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Drinking water quality in areas impacted by oil activities in Ecuador: associated health risks and social perception of human exposure

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Highlights

- A punctual enrichment in volatile hydrocarbons is observed in several private wells.
- Low mineralization of drinking waters is a special health issue in the Amazon region.
- The bacterial contamination of waters needs more attention by public policy.
- Local stakeholders are unable to provide effective water supply and water treatment systems.
- Social and economic constraints lead people to choose the risks they have to face.

Abstract

The unregulated oil exploitation in the Northern Ecuadorian Amazon Region (NEAR), mainly from 1964 to the 90’s, led to toxic compounds largely released into the environment. A large majority of people living in the Amazon region have no access to drinking water distribution systems and collects water from rain, wells or small streams. The concentrations of major ions, trace elements, PAHs (polycyclic aromatic hydrocarbons) and BTEX (benzene, toluene, ethylbenzene, xylenes) were analyzed in different water sources to evaluate the impacts of oil extraction and refining. Samples were taken from the NEAR and around the main refinery of the country (Esmeraldas Oil

Refinery/State Oil Company of Ecuador) and were compared with domestic waters from the Southern region, not affected by petroleum activities. In most of the samples, microbiological analysis revealed a high level of coliforms representing significant health risks. All measured chemical compounds in waters were in line with national and international guidelines, except for manganese, zinc and aluminium. In several deep-water wells, close to oil camps, toluene concentrations were higher than the natural background while PAHs concentrations never exceeded individually 2 ng.L^{-1}. Water ingestion represented 99% of the total exposure pathways for carcinogenic and non-carcinogenic elements (mainly zinc) in adults and children, while 20% to 49% of the Total Cancer Risk was caused by arsenic concentrations. The health index (HI) indicates acceptable chronic effects for domestic use according the US-EPA thresholds. Nevertheless, these limits do not consider the cocktail effects of metallic and organic compounds. Furthermore, they do not include the social determinants of human exposure, such as socio-economic living conditions or vulnerability. Most (72%) of interviewed families knew sanitary risks but a discrepancy was observed between knowledge and action: religious beliefs, cultural patterns, information sources, experience and emotions play an important role front to exposure.

Graphical abstract

Keywords (max. 6)

Oil activities; Domestic waters; Hydrocarbons; Metal(loid)s; Demineralized waters; Social risk perception.

1. Introduction

Since decades, the petroleum industry has become a significant component of the global economy, but with strong environmental and social impacts (WEC, 2016). A high record of accidents occurring throughout the history of oil production demonstrates the devastating effects and the risks involved for exposed populations (Amiard, 2017; Levy and Nassetta, 2011). In Ecuador, crude oil exportation represents the country's main source of income, exceeding 50% of the total exportations from 2004 to 2014, and settling down at 37% in 2017 (Banco Central del Ecuador (BCE), 2010; Calderon et al., 2016; MCE, 2017). On the other hand, Ecuadorian oil industry is characterized by a long-term history of social and environmental conflicts (Fontaine, 2007).

The oil industry exerted with inadequate environmental standards is strongly suspected of polluting surface and ground waters, particularly in Orellana and Sucumbíos provinces where 80% of total oil spills and 97% of waste pits were recorded (MAE, 2015). During the Texaco oil extraction period (1964-1992), formation waters were directly released into the environment, in forests and rivers (Buccina et al., 2013; Narváez, 2000). However, since the 2000's the Ecuadorian legislation prohibited these practices, binding oil companies to redirect these waters into ancient oil wells (Decree 1215, 2001). Nevertheless, local inhabitants testified that illegal polluting discharges are still released punctually and mainly during the night.

Oil hydrocarbons, in particular PAHs and BTEX, are considered a high environmental and health concern since most of them are persistent and carcinogenic (Amiard, 2017; Jørgensen and Fath, 2010). Certain PAHs and their metabolites can induce leukemia, hepatitis and immunotoxicity; they can also affect the reproductive system and cause genotoxic and carcinogenic effects (IARC, 2010). Twelve PAHs and BTEX have been classified as probable or suspected carcinogens in humans but only benzo(a)pyrene (BaP) and benzene are confirmed as carcinogenic (ATSDR, 2007; IARC, 2018). Despite their high toxicity, volatility and solubility in water, BTEX are not commonly studied; noting that they are responsible for acute and chronic effects including eyes and throat irritation, headaches, tachycardia, leukemia, affections to nervous system, cancer and death (Leusch and Bartkow, 2010; Mitra and Roy, 2011; Neghab et al., 2015). Previous epidemiological studies reported numerous chronic effects including psychological disorders, endocrine and reproductive affections and cases of cancers (Hurtig and San Sebastián, 2002; Levy and Nassetta, 2011; Ramirez et al., 2017; Clinica Ambiental, 2017; Webb et al., 2018). Genotoxic analyses on individuals from the NEAR showed that the exposure to hydrocarbons has caused DNA damage and increased the risk of developing cancer (Paz-y-Miño et al., 2008).

Although hydrocarbons constitute the main fraction (75%) of crude oil, petroleum is a complex mixture of elements, such as sulfur, major cations, anions, trace metal(oid)s (TME) and organic molecules. Major elements (potassium (K), magnesium (Mg), sodium (Na) and calcium (Ca)), and trace elements (nickel (Ni), vanadium (V), iron (Fe) and cooper (Cu)), are naturally present as salts or organometallic compounds in crude and formation waters, while barium (Ba), cobalt (Co), arsenic (As) and lead (Pb) salts are added during the extraction, transportation or storage (Amiard, 2017). Though certain metallic elements are essential for biological functions, most of them could be highly toxic, even at low concentrations, and can cause hazardous effects due to their tendency to bio-accumulate (Jørgensen and Fath, 2010). In 2017, IARC listed cadmium (Cd), As, Cr and Ni as probed carcinogenic elements for humans (lungs, prostate, bladder, kidney, liver and stomach cancers); they can also affect nervous, immune and reproductive systems and can induce abortions, anemia, and teratogenic effects (Fry, 2015).

Released in the environment, hydrocarbons, metals and metalloids can reach aquifers and surface waters throughout all the phases of petroleum production. These pollutants can integrate the dissolved phase or be sorbed onto sediment and particles as their speciation and distribution depend on the physicochemical characteristics of aquatic environments (US-EPA, 2000; Amiard, 2017; Boehler et al., 2017).

Extensive information has been compiled in relation with the signs and symptoms of health problems in oil extraction areas in Ecuador (Arana and Arellano, 2007; Hurtig and San Sebastián, 2005; O'Callaghan-Gordo et al., 2016; San Sebastian, 2001). However, to our knowledge, there is no quantitative determination of the risks involved by drinking water exposure, nor epidemiological studies that connect water quality to suspected cancer and non-cancer risks.

In Ecuador, access to a water supply network has increased in recent years but the percentages of the supplied population are very heterogenous over the territory; in the studied cantons, it varies from 26,6% in Aguarico, to 33,7% in Shushufindi, 41,6% in Joya de Los Sachas and 57,5% in Orellana (SENPLADES, 2014). Nevertheless, in the NEAR, the use of unreliable water sources for domestic purposes (drinking, cooking, bathing and washing clothes) is still a high-risk factor of exposure to toxic pollutants either by dermal contact or ingestion. In Esmeraldas city, along the Pacific coast, where refining activities take place, the water distribution system reaches most of the households but is unreliable supply and safety wise. Therefore, Amazonian and Esmeraldas inhabitants rely on different water sources such as rivers, springs, streams, rainwater, or private wells; sources that are not always safe. Most of the dug wells, in general deep wells (≥8 m), were drilled by oil companies as compensation for the disturbance caused by their activities. However, inhabitants dig private shallow wells (<8 m) given that they don’t trust the groundwater quality. Then, in this context of extended oil activities in a precarious environment, the presence of pollutants can reach the groundwater, as well as superficial waters (Wernersson, 2004; Webb et al., 2018), highlighting a serious health concern for local populations of the NEAR and Esmeraldas areas. But, in Ecuador, similar to most of countries, effective regulations for drinking waters do not exist or are still inadequate (Kayser et al., 2015; Pinto et al., 2012), mainly because they define thresholds for individual elements and not for the mixture of metallic and organic compounds present in water.

Then, this study aims to measure the drinking water quality level (by determining trace metal(loid)s and hydrocarbons contents) and the associated human health risk in areas influenced by oil industry (production and refining activities) in Ecuador**.** The two major questions that motivated our research were: i) are the punctual and local wastewater discharges by oil companies in the Ecuadorian Amazon traceable by chemical analysis? and ii) what are the social responses of local communities to face the perceived contamination of drinking water?

2. Materials and Methods

2.1 Study area

Two oil areas were selected for this study (Fig. 1). The first area is located in the Northern Ecuadorian Amazon Region (NEAR) between 76°30'20"-77°12'00"W and 00°05'01"N-00°55’11”S covering the Orellana and Sucumbíos provinces. This area was defined by the

Ecuadorian Ministry of Environment (PRAS Program, 2010) as the most affected socially and environmentally due to oil activities. The second area is located in the Northern Ecuadorian Pacific Coast (NEPC) between 79°39'27"-79°42'18"W and 00°53'52.00"-00°56'41"N in the Esmeraldas province where most of the crude oil refining is performed. During four decades, these zones were associated to a historic record of oil production and many environmental disasters (Buccina et al., 2013). Both regions were compared with two control areas located in the Morona-Santiago province (77°53'12"-78°34'51"W; 2°58'48"-3°31'20"S) in the Southern Ecuadorian Amazon Region (SEAR) and in the Manabí province (80°19'59"-80°20'57"W; 1°12'57"S-1°13'32" S) Pacific Coast (SEPC), where neither oil nor mining activities has taken place.

2.2 Water sample collection

From 2012 to 2016, we realized five sampling campaigns of surface and drinking waters in the whole study area: two during the low water stage, two during the flood and one during the falling water stage of the Napo River and tributaries. A total of 83 samples of domestic water were collected from local communities from 7 different sources: 15 samples from deep wells (≥ 8 m depth), 8 from shallow wells (< 8 m depth), 10 from rainwater stored in plastic or concrete tanks, 18 from springs, 10 from rivers, 5 from streams, and 17 of households tap water. Among these water samples, 15 were collected from the Pacific coast coming mainly from the water distribution system.

Water sampling was performed in Teflon bottles, using ultra-clean procedures. River waters were filtered (0.45 µm porosity) in the field on Teflon and cellulose acetate pre-cleaned membranes. Each sample was conditioned by addition of nitric acid (for major and trace elements analysis) and hydrochloric acid (for Hg analysis) and kept cooled (4°C) until analysis. For PAHs determination, samples were collected in glass vials (10 and 20 mL) previously calcined at 450°C overnight and kept frozen (-18°C) until analysis. Temperature (T), potential Hydrogen (pH), electrical conductivity (EC), oxide reduction potential (ORP) and dissolved oxygen (DO) were measured *in situ* by ®YSI probes.

