



# Co-occurrence, possible origin, and health-risk assessment of arsenic and fluoride in drinking water sources in Mexico: Geographical data visualization

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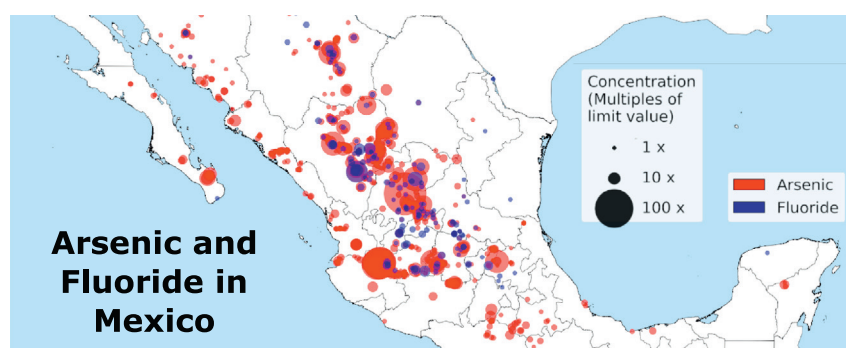
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## HIGHLIGHTS

- A groundwater arsenic-fluoride concentration map highlights enrichment zones.
- Volcanic glass is likely a primary source of arsenic-fluoride contaminated water.
- Evaporation in (semi)arid areas concentrates arsenic-fluoride in aquifers
- The states of Durango, San Luis Potosí, and Zacatecas have higher exposure risk

## GRAPHICAL ABSTRACT



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## ABSTRACT

Arsenic and fluoride in drinking water present a significant challenge to public health worldwide. In this study, we analyze the results of one of the largest surveys of drinking water quality in Mexico: 14,058 samples from 3951 sites, collected between January and December 2017. We use these data to identify the distribution and possible origin of arsenic and fluoride in drinking water throughout the country, and to estimate the associated health burden. The highest concentrations appear in alluvial aquifers in arid northern Mexico, where high-silica volcanic rock likely releases both arsenic and fluoride to the groundwater. We find fluoride contamination to be significantly correlated with aridity (Pearson correlation =  $-0.45$ ,  $p = 0.0105$ ), and also find a significant difference in fluoride concentrations between arid and humid states (Welch's  $t$ -test,  $p = 0.004$ ). We estimate population exposure by assigning to each town in Mexico the average concentration of any sampling sites within 5 km. Our results show that 56% of the Mexican population lives within 5 km of a sampling site, 3.05 million

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## 1. Introduction

Safe and readily available drinking water is an essential requirement for public health that concerns every country in the world (World Health Organization (WHO) and the United Nations Children's Fund (UNICEF), 2017). Arsenic (As) and fluoride (F) are contaminants present in many drinking water sources worldwide, and pose one of the greatest inorganic threats to public health (Kimambo et al., 2019). These elements, which generally occur naturally, are most often found in groundwater and therefore disproportionately affect countries that rely heavily on groundwater for irrigation and drinking.

Arsenic occurs in drinking water primarily as arsenate (As(V)), although in reducing environments significant concentrations of the more toxic form arsenite (As(III)) have also been reported. Other forms can also occur, among them organic arsenic and methylated arsenic (Gómez-Caminero et al., 2001; Limón-Pacheco et al., 2018). Inorganic As (arsenate plus arsenite) is the predominant form of arsenic in drinking water (Gómez-Caminero et al., 2001). World Health Organization (WHO) guidelines recommend that levels of As in drinking water should not exceed 10 µg/L (Fewtrell and Bartram, 2001). Exposure to 1500 µg/L of F reportedly causes discoloration of teeth, and exposure to 3000 µg/L may lead to fluorosis and more serious health problems (U.S. Public Health Service Recommendation for Fluoride Concentration in Drinking Water for the Prevention of Dental Caries, 2015).

The limit for F in drinking water that most countries have adopted is based on the WHO guideline of 1500 µg/L (World Health Organization (WHO), 2017). Health problems have been associated with chronic intake of F at this concentration, however, and thus many international organizations now recommend an even lower F limit of 700 µg/L (Amalraj and Pius, 2013). As and F co-occurrence in groundwater is generally associated with volcanic rocks (Ahmad and Bhattacharya, 2019; Kumar et al., 2016; Smedley and Kinniburgh, 2002). The health impacts of this co-occurrence are less well known, and may be different than the added effects of individual exposures to As and F.

