhabitants who reuse rainwater are mainly exposed to zinc, iron, aluminium and copper. High concentrations of these contaminants may be due to the chemical composition of major components of the system. Indeed, gutters are made with zinc, and some pipes are made with copper. Iron may come from terra cotta tile.

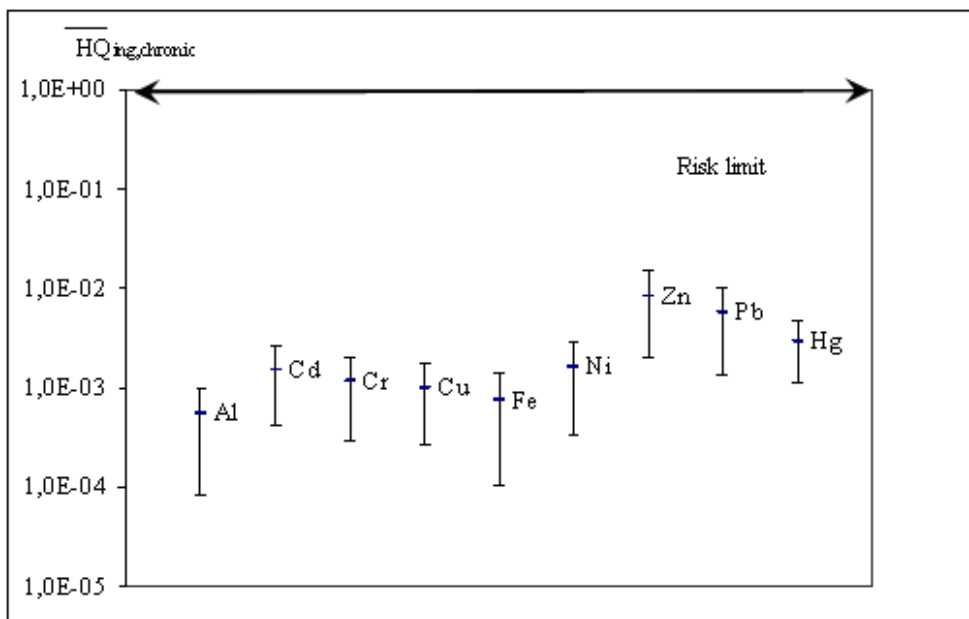


FIGURE 2. Hazard quotients for a person for chronic exposure during his or her entire life

## 4. CONCLUSIONS

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Our modelling of exposure to recuperated rainwater combined with a Monte Carlo simulation has shown that there is no health risk from trace metals in these experimental conditions. Whatever the scenario of exposure, the population is not endangered by the ingestion of rainwater during activities that do not include drinking water and in case of cross-connection. The concentration of trace metals highlighted that some elements with the highest concentrations must come from the system and not from rainwater itself. Other metals may be from atmospheric leaching by rainwater. However, only trace metals were taken into account. This modelling for HRA should be applied to other trace pollutants, such as PAHs or pesticides, and to microbiological parameters, as microbiologic risk is one of the most important risks to be considered.

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