2.3 Analytical methods

2.3.1 Major and trace elements concentrations

Analysis of fluorides, chlorides, sulfates and nitrates were performed by Ion Chromatography (Dionex® ICS 200). The cations Ca, K, Mg, Na and Si were determined by Optical Emission Spectrometry-Inductively Coupled Plasma (ICP-OES, Horiba® Jobin Yvon Ultima2). The determination of trace metal(loid)s elements (TME) was performed by Mass Spectrometry-Inductively Coupled Plasma (ICP-MS, Agilent 7500CE). Total Hg analysis were performed according to the US EPA Methods 1631 (US Environmental Protection Agency (US EPA), 2002) using oxidation-reduction with BrCl and $SnCl_2$, and finally analyzed by cold-vapor atomic fluorescence spectrometer (CV-AFS, Brooks Rand®). The quantification limits were calculated as 10 times the reagent blanks standard deviation (SD) and listed in the table SI_1. The recoveries were calculated from different River Water Certified Reference Standards; the percentages obtained for anions, trace metal(loid)s and cations were 104±5% (ION-915), 96±10 % (SLRS-5) and 105±4 % (SLRS-5 and EPOND) respectively. The average recovery for total Hg analysis reached 92±9 % (ORMS-5).

2.3.2 PAHs and BTEX concentrations

Solid-phase microextraction (SPME) was used in immersion mode in combination with gas chromatography-mass spectrometry (SPME-GC/MS) for the analysis of parent and alkylated PAHs and BTEX in water samples (8 mL and 10 mL for PAHs and BTEX, respectively).

An Agilent Technologies 7890A/5975C GC/MS (Palo Alto, CA, USA) was used for the analysis of BTEX. A MPS2XL autosampler (Gerstel®) was used to hold the 65 µm PDMS/DVB SPME fibre (Supelco) during extraction and injection. After the addition of perdeuterated surrogate standards, the fibre was immersed into the 10 mL water sample with agitation (250 rpm) at 40 °C during 30 min. After extraction, the fiber was thermally desorbed for 3 min into the liner of the GC injector port at 220°C in pulsed splitless mode (25 psi, 1.5 min). Helium (6.0) was used as the carrier gas at a flow rate of 2 mL.min^{-1}. Analytes were separated on a DB-624 column (30 m x 0.32 mm, 1.8 µm film thickness; Agilent Technologies, J&W Scientific). The column oven program was as follows: 0°C (5 min) (liquid nitrogen) to 150°C (1 min) at 5°C.min^{-1} and then to 260°C (8 min) at 30°C.min^{-1}. The MSD was operated with electron impact ionization (70 eV) in selected ion monitoring (SIM) mode. The transfer line, source and quadrupole temperatures were 260, 230 and 150°C, respectively. Parent and alkylated PAHs were analyzed using an Agilent Technologies 7890A/5973 GC/MS (Palo Alto, CA, USA) (Kanan et al., 2012). A GC Sampler 120 (Agilent Technologies) was used to hold the 100 µm PDMS SPME fibre fibre (Supelco) during extraction and injection. After the addition of perdeuterated surrogate standards, the fibre was immersed into the 8 mL water sample with agitation (250 rpm) at 40 °C during 60 min. After extraction, the fiber was thermally desorbed for 3 min into the liner of the GC injector port at 270°C in pulsed splitless mode (25 psi, 1 min). Helium (6.0) was used as the carrier gas at a flow rate of 1.3 mL.min^{-1}. Analytes were separated on a HP-5MS-UI column (30 m x 0.25 mm, 0.25 µm film thickness; Agilent Technologies, J&W Scientific). The column oven program was as follows: 50°C (2 min) to 300°C (5 min) at 10°C.min^{-1}. The MSD was operated with electron impact ionization (70 eV) in selected ion monitoring (SIM) mode. The transfer line, source and quadrupole temperatures were 280, 300 and 180°C, respectively.

PAHs and BTEX were quantified by their perdeuterated surrogate standards using their molecular ions (SIM mode). Analyzed compounds (and acronyms) and their limits of quantification (LQs) are listed in Table SI_2 and Table SI_4, respectively. A fiber blank was measured before each water sample by exposing the SPME fiber in an empty vial under the same conditions as the samples. A blank sample (reagents and surrogate standards) and a control sample (blank sample spiked with model compounds) were performed for each batch of experiments in order to check the background contamination and the analytical performance. Whole-method accuracy was determined on spiked water samples (at 10 ng.L^{-1} in Milli-Q water and 50 ng.L^{-1} in Vittel water for PAHs and BTEX, respectively) and remained between 100-105% (CV < 10%) for parent and alkylated PAHs and 89-98% (CV < 10%) for BTEX. Limits of quantification (LQs) were determined from a 10 signal to noise ratio (SNR) observed in low-spiked samples (Table SI_4), except for compounds present in blanks. For these latter compounds, LQs were determined using blank levels, assuming at least a factor three between the analyte mass in the sample and that found in the blank. LQs are in the range 0.3 – 5.0 ng.L^{-1} for parent and alkylated PAHs, and 2 – 9 ng.L^{-1} for BTEX, except for benzene (100 ng.L^{-1}).

2.3.3 Microbiological analysis

Water samples were collected in sterile flasks, refrigerated and analyzed within 12 hours of collection. One mL of raw sample was plated onto selective 3M™ Petrifilm™ 6404/6414/6444 plates (3M Microbiology Products, St. Paul, MN) and incubated following manufacturer instructions. The concentrations of total coliforms and *E. coli* were determined within 24-48 h of incubation time at 30°C. Confirmed coliforms are red or blue colonies with associated gas bubbles. Confirmed *E. coli* are blue colonies with associated gas bubbles. Results are accurate between 25-250 colonies of total coliforms per plate.

2.4 Sociological methods

"Risk culture" refers to the oil contamination risks awareness, behaviours and social practices to cope with it in everyday life, considering another set of risks (social, economic), and the capacity to think the building of future projects at both individual and territorial scale (Becerra et al., 2015). The sociological study was centred on two rural parishes of the NEAR, Dayuma (Orellana Province) and Pacayacu (Sucumbíos Province). We studied how people perceived and managed their environment and the associated contamination risks. Water resources were not the only issue considered in the interviews, while it appeared as the main concern. We focused on both mestizo[1] and indigenous communities. Primary sociological data were collected during five field surveys between 2012 and 2016, most of them during the water sampling campaigns. Our study is novel in interdisciplinary scope, from inception though application, combining faster environmental monitoring with slower-paced sociological studies that facilitate trust-based relationships with study participants. Were realized 160 qualitative interviews, according various levels

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[1] In other words, the individuals who consider that they don't belong to any ethnic group or native nationality because their parents have different geographical and cultural characteristics.

of exposure, living next to possible sources of contamination. Various generations as well as both genders were represented. Around 30 interviews were conducted with elected officials and community leaders. Interviews were built around 4 general topics: 1) History of life; 2) Living conditions (economic incomes), perceptions of oil activities impacts (positive and/or negative), perception of their regulation by local or national authorities; 3) Attitude towards the future (quality of life, future projects); and 4) Biographical data (age, nationality, education level, etc.).

2.5 Health risk assessment

Risk assessments were carried out according risk management measures proposed by the WHO (2014). Since human exposure to multiple chemical mixtures is ubiquitous (polluted air, soil and water), we used US-EPA guidelines (Callahan and Sexton, 2007) for assessing the cumulative risk related to these chemical cocktails and considering different pathways for toxic compounds into the body, with ingestion and dermal contact as main exposure routes (Barraza et al., 2018; US EPA, 2015a,b).

Mathematical equations of the Probabilistic Risk Assessment (as published by Barraza et al., 2018) allow us to evaluate the Hazard Index (HI) and the Total Cancer Risk (TCR), which are defined as the addition of individual quotients of non-carcinogenic (HQ) and carcinogenic (CR) elements respectively, obtained from the average daily dose (ADD), the lifetime average daily dose (LADD) and the references doses (RfD) (US Environmental Protection Agency (US EPA), 2015a, 2015b). For drinking water, we supposed a daily consumption rate of 3 L and 1.5 L for adults and children, respectively and an annual exposure frequency of 350 days per year, assuming that people moved 15 days every year in the Amazon region. Values of HI higher than 1 means that adverse health effects can occur. The TCR is the accumulation of cancer risk values (CR) calculated for carcinogenic elements (As, Cd, Cr, Ni) and PAHs by the product of slope factor (SF) and LADD. The TCR is then compared with the guidelines given by the US EPA meaning the probability of providing cancer risks of 10^{-4} (1 in 10 000) or 10^{-6} (1 in 1 000 000).

2.6 Statistical analysis

Software ©Rstudio (Version 3.5.1) was used for statistical analysis and box plots figures. One sample t-test was used when data showed a normal distribution (Shapiro-Wilcoxon test). In other cases, variables were log-transformed to follow a normal distribution. A confidence level of 0.95 was chosen. Pearson coefficient correlations were calculated to check the significance of the correlations between the parameters.

3. Results

3.1 Physico-chemical parameters

Physico-chemical parameters of drinking waters (Table 1) were compared with the different water quality guidelines (WQGs) reported in the Supplementary Information (Table SI_3).

In the drinking water sources of the Amazon region, the pH values ranged from 3.5 to 8.7 (average of 5.8±1.3) with an extreme minimum value of 2.5 in a forest spring (Shuara 09 oil camp). Most pH values (73%) were in the range of the Ecuadorian quality guidelines. Several water sources were found to be acidic: the river water from the SEAR (4.80±1.47), and the rainwater (pH of 4.84±1.02). The only neutral sources were stream and distribution system waters (7.02±1.38 and 7.16±0.51, respectively).

The EC range (3-1479 µS.cm^{-1}; average: 122±204 µS.cm^{-1}) showed that all the samples were lower than the maximum recommended value (1100 µS.cm^{-1}), except in a deep well (26 m) in Pacayacu (NEAR). Only 19% of the samples were in line with the WQG used in France (200-1100 µS.cm^{-1}): most of the water samples (78%) showed conductivity lower than 200 µS.cm^{-1}, and can be considered as demineralized waters. In the Amazon Region, rainwaters showed the lowest conductivity in both, the oil (11.9±12.9 µS.cm^{-1}) and control area (5.21 µS.cm^{-1}). Only the deep wells (272.2±348.5 µS.cm^{-1}) and system waters (224.3±152.2 µS.cm^{-1}) in the NEAR, and river surface water (237.7±139.2 µS.cm^{-1}) in the SEAR showed conductivity higher than the recommended value (200 µS.cm^{-1}). However, in Esmeraldas, the tap water conductivity never reached the minimum WQG (200 µS.cm^{-1}). Concerning the total dissolved salts (TDS) content of the samples, it was found to be very low falling below the minimum concentration of 100 mg. L^{-1} and the optimal range of 250-500 mg. L^{-1} set by WHO. Only water from deep wells and the distribution system met the minimum recommended value.