### 1.1. Arsenic and fluoride exposure worldwide

Exposure to As in drinking water has been reported for decades in regions where As is a contaminant of concern. >300 million people in 105 countries are estimated to be chronically at risk from drinking water with As concentrations above the 10 µg/L WHO limit (Jiménez-Córdova et al., 2019; Limón-Pacheco et al., 2018; Shakoor et al., 2017). The highest exposures have been reported for India (West Bengal), Bangladesh, Nepal, Pakistan, China, Taiwan, Japan, Cambodia, Vietnam, Australia, Hungary, and Romania. In the Americas, As is mainly found in the arid and semi-arid regions of the United States, Mexico, Bolivia, Nicaragua, El Salvador, Peru, Chile, and Argentina (Chakraborti et al., 2016; Kimambo et al., 2019; Kumar et al., 2016; Shakoor et al., 2017). Exposure to F in drinking water has been reported in >25 countries. An estimated 200 million people rely on water sources with F above the WHO limit of 1500 µg/L (Kimambo et al., 2019).

Asian countries with high ground- and surface water F concentrations include India, China, Korea, Thailand, Sri Lanka, Indonesia, Yemen, Pakistan, Iraq, Turkey, Syria, Jordan, Palestine, Bangladesh, Iran, and Saudi Arabia (Biglari et al., 2016; Kimambo et al., 2019; Mumtaz et al., 2015). Though the origin of As and F in drinking water is mostly geological, there is some evidence that prolonged droughts

increase the concentration of these contaminants (Ali et al., 2019; Dehbandi et al., 2019; Podgorski et al., 2018; Reyes-Gómez et al., 2015). Some other human activities, notably mining (As) and the use of phosphate pesticides (F), can also contribute to contamination of aquifers and surface water (Navarro et al., 2017).

### 1.2. Arsenic and fluoride in Mexico

Groundwater is the source of an estimated 39% of drinking water in Mexico (Comisión Nacional del Agua (CONAGUA), 2017). In this country's arid regions, groundwater is the main (and often only) source of water for household use. As and F are contaminants of concern, as they have been reported in high concentrations in various Mexican aquifers (Alarcón-Herrera et al., 2013; Gutierrez et al., 2009; Kumar et al., 2016). Many studies have highlighted the public health challenge that As and F represent for central and northern Mexico (e.g., Chiprés et al., 2009; Mahlknecht et al., 2008; Reyes-Gómez et al., 2013). The geological sources of these contaminants have been given less research attention, in contrast, even though F-enriched mineral deposits have long been identified as part of the "Mexican Tin Belt" (Huspeni et al., 1984). The Mexican Tin Belt is a mineralized area on the eastern flank of the Sierra Madre Occidental (Fig. 2) that is mostly composed of rhyolites and ignimbrites, and which was created after a succession of ignimbrite flare-up episodes between 32 and 30 million years ago (Ferrari et al., 2002; Gómez-Tuena et al., 2007; Huspeni et al., 1984). This volcanism relates to the subduction and detachment of the Farallon Plate, which also resulted in the development of grabens and half-grabens to the east. Volcanic emissions, volcanic ash and volcanic glass from other sites (e.g., the Andes mountains, Mount Etna (Bhattacharya et al., 2016; Ferrari et al., 2002)) have been shown to contain large amounts of As and F (Alvarez and Carol, 2019; Kimambo et al., 2019; Mukherjee et al., 2014). Attempts to pinpoint geologic sources of As and F in Mexico include work by (Armienta and Segovia, 2008; Carrillo-Rivera et al., 2002; Reyes-Gómez et al., 2015).

It is estimated that at least 1.5 million people in Mexico consume water with As above 25 µg/L, and about 150,000 people are exposed to much higher concentrations still (75 to 530 µg/L; (Alfaro de la Torre et al., 2018)). Similarly, about 20 million people in the country drink water with F concentrations above 1500 µg/L. About 900,000 of those—mostly located in the states of San Luis Potosí, Durango, Zacatecas, Jalisco, Chihuahua, and Sonora—are exposed to even higher levels (4500 to 29,600 µg/L; (Alfaro de la Torre et al., 2018)). The magnitude of this problem is such that about 6.5 million children are believed to be exposed to concentrations of As or F high enough to cause health problems (Jiménez-Córdova et al., 2019; Limón-Pacheco et al., 2018).