3.2 Major and TME concentrations

The analyzed anions (Table 1) are present in concentrations compliant to the local and international standards but lower than the recommended WHO guidelines. For example, all analyzed sources were deficient in fluoride based on the minimum level of 0.5 mg.L^{-1} recommended by WHO (2005) for dental health. Similarly, most of the major cations' concentrations (for example Ca and Mg) were lower than the minimum 2005 WHO guidelines (Fig. 2).

Regarding Al concentrations, all the water sources sampled in the Amazon region (SEAR and NEAR) showed values in a similar and acceptable range (WHO, 2005). While in the public water system (Fig. 3), these concentrations exceeded regularly 200 $\mu g.L^{-1}$, explained by the utilization of aluminum sulfate as flocculent during the water treatment. A similar behavior was observed with Fe concentrations, both elements presenting a common origin from the soil weathering; tropical, ferralitic soils are characterized by elevated Al and Fe contents (Mainville et al., 2006).

Manganese concentrations in drinking water sources ranged between 0 and 2373 $\mu g.L^{-1}$, and averaged 72 $\mu g.L^{-1}$; the highest concentrations being measured in a deep well close to Pacayacu village in a private property (Pichincha 11, Libertador oil camp). In the majority of private water wells, Mn concentrations exceeded the recommended values by Ecuador (100 $\mu g.L^{-1}$), and *a fortiori* France and Canada (50 $\mu g.L^{-1}$), while in rivers, springs and water systems, they were acceptable (Fig. 3).

The Zn concentrations in samples collected from rainwater tanks were significantly higher than other sources (Fig. 3). Even though high concentrations of this element are not of health concern, a limit of 500 µg/L was considered as WQG in France, being the most restrictive value among the legislations consulted; all rain waters exceeded this limit. In isolated areas of the Amazon basin, people collect rainwater from their home's roof. The acidic rain water passing through the roofs leached, dissolved and transported dissolved Zn to water tanks.

The elements As, Cd, Cr, Ni and Pb and Co are considered carcinogenic (group 1 and 2A), while oxides of Sb, V and Mo are defined as possibly carcinogenic (2B) by the IARC.

System water and (deep and shallow) wells presented the highest values of As, Ni, Cr, Pb, Sb, and V without exceeding the WQGs. Chromium concentrations were very low (average of 0.56±5.44 µg.L^{-1}) and the maximum value (10.57 µg.L^{-1}), lower than the WQGs, was measured in the Dayuma tap water. The increase of these carcinogenic elements may come from the pipework quality of the water distribution system. Antimony in rainwater presented also a mean concentration (0.087±0.097 µg.L^{-1}) higher than in the other sources, but lower than the recommended limits.

3.3 PAHs concentrations

All aromatic organic compounds (PAHs and BTEX) concentrations measured in the analyzed drinking waters (Table SI_4) were in line with the WQGs, even in the NEAR. The benzo(a)pyrene considered as carcinogenic (group 1) was under the quantification limit in all the samples. However, high values of toluene (2000 to 6000 ng.L^{-1}) were measured in deep wells of the "12 de Febrero" community in San Carlos village located on the Sacha oil field. Benzene (also classified as carcinogenic group 1) was present in the water from the distribution system in Esmeraldas (NEPC) and Portoviejo (SEPC) sampled in 2016 but these significant concentrations (490 to 710 ng.L^{-1}) close to the WQG (1000 ng.L^{-1}), were not detected in 2017. The results of 2016 can be due to the contamination of the water stored by residents in plastic or metallic tanks as a solution for the frequent water network shortages. Our results show that there is a risk of accidental and local contamination of drinking water by benzene in collection tanks from traffic or industrial emissions in Esmeraldas city.

3.4 Bacteriological results

One of the main health concerns was the presence of *Escherichia Coli* and total coliforms in water sources sampled in the Amazon region which could reach in deep wells up to 40 and 700 CFU.100 mL^{-1}, respectively, except in the public water distribution systems. According to international regulations on drinking water quality, *Escherichia Coli* and total coliforms must be absent in drinking water. Untreated wastewaters, agricultural runoffs, migration and development of microorganisms in wells and shallow surface waters exposed to light and high temperatures can contribute to the development of coliforms. Therefore, it would be crucial to disinfect (e.g. boiling, chlorination) any kind of water sources in the Amazon region prior to consumption to prevent any risk of water-borne diseases.

3.5 Human health risks assessment

For both adults and children (Table 2), the ingestion hazard index (HIing) showed a contribution of 99 to 100% to the non-carcinogenic total health risk (THI). Compared to the reference value (US EPA, 2015), 7% and 27% of the THI obtained for deep well water sources was higher for adults and children, respectively.

For adults, the HI values ranged from $3.4x10^{-02}$ to 2.6 and from $1.4x10^{-04}$ to $2.0x10^{-02}$ for ingestion and dermal routes, respectively. From the sites that shows HQ values > 1 (Table SI_5), two samples came from the NEAR, with the highest value (THI=2.6) in Dayuma due to high aluminum concentrations (HQ=1.8) in the system water, and in a deep well (HI=1.1) with an elevated hazard quotient in Mn (HQ=0.75). Along the Pacific coast (NEPC), only one sample showed a HQ value > 1, corresponding to a tap water collected from a pumping and storage station in front of the discharge channel of the main refinery of the country; this elevated HI (1.8) is mainly explained by the Cd content (HQ=1.5).

For children, no contribution of dermal exposure was found in the total hazard index. The ingestion route in the THI ranged from $7.5\text{x}10^{-02}$ to 5.5. From all the samples, 9 (11%) were higher than the reference value (US EPA, 2015). The highest THI values were found in deep wells waters of the NEAR, two in Pacayacu (HI =1.6 and 0.61 due to Mn concentrations), and two in Dayuma (HI=1.32 and 1.38 explained by As concentrations), in a private well and in the stadium, respectively. Individually, hazard quotients >1 in rainwater were found in the whole Amazon region, being Zn the element that contributed to most of the total hazard index (HQ of Zn=1.01 and 1.06, in the NEAR and SEAR respectively). Finally, in the system water from Dayuma (NEAR), we found one point with a high concentration of Al in Auca 09 field corresponding to THI equal to 3.9. In the NEPC, the highest risk explained by Cd (HI=3.13) and As (HI=0.38) concentrations were found in Esmeraldas, in the potable water pumping station and in a particular house, respectively.

For both, adults and children, water from a deep well located in the stadium of Dayuma (NEAR) presented the highest contribution (93%) of carcinogenic molecules in the dermal-HI, while regarding the ingestion risk contribution, system water from the pumping station close to the refinery exhibited a contribution of 88% to the total HI.

It is interesting to notice that, the hazard quotient of arsenic represents between 20% and 49% of the total ingestion HI (Fig. 4), while As concentrations never exceeded 3 $\mu\text{g.L}^{-1}$. In the Total Cancer Risk (TCR) calculated for carcinogenic elements (As, Cd, Cr and Ni), arsenic contributes with 51% to 100% in 65 points (78% of the total sampling points). In Esmeraldas, Cd contributes with 56% to 94% of the TCR in 4 system water samples.

4. Discussion

4.1 Impacts of oil activities on drinking water quality

Vanadium, Ba, Ni, Co, Mo and PAHs (except AC, IP, and Pe) are the main elements and compounds that can trace oil activities impacts in the hydrological system, due to their high contents in the Ecuadorian Amazon crude oil (Table SI_6). High concentrations of V (305±36 $mg.kg^{-1}$), Ba (133±5 $mg.kg^{-1}$), Ni (99±11 $mg.kg^{-1}$), Co (1.82±0.04 $mg.kg^{-1}$), and Mo (0.93±0.02 $mg.kg^{-1}$) were measured in a topsoil sampled in a waste pool (from the Auca 08 oil well, NEAR). These values were slightly higher than the concentrations measured in fresh crude oil from the same oil camp (Auca 03), except for barium (7±1 $mg.kg^{-1}$). However, the crude from the national oil pipeline showed slightly lower values of toxic compounds than these in the waste pool soil except for PAHs (Table SI_6). Barium is then considered as a relevant element to trace oil activities since barite (barium sulfate) is commonly used in drilling activities. This element in surface and groundwaters mainly comes from the soil drainage, as in Ecuador these sources are defined as calcium bicarbonate waters where barium salts may precipitate easily (Kabata-Pendias and Szteke, 2015). All the Ba values measured in the study drinking waters were in line with the WQGs. The only elements that exceeded the WQGs thresholds were Al, Fe, Mn, Cu and Ba; these elements mainly originate from the natural soil erosion and are not of health concern at observed concentrations except for Mn. The water sample from Pacayacu (Pichincha 11 oil camp) presented concentrations in most of the analyzed elements and compounds 10 to 100 times higher than the sampling points average. Manganese concentrations were higher than 50, 100 and 500 µg.L^{-1} in 11%, 9% and 4% of the samples, respectively. The highest concentrations were all observed in private or public deep wells. Manganese can be related with agriculture practices, as it is one of the main components in fertilizers, animal food, and pesticides. As proposed by Van Wendel de Joode et al. (2016), drinking water risk assessment should consider Mn as a health hazard. Several studies have shown a negative association between elevated Mn concentrations in drinking waters and children's neurodevelopment, behavior and academic achievement (Bouchard et al., 2011; Van Wendel de Joode et al., 2016).

Regarding organic toxic compounds, no pollution by hydrocarbons was found in the study water samples. The unique concern highlighted in drinking water sources which can originate from the oil activities, was the toluene and the sum of methylnaphthalene concentrations measured in several deep wells in the San Carlos community (located on oil camps managed by the Rio Napo Company), nevertheless without exceeding individually, national and international guidelines. Individual alkylated PAHs have been observed to have potentially mutagenic, tumor-promoting, or carcinogenic activity and their toxic potential may easily surpass that of the parent compounds (Andersson and Achten, 2015). However, the concentrations of di- and tri-methylnaphthalenes were very low, but except for 1- and 2-methylnaphthalene, insufficient toxicity data is available to quantify cancer risk from chronic exposure to individual or mixtures of alkylated PAHs. Considering the concentrations ranges of carcinogenic PAHs, we may suppose that direct ingestion of water and dermal contact are probably not the main exposure routes. This hypothesis was confirmed by the Total Cancer Risk indexes calculated for parents and alkylated-PAHs (section 3.5).

On the other hand, we observed just two significant correlations between toxic compounds and oil activities impacts. These moderate and positive correlations were found between the number of pits in the vicinity area of the sampling points and MA (2-methylanthracene; r=0.43) and total dissolved Hg (r=0.47) concentrations. However, no correlation was found with the oil spills frequency, nor with the number of flares or oil waste pools.

4.2 Drinking water risks due to low mineralization

In the whole Ecuadorian Amazon Region and along the Pacific Coast, the study water sources presented very low mineralization levels, till 10 to 100 times lower than the WHO's minimum recommended concentrations (WHO, 2006), with certain exceptions in deep wells. Although high concentrations of certain major elements can affect the human health (Nerbrand et al., 2003), it is very important to pay attention to acid and low mineralized water sources as well. Acid pH water can accelerate the leaching of toxic metals and other compounds from roofs or distribution pipes and modify the taste. Acidic pH is characteristic of rainwaters which feed spring and shallow groundwater and is the main source of drinking water for local people. The percolation of rain waters in tropical soils of the Amazonian plain does not change the water pH as these soils are acidic and highly weathered. Additionally, the lack of minerals can cause direct health effects like tooth caries due to the lack of fluoride, malfunction of the intestines, reduction of calcium and magnesium intake and other essential elements as well as an increase of intake of toxic elements, due to their dissolution in acid waters during the cooking for example (WHO, 2006). For about 50 years, epidemiological studies in many countries all over the world have reported that soft water (i.e., water low in Ca and Mg) is associated with increased morbidity and mortality from cardiovascular disease (CVD) compared to hard water and water high in Mg (Kozisek, 2005). Epidemiological studies of an ecologic design among Russian populations supplied with water varying in TDS suggested that low-mineral drinking water may be a risk factor for hypertension and coronary heart disease, gastric and duodenal ulcers, chronic gastritis, goiter, pregnancy complications and several complications in newborns and infants, including jaundice, anemia, fractures and growth disorders (Turnlund, 2002). The intake of low water-content in Ca, is associated with a higher risk of fracture in children, certain neurodegenerative diseases, pre-term birth and low weight at birth and some types of cancer (Kozisek, 2005). The WHO established in 1980 minimum and optimum levels of minerals and recommended that demineralized water contains: for Mg and Ca, a minimum of 10 and 20 $mg.L^{-1}$, respectively, and an optimum range of 20-30 and 40-80 $mg.L^{-1}$, respectively. For total water hardness, the sum of Ca and Mg should be 2 to 4 $mmol.L^{-1}$ (Kozisek, 2005). In rainwater, the most important drinking water source for Amazonian communities, the Mg and Ca average concentrations were less than 0.01 and 1.28 $mg.L^{-1}$, respectively. Average concentration of Na in rainwater was 0.12 $mg.L^{-1}$, 2000 less than the minimum recommended value (WHO, 1980).

Another issue caused by the regular domestic use of low mineralized water is the loss of essential minerals from food during the cooking, mainly of Mg and Ca and in a less extent of essential microelements (Cu, Mn and Co). A low diversified diet due to precarious living conditions coupled to the domestic use of demineralized water reduce the nutrients intake by local populations and participate to the child undernutrition.

It appears that the health risk tied to the chronic exposure to low mineralized water can be as important or in some cases, more important than the health risk related with current oil activities. Though, it is necessary to pay attention to specific sampling points (deep wells) were the total hazard index and total cancer risk denote that inorganic elements related with oil production (as V or Mo in surface water, Table SI_5) can contribute significantly to the sanitary risk.

However, the quality of rainwater may subsequently deteriorate during harvesting, storage and household use (WHO, 2011). In the Amazon region, many families collect rainwater from their house' roof, explaining the very high concentrations of Zn measured in this water source. Later, they store it in large plastic or metallic tanks without any cover where wind-blown dirt, leaves, insects, fecal particles, or particles from the atmosphere or from burning materials (e.g. trash or oil flares) can contaminate the stored rainwater.

4.3 Limits of Ecuadorian Regulations

We suggest three types of limits in the National environmental regulations: legal, technical and political.

4.3.1 Legal limits

In Ecuador, two regulations have been set up: the INEN 1-108 and the TULSMA (Texto Unificado de Legislación Secundaria del Ministerio del Ambiente). The Ecuadorian regulation INEN 1-108 is related to treated water for human consumption, and certain parameters, such as pH, conductivity or oxygen percentage were omitted from the 2014 latest version compared to the 2006 version (Table SI_3). Regarding the inorganic compounds, from 34 parameters considered in the 2006, only 18 were proposed in 2014. For example the Mn threshold values increased from 0.1 $mg.L^{-1}$ in 2006 to 0.4 $mg.L^{-1}$ in 2011, to completely be removed in 2014. From 5 PAHs regulated in 2006, INEN reduced to one molecule in 2014, the Benzo(a)Pyrene, with a maximum permissible value (MPV) higher than in the 2006 version. For BTEX molecules, toluene MPV became more permissive, and ethylbenzene regulation disappeared. A restriction of the maximum permissible limit for microbiological parameters was also observed, reducing the MPV for fecal coliforms from 2 NMP.100 ml^{-1} in 2006 to 1.1 NMP.100 ml^{-1} currently, and eliminating total coliforms guidelines.

The second Ecuadorian regulation, TULSMA, is applicable to natural water sources for human consumption prior to any treatment. In its last version (November 2015), this regulation only considered treated-clean waters and raw water that needed a physical treatment and presented a lower number of parameters than the former text. The MPV of some elements, such as Cd, increased from 0.003 mg.L^{-1} in February, 2015 to 0.02 mg.L^{-1} in November, 2015. Concentrations of PAHs and BTEX in drinking water sources are not regulated. The current version for fecal coliforms set up a lower concentration (1000 NMP.100 ml^{-1}) than the previous ones (2000 NMP/100 ml) but it still remains very high.

Then, during the last decade, both Ecuadorian regulations, INEN and TULSMA, have not just reduced the number of control parameters but also proposed less restrictive values without considering international recommendations. Since international regulations (Santé-Canada, WHO, US-EPA and European Union) are based on epidemiological and risk assessment studies, we considered them in this study as more adequate water quality references for the health risk assessment discussion. Nevertheless, concentrations thresholds are related to individual elements or molecules, or for groups of specific metal or organic contaminants but not for chronic exposure to cocktails of both categories of toxic elements and compounds.

4.3.2. Technical limits

These limits are mainly due to the weakness of technical capacities of local stakeholders. In the different visited communities, we found collective water supply systems installed by local autonomous governments that were often ineffective. Many of these systems were abandoned because of the lack of water availability, and/or of pump maintenance. This ineffectiveness reflects two realities combined in the Amazon region: i) the lack of technical ability of the projects managers and ii) lack of proper management of public resources (Juteau-Martineau, 2019). Sometimes, under the pretext of public utility projects, public funds can be redirected to non-public matters.

4.3.3. Political limits: political ability to relay the social demands

People in charge of community relationships between public oil companies and local inhabitants have always negated the link between river's contamination and oil activities. According to them and policy makers, water quality degradation is mainly due to the presence of total and fecal coliforms, given that wastewater treatment is not available in the country, except in big cities. They don't constitute any opposition force that would set up a water quality monitoring or at least the compliance of safety regulations. The latter does not reflect the social experience of inhabitants of the oil region, with what they observed in their own farm or in the close river, this experience leads them to ask for remediation, compensation and indemnity. The local political ability to relay the social demands varies from place to place. For example, in the Management Plan of Pacayacu, oil activities are not accounted as responsible for the poor water quality, in contrast to the Dayuma Plan that holds oil companies responsible for the contamination of water resources, even during environmental remediation activities.

4.4 Social perception of the risk and human exposure

Perceived environment and perceived exposure to health risks

In the Amazon basin, traditionally, indigenous and mestizo use water sources close to their home, from rivers, streams, rainwater or shallow wells. But in the oil Amazon basin, the lack of access to drinking water is the major concern expressed by communities living close to oil infrastructures (Fig. 5), not only for inhabitants but also for cattle. The social experience of the environmental pollution is stable with time and the general resentment is that “everything is polluted and oil companies are lying”. Contamination is mainly perceived empirically, directly from the farms or indirectly from a decrease in aquatic biodiversity.

The interviews conducted in the NEAR showed 2 main social responses to perceived contamination of local water resources, *Exit* and *Voice* (as proposed by Hirschman, 2011): *Exit* means leaving the current water supply. At the household level, harvesting rainwater is today perceived as the safest and most effective way to collect water even if in fact various sources are combined (rainwater, wells, rivers or spring). Harvesting rainwater has undergone a rapid development thanks to the social compensation programs systematically implemented by the National oil Company, Petroecuador, since the 2000’s. During oil exploration campaigns or when opening new platforms, the public company offered zinc plates to cover the roofs of many communities’ houses and water tanks to store the clean water. Later filtration systems were offered by NGOs to specific families involved in the protestation against oil activities. At the community scale, collective action took place: for instance, two communities close to the urban centre of Pacayacu developed their own drinking water supply system within a "*minga*", an indigenous practice of cooperative and voluntary work for the common good. One of them has failed due to the lack of technical and financial maintenance.

Voice: In the centre of Pacayacu village, most of the inhabitants contrary to the position of the leaders, do not trust the quality of the drinking water system and choose the « voice » thanks to the help of the NGO “Acción Ecológica”. The mobilization, coordinated by this same NGO, operates in different ways: creation in 2009 of the “environmental clinic” as an alternative program to restore the environment and livelihoods of about 30 families living in the oil impacted area; in 2011, opening of a defense committee of families that perceived a local contamination of their water resources, and organization of “Toxic-tours” in Atacapi and Libertador oil camps.

Facing the risk but to which risk high priority is given?

The perception of the contamination risk was largely shared in the interviewed families as 72% of them knew about sanitary risks linked to bad water quality. But there was a discrepancy between knowledge and action, as social investigations underlined for other risks (Weiss et al., 2011): existence of the first (knowledge) does not guarantee the second (action). It is important to distinguish between the drinking water supplies: primarily public water system in urban centres and rain, river or private wells in rural areas. Paradoxically to the perceived sanitary risk, the adopted strategy is not always to reduce the exposure. An oil spill in a private farm is generally conserved as a proof of oil activities damages and used to ask for material compensation (job or material good) or money (Becerra et al., 2016). Similarly, a riparian who saw his water resource damaged by an oil spill for example, won’t necessary look for other water sources for domestic use.

The role of religious beliefs, cultural ideals, experience and emotions were evaluated in order to understand why people resist to change their practices to face supposed polluted water sources.

Religious beliefs. Data collected from interviews with mestizo people showed that culture risk is highly influenced by religious beliefs in at least two ways. First, faith can bring a certain form of fatalism towards one's own existence. Some individuals could also stop worrying about the future since God decides for them. Data collected gave numerous references to biblical scriptures.

Cultural ideals and practices. Change to prevent sanitary risk depends first on social perceptions of the risks, and secondly on a cultural process, but also depends on the existence of other available alternative ways and on adequate financial resources. In the Amazon forest, Shuar and Kishwa communities have a strong reliance on nature and surface waters because of their ancestral culture. Their spirituality is based on the principle of Mother Earth as well as deities of the jungle, the agricultural garden ¨*chacra*¨ and the water. They usually leave far from towns or oil infrastructures. Even if these groups tend to evolve toward a cultural hybridism (for instance integrating modern food to their diet), they prefer natural food and water from the farm, rivers or mountains (Beguet, 2015). Furthermore, in indigenous groups, environmental pollution is unequally perceived. We observed in Shuar and Kischwa groups that generally, this perception by women is less noticeable than by men: maybe because women usually don't work for oil company (Racines, 2017).