The Mexican national limits for As and F in drinking water are 25 and 1500 µg/L, respectively. The federal guideline that sets this limit, NOM-127, was reviewed in 2000 (Secretaría de Salubridad y Asistencia, 2000). An update of this guideline is projected which will set the As limit to 10 µg/L and gradually decrease the limit for F until it reaches 1000 µg/L (Secretaría de Salubridad y Asistencia, 2000). Enactment of this new guideline will increase the difficulty of providing drinking water that meets the federal standard.

### 1.3. Health effects of arsenic and fluoride

The health effects of long-term exposure to arsenic have been widely documented; they include peripheral neuropathy,

gastrointestinal symptoms, diabetes, renal system effects, enlarged liver, bone marrow depression, high blood pressure, and cardiovascular disease (Alfaro de la Torre et al., 2018; Limón-Pacheco et al., 2018; World Health Organization (WHO), 2010). Inorganic arsenic is also one of the few substances that have been shown to cause cancer in humans through drinking water exposure. The International Agency for Research on Cancer (IARC) classifies arsenic in drinking water as a Group 1 human carcinogen (International Agency for Research on Cancer (IARC), 2004).

Chronic ingestion of fluoride at high doses causes dental, skeletal, reproductive, renal, neurological, gastrointestinal, and endocrine effects. High F increases the rates of dental and skeletal fluorosis, bone fractures, and kidney stones. It also decreases birth rates, thyroid function, and glucose tolerance (Mohammadi et al., 2017; Ozsvath, 2009; Verma et al., 2018). In addition, both a 2012 meta-analysis (Choi et al., 2012) and a 2018 dose-response meta-analysis (Duan et al., 2018) found that high water fluoride is significantly associated with lower IQ levels.

#### 1.4. Health effects of co-exposure

Little is known about the toxic effects caused by co-exposure to As and F (Jiménez-Córdova et al., 2019; Limón-Pacheco et al., 2018), though recent studies have explored their effects on immune cells in human populations with chronic exposure to both pollutants through drinking water (Ilizaliturri et al., 2009; Ortiz-Pérez et al., 2003). Exposure to either As or F has shown to reduce IQ levels and intellectual functionality among children (Limón-Pacheco et al., 2018; Ortiz-Pérez et al., 2003; Salgado-Bustamante et al., 2010; Wasserman et al., 2014). The potential role of F in health effects, previously attributed to As alone, should be systematically studied. In order for these studies to be carried out, the sources of both pollutants in drinking water must be better understood. Previous studies have found that about half (47%) of locations in Mexico where either As or F are present above guidelines (10 µg/L As, 1500 µg/L F) are simultaneously exposed to both contaminants (Limón-Pacheco et al., 2018). The aim of the present study is to spatially analyze one of the largest datasets yet collected of drinking water quality for the whole country. We focus on As and F, and seek to determine their distribution, degree of co-occurrence, possible origin, and the magnitude of the population exposed to them.

## 2. Methods

The present study is based on water monitoring data reported by the Mexican National Water Commission (CONAGUA) in 2017, concerning groundwater and other sources of drinking water in Mexico (Comisión Nacional del Agua (CONAGUA), n.d.). The original CONAGUA dataset contained water quality information for 14,058 samples from 3951 sites throughout the country, collected between January and December of 2017. For our analysis of the population exposed to these contaminants, we used a list of towns, settlements, and cities (collectively, towns) published by the National Institute for Geography and Informatics (INEGI) in 2010 and made accessible by the Mexican National Commission for the Knowledge and Use of Biodiversity (CONABIO) (Instituto Nacional de Estadística Geografía e Informática (INEGI), 2010).

We cross-referenced INEGI town data with CONAGUA water quality data. Our analysis considers only towns within 5 km of a sampling site from the CONAGUA dataset. The population in these towns adds to 69 million people, or 59% of the total population of Mexico (2010 population data). We assumed that the population living in these towns is exposed to the average contaminant levels of the sampling sites within 5 km. We calculated daily doses of contaminant exposure using the following standard formula (US Environmental Protection Agency,

2010):

$$\text{daily dose} \left[ \frac{\text{mg}}{\text{kg} \cdot \text{day}} \right] = \text{concentration} \left[ \frac{\text{mg}}{\text{L}} \right] * \frac{\text{consumption} \left[ \frac{\text{L}}{\text{day}} \right]}{\text{body weight} [\text{kg}]}$$