Sensitive experience. Danger is learned by sensitive experiences which could be traduced by: "*the kick warns you and the damage teaches you*". Since health effects of environmental exposure are not immediate, people tend not to change their practices and the collective action stops to the suspicion or denunciation. Social determinants such as the high rates of unemployment and poverty (Larrea and Camacho, 2013) explain the difficulty to get out from this situation of "environmental suffering". People respond to the contamination of water in a confused way. As explained by Auyero and Swistun (2007) often people under these conditions have "toxic confusion", as awareness of their situation is distorted by lack of knowledge of the real causes which leads to inappropriate behaviors.

Chronic and historical distrust of oil companies.

Due to numerous social liabilities between oil companies and Amazonian communities, health issues related to water contamination are attributed by inhabitants to the impact of oil activities which obscure the complexity of the causes neglecting all the historical and political context that is responsible for poor human health, limits the perception of their own responsibilities and inhibits practices changes. Because of this situation that despite the awareness of rivers and water being contaminated, people are not changing their daily practices based on the hope that finally, there will be some recognition and retribution due to the damage caused by the oil companies during the last decades. Each emergence of cancer cases, whatever its origins, nourished the proofs list against oil companies' activities. The social liability acts as a psychological lock that convinces inhabitants and NGOs that only oil companies are responsible of the environmental and sanitary hazards of the region.

5. Conclusion and recommendations

Contrarily to the common prejudices, this study showed for the first time that the health risks due to drinking water exposure could be more impacted by the precarious living conditions than by the oil activities in the Ecuadorian Amazon region. Local people routinely chose the water sources based on the end-use, availability and some of them regarding their perception of contamination risks especially after an accident (oil spill).

International and national regulations are generally based on maximum acceptable concentrations of inorganic and organic compounds because water may contain elements and molecules that are undesirable. Main of the different water sources sampled in Ecuador can be considered as suitable for human consumption. It is important to control the quantity of aluminum sulfate in the system during the water treatment as well as the way of collecting and storing rainwater. Regarding Mn, a well-known neurotoxin, the high concentrations observed in private wells can also be related to agriculture (use of pesticides; van Wendel de Joode et al., 2016). Arsenic which contributes from 20% to 49% to the ingestion health index, is naturally present in high concentrations in soils and waters of most of Latin America regions (Bundschuh et al., 2012). But the novelty of this study is the low mineralization of the drinking water sources which could be an important health issue due to the deficiency in essential minerals like Ca, Mg and F. The addition of salt enriched with fluoride and iodine into the cooking water is a common practice, but not used everywhere in Ecuador, especially in remote areas. Most of the symptoms as fatigue, stomachaches or dizziness, often signaled by the population can be related with the low concentrations of minerals in their domestic water.

In Ecuador, agricultural runoffs and discharges of household effluents into water bodies without previous waste treatment seems to be the main source of microbiological pollution, confirmed by the presence of fecal coliforms in almost all the drinking water samples. It appears primordial before consumption, to disinfect or boil any kind of water sources before use in areas where there is no access to treated clean water. At the national level, public policies should address this problem by extending rural potable water supply and waste water treatment systems to the communities living in remote areas of the Amazon region.

Although no toxic compounds in drinking water samples exceeded the international water quality guidelines, human co-exposure to low mineralized water and volatile organic molecules needs more attention in oil impacted areas. Local people living in these areas still endure the insufficient recognition of historical impacts of oil exploitation by the Ecuadorian government, confirming that information can be considered as a sort of power that should be shared. Till now, without any satisfactory processing of the historical impacts of the oil activities in the country, inhabitants of the NEAR focus their problems on oil companies ignoring other possible sources, such as agriculture practices or the lack of water treatment. The belief held by many of the inhabitants that oil companies are the only reason for water pollution is part of a symbolic process by which water pollution crystallizes the set of social and environmental claims in front of oil industry. Water quality preservation is the most shared argument that can become a symbol of the oil companies' environmental debt and of the government to solve this issue.

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Figures captions

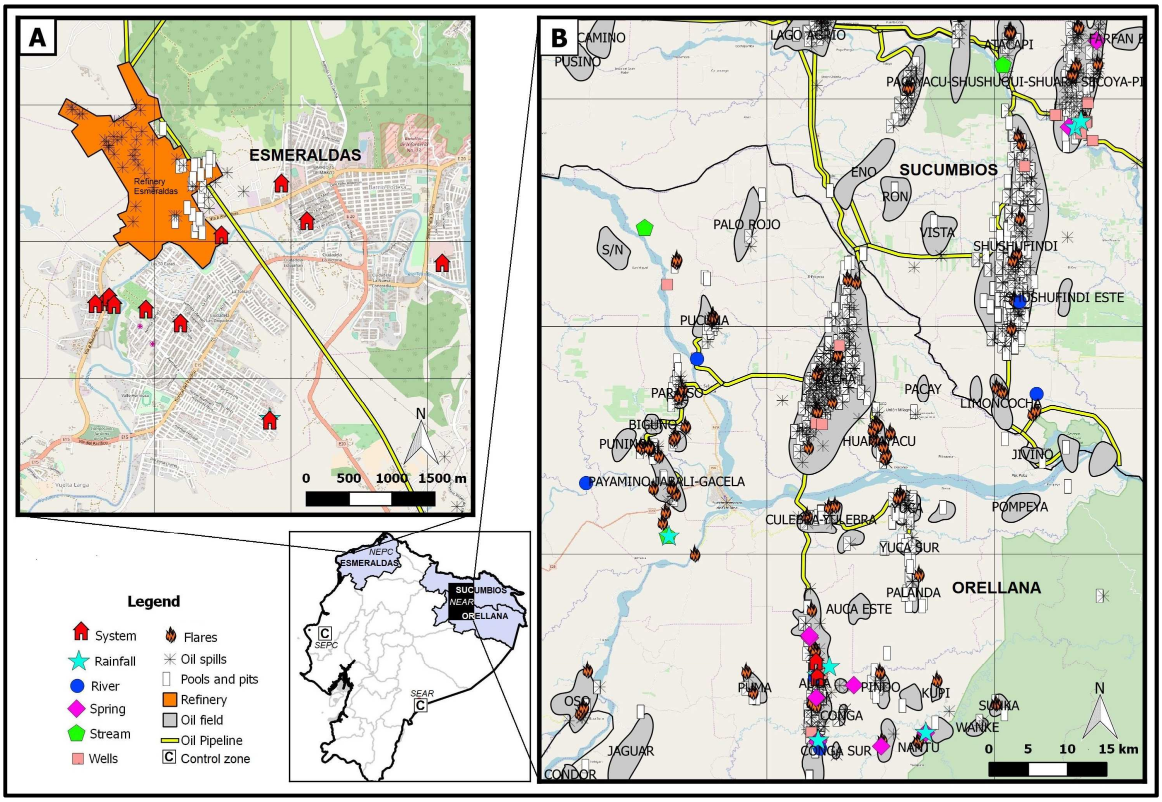
Figure 1. Hazards description (flares, oil spills, pools and pits, refinery, oil fields and pipelines) and sampling points of drinking waters (from various sources: rainfall, river, spring, stream, well) in four study areas: A) Northern Ecuadorian Pacific Coast (NEPC), B) Northern Ecuadorian Amazon Region (NEAR) and C) in two control zones (Southern Ecuadorian Amazon Region (SEAR) and Southern Ecuadorian Pacific Coast (SEPC)).

Figure 2. Major element concentrations in drinking water sampled during the 2012-2016 period, in (A) the Northern Ecuadorian Amazon Region (NEAR) and in (B) the control zone in the Southern Ecuadorian Amazon Region (SEAR); the red and blue lines report maximum and minimum values recommended by the World Health Organization for drinking water (2005) respectively; international water quality guidelines (WQG) are reported.

Figure 3. Al, Fe, Mn, Zn and Ba concentrations in drinking water sampled during the 2012-2016 period, in (A) the Northern Ecuadorian Amazon Region (NEAR) and in (B) the control zone in the Southern Ecuadorian Amazon Region (SEAR). The red and blue lines report maximum and minimum values recommended by the World Health Organization for drinking water (2005), respectively; international quality guidelines (WQG) are reported.

Figure 4. Distribution of ingestion and dermal contact Hazard Index (HI) for the Amazon and Pacific Ecuadorian regions, followed by the percentage of samples that overcome the Total HI reference value (US EPA, 2015b) in children and adults.

Figure 5. The 20 most cited words in Spanish by local people living in Dayuma and Pacayacu villages (©NVivo simulation).



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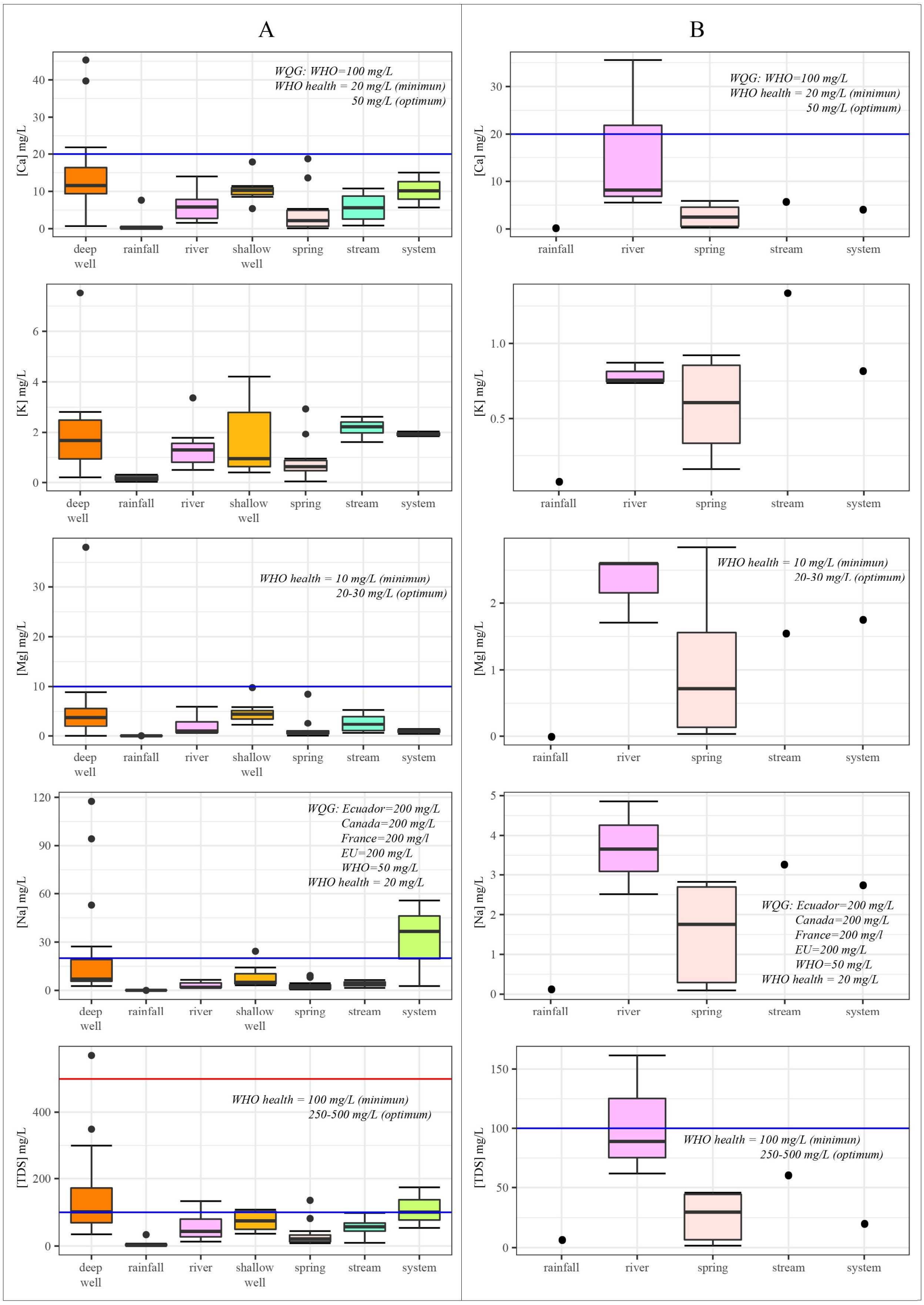

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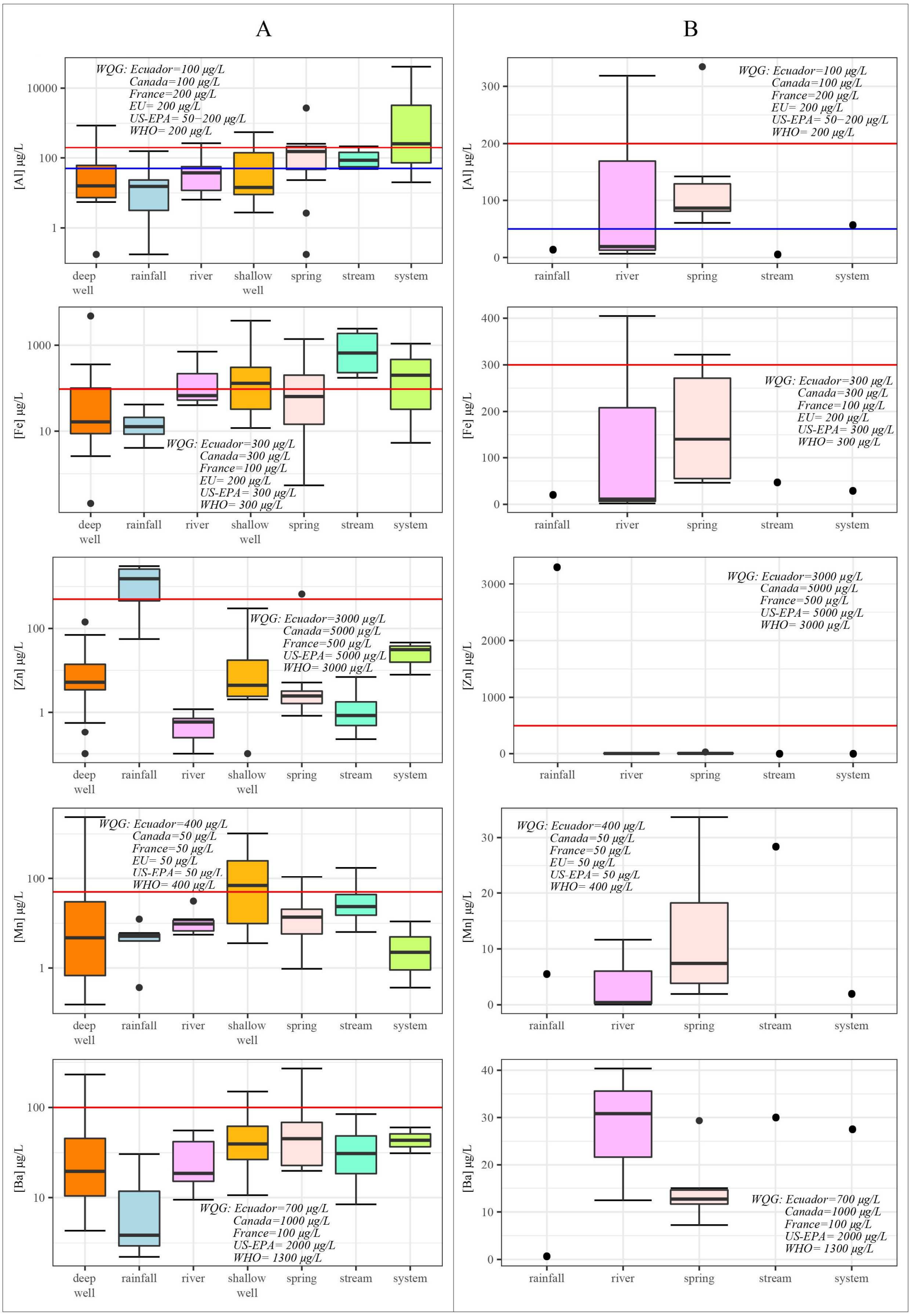

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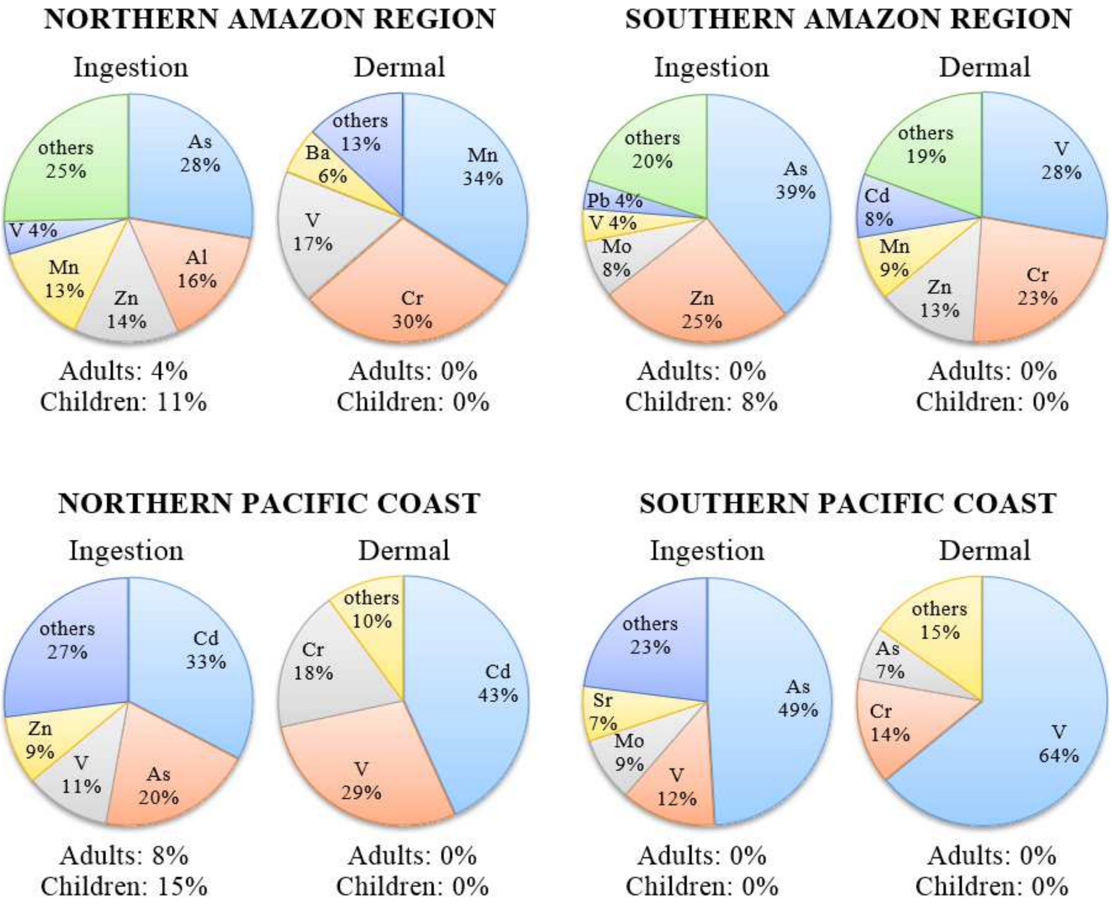

NORTHERN AMAZON REGION

SOUTHERN AMAZON REGION

NORTHERN PACIFIC COAST

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Start------Table 1

Table 1. Local metrics$^{\#,\#\#,b,c}$, inorganic major element ^{b} and metal(oid)s average concentrations (±Standard Deviation)^{d,e} in different sources of domestic water in the Northern and Southern Ecuadorian Amazon Region (NEAR and SEAR, respectively) and in the Northern and Southern Ecuadorian Pacific Coast (NEPC and SEPC, respectively).