Arsenic exposure is carcinogenic. For a person that drinks contaminated water every day, the associated lifetime cancer risk is proportional to their daily dose of As exposure. This daily dose depends on three factors: As concentration in drinking water, daily water intake, and the person's body mass. In order to estimate the daily doses for the population of each town, we assumed that each town has the same age distribution as the whole country (71% adults, 29% children) (United Nations Department of Economic and Social Affairs, 2018) and we also assumed common reference values for body mass (70 kg for adults, 25 kg for children) and water intake (2 L/day for adults, 1 L/day for children). Each person's additional lifetime cancer risk is linearly related to their daily exposure through the cancer slope factor (CSF):

$$\text{individual cancer risk} \left[ \frac{1}{\text{person}} \right] = \text{daily dose} \left[ \frac{\text{mg}}{\text{kg} \cdot \text{day}} \right] * \text{CSF} \left[ \frac{\text{kg} \cdot \text{day}}{\text{mg}} \right]$$

We used a CSF value of 1.5 (kg · day)/mg in order to estimate the additional cases of cancer to be expected in each town from a lifetime of As exposure (Donohue and Lipscomb, 2002; US Environmental Protection Agency, 2010). For cancer estimations, we assumed that the CSF applies to all people exposed to As above the limit of 10 µg/L.

These data were processed using several open-source data analysis tools based on the Python programming language. Data analysis was carried out using pandas (Python Data Analysis Library (pandas), n.d.) and geopandas (GeoPandas, n.d.), with plotting enabled by matplotlib (matplotlib, n.d.) and task codification using Luigi (Luigi, n.d.).

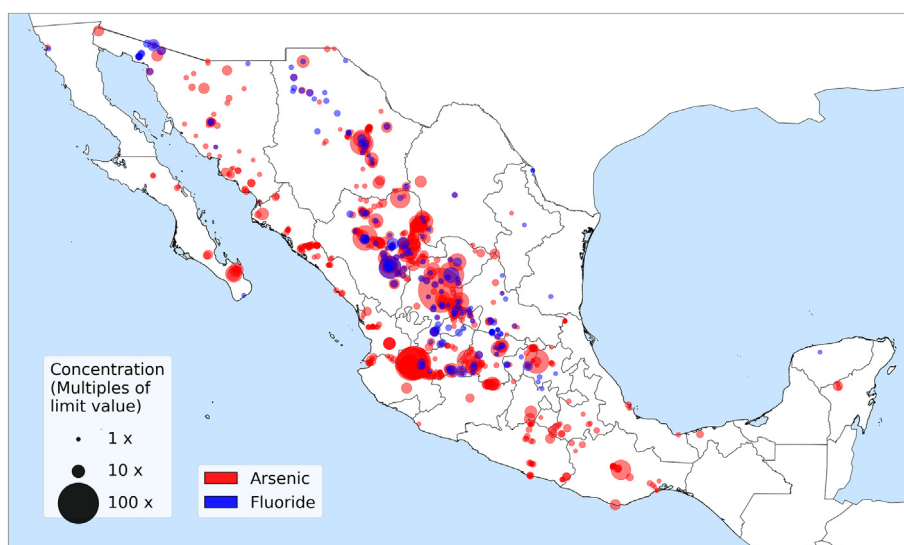
## 3. Results

### 3.1. Co-occurrence of arsenic and fluoride

As and F co-occur over the established limits throughout the country, and particularly in aquifers along a northwest-southeast axis in the arid and semiarid plateaus of north-central Mexico (Fig. 1).

Alluvial aquifers in these areas are the main source of drinking and irrigation water, and are located in the vicinity to the fluoride-rich "Mexican Tin Belt" (Huspeni et al., 1984) (Fig. 2).

In agreement with (Mukherjee et al., 2014), who report an association between arsenic and sedimentary basins adjoining major orogenic belts, As is found enriched in alluvial aquifers adjoining the Sierra Madre Occidental. The sediment in these aquifers is comprised of the erosional fragments of the surrounding rocks. Although outcrops contain all three types of rocks—igneous, metamorphic and sedimentary—, volcanic rocks dominate (e.g., rhyolitic ignimbrites) (Ferrari et al., 2007; Reyes-Gómez et al., 2013). These volcanic rocks were emplaced during a volcanic event that formed the Sierra Madre Occidental, a mountain range massive enough to be considered the world's largest continuous rhyolitic province. Ignimbrites and rhyolites are volcanic rocks that form after fast cooling of high silica magma, and which may contain a large amount of volcanic glass (Ferrari et al., 2007). Trace elements present in the magma, including As and F, become trapped in the glass during solidification (Bhattacharya et al., 2006; Nicolli et al., 2010; Ruggieri et al., 2011). Nicolli et al. (2010) report a strong association between As and F in volcanic glass as well as highly variable content, with median values of 6 and 722 mg/kg, respectively. (Ruggieri et al., 2011) report As and water-leached F content in volcanic ash of a rhyolitic magma to be 38 and 220 mg/kg, and both of these elements to be highly mobile.



**Fig. 1.** Co-occurrence of arsenic (red) and fluoride (blue) in Mexico. Marker size is proportional to the number of times by which that site exceeds the safe limit. Only sites at or above the limit are shown (10 µg/L for As; 1500 µg/L for F). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

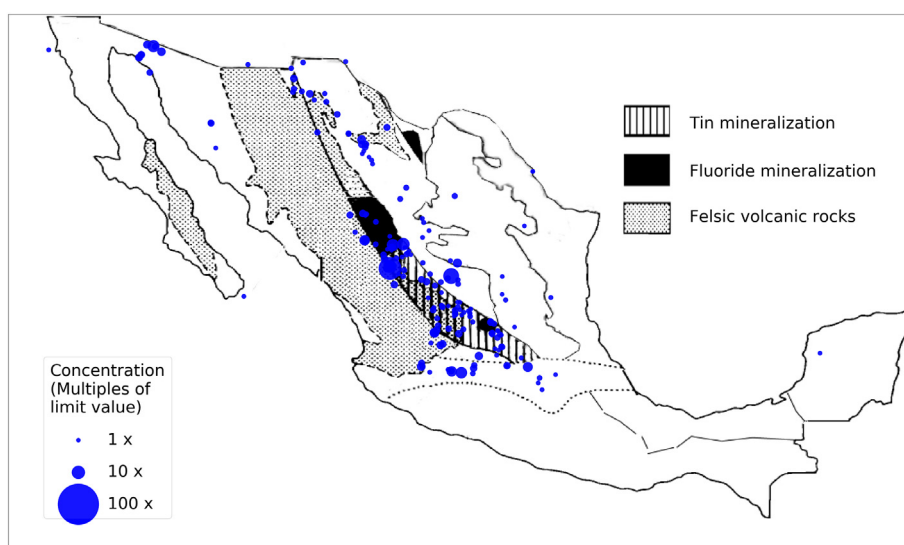
As and F are released upon weathering of the rock (Nicolli et al., 2010). Dissolution of amorphous silica in volcanic glass is slow, but proceeds under alkaline conditions and is enhanced by the presence of  $\text{Na}^+$  (or  $\text{K}^+$ ) (Alvarez and Carol, 2019; Crundwell, 2017). Released As quickly adsorbs onto Fe- and Mn oxyhydroxides, clays, and organic matter and becomes part of the solid fraction again, where it can remain stable for a long time (especially if adsorbed to iron oxyhydroxides). These adsorption-desorption reactions are strongly affected by pH; under alkaline conditions, As desorbs from Fe oxyhydroxides and other sorbents, and moves back into the water (Alvarez-Ayuso et al., 2008; Alvarez and Carol, 2019; Nicolli et al., 2010).

Groundwater in the arid regions is predominantly oxidizing with alkaline pH, a characteristic that favors both the dissolution of the silica in volcanic glass and the desorption of the elements retained in the iron oxyhydroxides, with desorption rates being considerably higher than silica dissolution (Alvarez and Carol, 2019). High evaporation, characteristic of the arid conditions of the eastern side of the Sierra Madre Occidental, further concentrates As and F in the shallower parts of the aquifers (Nicolli et al., 2010). By contrast, the western flank of the Sierra Madre Occidental receives more precipitation and has a steeper drop in

topography to reach sea level, which deters the As—F concentration effect.