PARAMETER	**NEAR**							**SEAR**					**NEPC**		**SEPC**	
	deep well	**rainfall**	**river**	**shallow well**	**spring**	**stream**	**system**	**rainfall**	**river**	**spring**	**stream**	**system**	**rainfall**	**system**	**system**	**shallow well**
Elevation$^{\#}$	281±17	276±18	264±10	264±6	283±12	293±36	266±6	321*	301±22	497±278	850*	303*	108*	32±25	65*	74*
T $^{\#\#}$	27±3	28±4	25*	26±1	26±2	27±1	29±3	24*	25±1	25±2	23*	25*	23*	27±2	29*	29*
EC ^{a}	272±349	12±13	82±91	156±59	60±67	86±66	224±152	5*	238±139	31±28	60*	49*	17*	144±63	210*	1319*
pH	6.11±1.03	4.84±1.02	6.12±2.06	5.61±0.42	5.2±1.43	7.02±1.38	7.16±0.51	7.11*	4.80±1.47	5.64±1.17	7.26*	6.69*	7.11*	7.01±0.19	6.82*	6.7*
OD ^{b}	3.25±2.42	4.89±2.34	5.37±1.99	3.33±2.68	5.24±1.9	5.97±2.78	5.96±1.76	2.56*	5.57±1.87	2.82±2.13	2.02*	2.05*	--	3.62±0.92	4.34*	3.12*
OD ^{c}	43.9±27	63±28	68.4±17.6	42.4±33.7	66.8±22.3	71.5±28.5	77.9±19.4	29.9*	66.6±22.6	33.6±24.8	23.8*	24.8*	--	46.9±10.9	57.5*	42.9*
$CaCO_3$ ^{b}	75.5±49.7	5.76±7.69	29.2±22.2	45.7±14.8	23.9±27.2	34.2±24.5	46.1±21.7	4.42*	47.2±29.3	13.9±13.2	28*	23.3*	2.14*	30.9±18.5	66*	239.2*
Bicarbonates^{b}	92.1±60.7	7.03±9.38	35.7±27.1	55.7±18.1	29.2±33.2	41.8±29.9	56.2±26.5	5.4*	57.6±35.8	16.9±16.1	34.2*	28.5*	2.61*	37.8±22.6	80.5*	291.8*
Fluorides^{b}	0.12±0.17	<0.02	0.04±0.04	0.03±0.02	0.03±0.04	0.06±0.04	0.07±0.04	<0.02*	0.08±0.02	0.04±0.03	0.07*	0.07*	<0.02*	0.08±0.03	0.17*	0.36*
Chlorides ^{b}	25.28±83.48	0.19±0.11	0.96±0.83	10.65±11.29	0.92±1.39	0.55±0.43	36.61±32.95	0.19*	1.64±1.74	0.09±0.03	0.65*	0.09*	3.18*	4.22±2.06	6.09*	18.6*
Sulfates ^{b}	1.13±1.26	0.62±0.33	1.88±3.06	2.5±2.67	0.46±0.75	0.36±0.11	2.56±3.32	0.11*	7.38±7.35	0.27±0.25	0.34*	0.19*	2.07*	20.99±6.12	14.76*	253.81*
Nitrates ^{b}	5.38±4.71	0.81±0.52	1.32±1.23	5.18±5.51	1.69±2.2	0.31±0.22	0.74±1.13	0.17*	0.13±0.19	0.35±0.19	0.16*	0.47*	0.06*	2.3±1.61	0.56*	22.17*
Ca ^{b}	15.27±12.22	1.28±2.81	6.1±4.34	10.55±3.79	4.53±5.8	5.71±4.52	10.30±4.69	0.06*	16.42±16.63	2.71±2.45	5.53*	3.94*	0.56*	12.02±7.76	21.19*	104.13*
K^{b}	1.99±1.73	0.18±0.11	1.41±0.96	1.77±1.48	0.86±0.81	2.17±0.43	1.93±0.1	<0.08*	0.79±0.07	0.58±0.33	1.34*	0.81*	0.5*	2.18±0.54	3.57*	7.6*
Mg ^{b}	6.04±9.23	<0.01	2.1±1.99	4.83±2.49	1.38±2.32	2.66±2.15	0.95±0.5	<0.01*	2.3±0.52	1.02±1.11	1.55*	1.76*	0.08*	2.87±0.84	5.61*	34.9*
Na ^{b}	24.05±35.88	0.12±0.05	3.25±2.08	8.75±7.81	3.09±2.86	4.07±2.07	31.68±26.85	<0.12*	3.68±1.17	1.54±1.33	3.27*	2.74*	0.64*	7.37±3.13	8.48*	68.79*
Si ^{b}	18.65±8.18	0.44±0.72	13.85±11.25	11.88±3.85	11.47±7.63	19.79±12.14	7.46±3.87	0.02*	8.45±4.53	6.19±4.66	10.69*	9.44*	0.10*	13.74±0.55	15.64*	39.73*
SiO_2^{b}	39.90±17.50	0.95±1.54	29.64±24.07	25.41±8.23	24.54±16.32	42.35±25.98	15.96±8.29	0.04*	18.07±9.7	13.23±9.98	22.87*	20.2*	0.21*	29.4±1.18	33.45*	85.01*
Hardness**	0.63±0.62	0.03±0.07	0.24±0.19	0.46±0.10	0.17±0.24	0.25±0.20	0.30±0.11	<0.01*	0.51±0.39	0.11±0.10	0.20*	0.17*	0.02*	0.42±0.19	0.76*	4.03*
TDS^{b}	211±155	8±12	82±58	125±33	64±62	100±64	157±94	6*	108±50	37±31	70*	59*	7*	119±43	174*	887*
Hg^{d}	2.1±3.5	1.9±1.1	1.7±0.7	0.8±0.7	2.6±2.5	1.5±1.5	8.0*	--	--	--	1.6*	--	--	--	--	--
Al^{e}	110±230	32±55	64±93	126±202	340±755	113±79	13693±23475	14*	115±177	132±103	5.43*	55*	129*	318±249	168*	6.93*
Ti^{e}	0.91±1.26	0.27±0.26	1.17±1.53	5.84±10.65	10.71±32.78	4.15±3.96	29.47±50.68	0.11*	0.32±0.35	1.11±0.56	0.03*	0.30*	0.23*	2.31±2.64	1.82*	<0.02
V^{e}	1.79±2.25	0.35±0.34	2.37±2.68	0.84±0.71	1.26±1.44	3.11±2.89	4.87±6.95	0.06*	2.06±0.4	0.9±0.95	0.49*	1.12*	19.8*	4.96±3.14	4.9*	9.13*
Cr^{e}	0.62±1.42	0.23±0.18	0.26±0.34	0.14±0.2	0.61±0.7	0.21±0.1	3.67±5.98	0.05*	0.32±0.21	0.18±0.1	0.06*	0.11*	2.14*	0.7±1.47	0.16*	0.46*
Mn^{e}	230.4±635.1	5.3±3.6	11.9±8.8	231.8±366.7	21.8±29.5	57.2±79.1	4.5±5.6	5.3*	4±6.6	12.5±12.5	28.3*	1.8*	6.7*	7±9.7	11*	0.7*
Fe^{e}	392±1223	17±13	208±258	669±1362	255±430	1146±1115	432±579	18*	139±230	165±127	47*	30*	69*	290±630	64*	10*
Co^{e}	0.23±0.5	0.04±0.04	0.11±0.13	0.6±0.53	0.32±0.31	0.38±0.32	0.09±0.09	0.03*	0.09±0.14	0.13±0.1	0.19*	0.03*	0.1*	0.1±0.09	0.06*	0.01*

Ni^{e}	0.57±0.93	0.26±0.1	0.18±0.13	0.38±0.54	0.77±0.6	0.54±0.68	0.82±0.56	0.78*	0.26±0.4	0.25±0.21	0.2*	0.18*	6.55*	0.92±1.04	0.57*	6.78*
Cu^{e}	7.76±12.63	1.52±1.62	0.36±0.29	8.87±15.31	1.07±1.28	0.71±0.86	6.82±9.15	0.61*	2.56±2.8	8.66±13.34	0.51*	2.01*	0.65*	3.9±2.86	15.42*	7.76*
Zn^{e}	20.3±38.1	1629±1249	0.5±0.4	49.9±111.5	57.5±190.9	2.2±3.2	28.5±19.3	3316.8*	3.5±1.2	8.5±11	0.2*	4*	999.3*	144.7±338	14.3*	25.5*
As^{e}	0.87±1.46	0.03±0.03	0.22±0.11	0.41±0.56	0.17±0.13	0.62±0.6	1.19±1.51	<0.02*	0.61±0.55	0.34±0.28	0.5*	0.85*	0.04*	0.51±0.3	1.03*	1.36*
Se^{e}	1.66±5.61	0.27±0.36	0.36±0.44	1.23±1.96	0.44±0.42	0.3±0.45	0.21±0.19	<0.06*	0.08±0.09	<0.06	0.07*	<0.06*	0.07*	0.21±0.14	0.31*	2.06*
Sr^{e}	206.6±197.1	3.8±5.3	70.9±52.8	125.6±62.5	73.7±96.4	75.5±50.3	67.6±27.4	0.3*	56.5±18.8	24.2±21.2	58.7*	26.4*	2.3*	80.4±33.8	126.6*	569.9*
Mo^{e}	2.59±5.48	0.03±0.03	0.55±1.04	0.07±0.05	0.07±0.19	0.19±0.13	0.32±0.39	<0.02*	5.5±9.42	0.02±0.02	0.12*	<0.02*	0.14*	1.25±1.08	0.27*	6.59*
Cd^{e}	0.036±0.046	0.022±0.014	0.019±0.023	0.085±0.078	0.042±0.035	0.040±0.064	0.015±0.012	0.036*	0.089±0.147	0.039±0.076	0.003*	0.001*	0.02*	1.381±4.711	0.006*	0.071*
Sb^{e}	0.024±0.029	0.087±0.097	0.013±0.009	0.028±0.028	<0.011	0.023±0.022	0.055±0.03	0.23*	0.028±0.026	0.017±0.013	0.033*	<0.011*	0.163*	0.279±0.312	0.152*	0.076*
Ba^{e}	54.7±74.2	9.4±10.2	28.4±18.3	54.1±46.6	62.7±68.4	39.3±32.9	44.9±14.6	0.5*	27.9±14.2	14.8±7.6	29.8*	27.5*	0.5*	23.9±7.9	33.1*	24.2*
Pb^{e}	0.509±0.639	1.169±1.994	0.084±0.15	1.536±2.796	0.247±0.293	0.129±0.117	0.948±0.846	0.151*	0.246±0.101	0.808±1.023	0.003*	0.090*	0.146*	0.453±0.684	0.768*	0.451*
U^{e}	0.071±0.116	0.004±0.009	0.027±0.021	0.022±0.019	0.068±0.077	0.069±0.089	0.396±0.639	0.001*	0.042±0.068	0.008±0.004	0.009*	0.004*	0.001*	0.052±0.092	0.083*	0.977*

NEAR=North Ecuadorian Amazon Region; SEAR= South Ecuadorian Amazon Region; NEPC= North Ecuadorian Pacific Coast; SEPC= South Ecuadorian Pacific Coast; #=masl (meters above sea level); ##=°C; * single value (n=1) ; **Ca+Mg (mmol.L^{-1}) ; a=µS.cm^{-1}; b= mg.L^{-1}; c=%saturation; d=ng.L^{-1} ; e= µg.L^{-1}.

End------Table 1

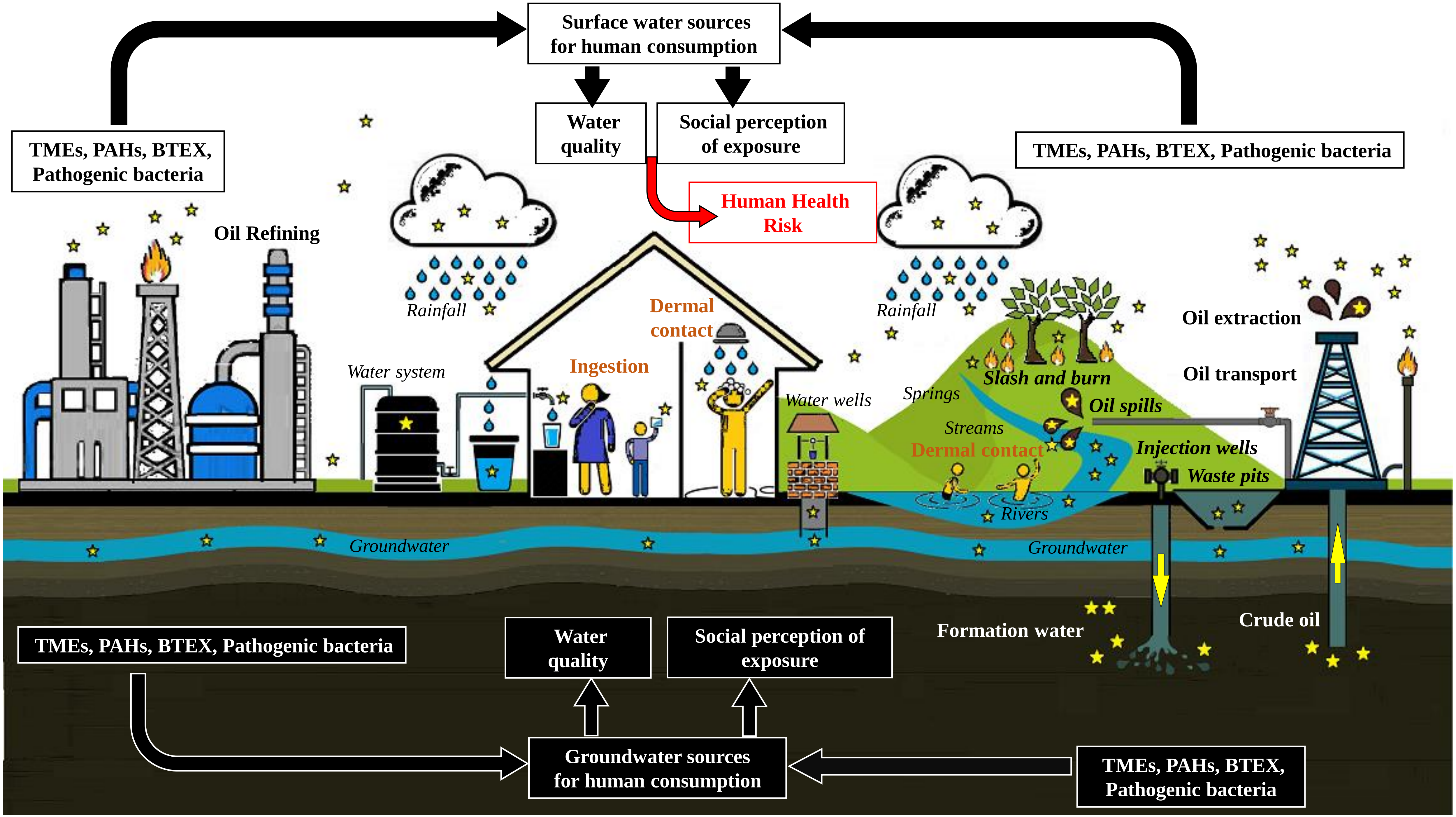
Start---Table 2

Table 2. Health Index for ingestion and dermal exposure pathways (HI*ing* and HI*der*), Total Hazard Index (THI), Cancer Risk for ingestion and dermal exposure pathways (CR*ing* and CR*der*), Total Cancer Risk (TCR), and percentage of samples (%) higher than the reference value for non-cancer risk (THI:1) and for cancer risk (TCR:10^{-6}-10^{-4}).

POPULATION	**RISK INDEX**	**NEAR**							**SEAR**					**NEPC**		**SEPC**	
		deep well	**rainfall**	**river**	**shallow well**	**spring**	**stream**	**system**	**rainfall**	**river**	**spring**	**stream**	**system**	**rainfall**	**system**	**system**	**shallow well**
ADULTS	**HI*ing***	$3.17x10^{-1}$	$2.88x10^{-1}$	$8.34x10^{-2}$	$2.29x10^{-1}$	$1.09x10^{-1}$	$1.69x10^{-1}$	$9.27x10^{-1}$	$5.27x10^{-1}$	$1.92x10^{-1}$	$1.01x10^{-1}$	$1.05x10^{-1}$	$1.51x10^{-1}$	$3.58x10^{-1}$	$3.45x10^{-1}$	$4.52x10^{-1}$	$2.67x10^{-1}$
	HI*der*	$2.16x10^{-3}$	$8.03x10^{-4}$	$7.23x10^{-4}$	$1.69x10^{-3}$	$9.79x10^{-4}$	$1.12x10^{-3}$	$3.77x10^{-3}$	$1.15x10^{-3}$	$8.05x10^{-4}$	$4.77x10^{-4}$	$3.94x10^{-4}$	$4.14x10^{-4}$	$5.06x10^{-3}$	$3.13x10^{-3}$	$2.13x10^{-3}$	$1.21x10^{-3}$
	THI	$3.20x10^{-1}$	$2.89x10^{-1}$	$8.41x10^{-2}$	$2.31x10^{-1}$	$1.10x10^{-1}$	$1.70x10^{-1}$	$9.30x10^{-1}$	$5.29x10^{-1}$	$1.93x10^{-1}$	$1.02x10^{-1}$	$1.06x10^{-1}$	$1.51x10^{-1}$	$3.63x10^{-1}$	$3.48x10^{-1}$	$4.54x10^{-1}$	$2.68x10^{-1}$
	> 1	7%	0%	0%	0%	0%	0%	33%	0%	0%	0%	0%	0%	0%	8%	0%	0%
	CR*ing*	$2.46x10^{-5}$	$9.53x10^{-7}$	$6.28x10^{-6}$	$1.17x10^{-5}$	$4.83x10^{-6}$	$1.77x10^{-5}$	$3.38x10^{-5}$	$2.35x10^{-7}$	$1.72x10^{-5}$	$9.71x10^{-6}$	$1.42x10^{-5}$	$2.42x10^{-5}$	$1.27x10^{-6}$	$1.45x10^{-5}$	$3.87x10^{-5}$	$2.93x10^{-5}$
	CR*der*	$6.52x10^{-9}$	$2.52x10^{-10}$	$1.66x10^{-9}$	$3.10x10^{-9}$	$1.33x10^{-9}$	$4.70x10^{-9}$	$8.95x10^{-9}$	$6.22x10^{-11}$	$4.56x10^{-9}$	$2.57x10^{-9}$	$3.76x10^{-9}$	$6.40x10^{-9}$	$3.35x10^{-10}$	$3.84x10^{-9}$	$1.02x10^{-8}$	$7.75x10^{-9}$
	TCR	$2.46x10^{-5}$	$9.53x10^{-7}$	$6.28x10^{-6}$	$1.17x10^{-5}$	$4.83x10^{-6}$	$1.77x10^{-5}$	$3.38x10^{-5}$	$2.35x10^{-7}$	$1.72x10^{-5}$	$9.71x10^{-6}$	$1.42x10^{-5}$	$2.42x10^{-5}$	$1.27x10^{-6}$	$1.45x10^{-5}$	$3.87x10^{-5}$	$2.93x10^{-5}$
	$>10^{-4}$	13%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
	10^{-6}-10^{-4}	73%	29%	100%	86%	100%	100%	100%	0%	100%	100%	100%	100%	100%	100%	100%	100%
	$<10^{-6}$	13%	71%	0%	14%	0%	0%	0%	100%	0%	0%	0%	0%	0%	0%	0%	0%
CHILDREN	**HI*ing***	$6.88x10^{-1}$	$6.24x10^{-1}$	$1.81x10^{-1}$	$4.97x10^{-1}$	$2.37x10^{-1}$	$3.65x10^{-1}$	$2.01x10^{-0}$	$1.14\ x10^{-0}$	$4.16x10^{-1}$	$2.19x10^{-1}$	$2.28x10^{-1}$	$3.27x10^{-1}$	$7.75x10^{-1}$	$7.47x10^{-1}$	$9.79x10^{-1}$	$5.78x10^{-1}$
	HI*der*	$3.75x10^{-4}$	$1.39x10^{-4}$	$1.25x10^{-4}$	$2.93x10^{-4}$	$1.70x10^{-4}$	$1.94x10^{-4}$	$6.53x10^{-4}$	$1.99x10^{-4}$	$1.40x10^{-4}$	$8.27x10^{-5}$	$6.82x10^{-5}$	$7.18x10^{-5}$	$8.78x10^{-4}$	$5.42x10^{-4}$	$3.69x10^{-4}$	$2.10x10^{-4}$
	THI	$6.88x10^{-1}$	$6.24x10^{-1}$	$1.81x10^{-1}$	$4.97x10^{-1}$	$2.37x10^{-1}$	$3.65x10^{-1}$	$2.01x10^{-0}$	$1.14x10^{-0}$	$4.16x10^{-1}$	$2.20x10^{-1}$	$2.28x10^{-1}$	$3.27x10^{-1}$	$7.76x10^{-1}$	$7.48x10^{-1}$	$9.79x10^{-1}$	$5.78x10^{-1}$
	> 1	27%	14%	0%	0%	0%	0%	33%	100%	0%	0%	0%	0%	0%	17%	0%	0%
	CR*ing*	$1.07x10^{-5}$	$4.13x10^{-7}$	$2.72x10^{-6}$	$5.08x10^{-6}$	$2.09x10^{-6}$	$7.69x10^{-6}$	$1.47x10^{-5}$	$1.02x10^{-7}$	$7.46x10^{-6}$	$4.21x10^{-6}$	$6.16x10^{-6}$	$1.05x10^{-5}$	$5.49x10^{-7}$	$6.28x10^{-6}$	$1.68x10^{-5}$	$1.27x10^{-5}$
	CR*der*	$2.82x10^{-9}$	$1.09x10^{-10}$	$7.19x10^{-10}$	$1.34x10^{-9}$	$5.53x10^{-10}$	$2.03x10^{-9}$	$3.87x10^{-9}$	$2.69x10^{-11}$	$1.97x10^{-9}$	$1.11x10^{-9}$	$1.63x10^{-9}$	$2.77x10^{-9}$	$1.45x10^{-10}$	$1.66x10^{-9}$	$4.42x10^{-9}$	$3.35x10^{-9}$
	TCR	$1.07x10^{-5}$	$4.13x10^{-7}$	$2.72x10^{-6}$	$5.08x10^{-6}$	$2.09x10^{-6}$	$7.69x10^{-6}$	$1.47x10^{-5}$	$1.02x10^{-7}$	$7.46x10^{-6}$	$4.21x10^{-6}$	$6.16x10^{-6}$	$1.05x10^{-5}$	$5.49x10^{-7}$	$6.28x10^{-6}$	$1.68x10^{-5}$	$1.27x10^{-5}$
	$>10^{-4}$	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
	10^{-6}-10^{-4}	53%	0%	100%	71%	75%	100%	100%	0%	100%	100%	100%	100%	0%	100%	100%	100%
	$<10^{-6}$	47%	100%	0%	29%	25%	0%	0%	100%	0%	0%	0%	0%	100%	0%	0%	0%

NEAR=North Ecuadorian Amazon Region; SEAR= South Ecuadorian Amazon Region; NEPC= North Ecuadorian Pacific Coast; SEPC= South Ecuadorian Pacific Coast; HI*ing*=hazard index for ingestion exposition; Hi*der*= hazard index for dermal exposition; THI=total hazard index.

End-----Table 2


Surface water sources for human consumption
Water quality
Social perception of exposure
Human Health Risk
TMEs, PAHs, BTEX, Pathogenic bacteria
TMEs, PAHs, BTEX, Pathogenic bacteria
Oil Refining
Rainfall
Rainfall
Dermal contact
Ingestion
Water system
Water wells
Springs
Slash and burn
Oil spills
Streams
Dermal contact
Oil extraction
Oil transport
Injection wells
Waste pits
Rivers
Groundwater
Groundwater
Formation water
Crude oil
TMEs, PAHs, BTEX, Pathogenic bacteria
Water quality
Social perception of exposure
Groundwater sources for human consumption
TMEs, PAHs, BTEX, Pathogenic bacteria