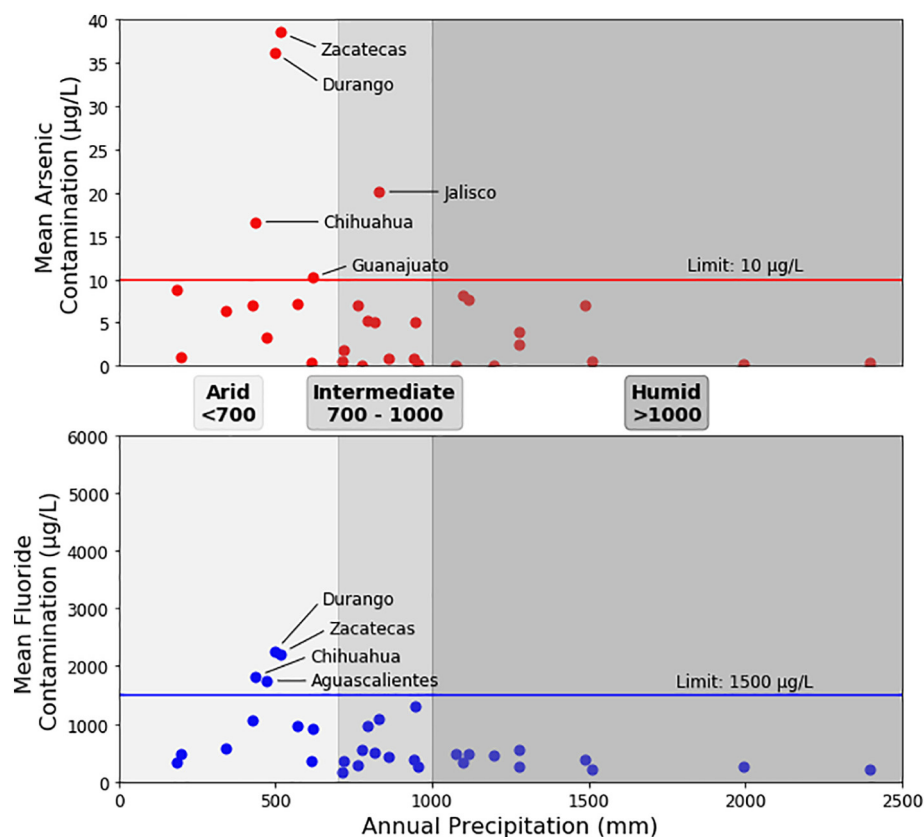
Although As contamination is much more widespread than F (Fig. 1), the highest As values are found in the regions where they co-occur. Of the 3951 total CONAGUA sites, 639 (16.2%) had As levels above the limit of 10 µg/L. Contaminated sites included groundwater (326; 51%), rivers (179; 28%), and other surface water sites (134; 21%). F, meanwhile, is mostly concentrated in the plateaus in the north-central part of the country (Fig. 3). Of the 3951 total CONAGUA sites, 184 (3.8%) had F levels above the limit of 1500 µg/L. F was only found in groundwater samples.

We expected that the higher evaporation rates associated with arid climate would be an important enrichment factor for As and F. Thus, we classified the Mexican states into three groups (arid, intermediate, and humid) and applied Welch's *t*-test to measure whether states with different degrees of humidity had significantly different As and F concentrations (we considered differences to be significant when  $p < 0.008333$ ; this threshold corresponds to a significance of 0.05, Bonferroni-corrected for testing 6 hypothesis). The results, shown in Table 1, show a significant difference in only one comparison: fluoride



**Fig. 2.** Extent of volcanic rock and locations of sampling sites above the limit (1500 µg/L) for fluoride. (Modified after (Huspeni et al., 1984).





**Fig. 3.** Relationship between annual precipitation and groundwater contamination with arsenic (top) and fluoride (bottom) in Mexican states. Precipitation correlates inversely with contamination.

concentrations between arid and humid states ( $p = 0.004$ ). That difference is relevant to this discussion because arid regions depend more on groundwater for drinking and irrigation, and are therefore at higher risk for exposure to fluoride.

The inverse correlation between aridity and As/F contamination can also be observed in a scatterplot of these contaminants against precipitation, Fig. 3. With a significance threshold of  $p = 0.05$  (Bonferroni-corrected to  $p = 0.025$  for two hypotheses), the Pearson correlation coefficient (PCC) between contaminant and annual precipitation is negative for both substances and significant for fluoride (PCC =  $-0.45$ ,  $p = 0.0105$ ), though not for arsenic (PCC =  $-0.34$ ,  $p = 0.0570$ ).

### 3.2. Cancer incidence related to arsenic

Arsenic distribution, the affected population, and the number of cancer cases that would result from a lifetime of exposure to this contaminant, are shown in Fig. 4 and Table 2. It is interesting to note from Fig. 4

that regions with high concentration do not necessarily correspond to a higher exposure and vice versa. Altogether, about 8.81 million people in 7263 towns live within 5 km of a CONAGUA sampling site with  $>10 \mu\text{g/L}$  of As, and an additional 13,070 lifetime cases of cancer are expected from this exposure. This estimate only accounts for the 3951 sites in the CONAGUA dataset, which is an underestimation of the real burden of exposure and disease for the country as a whole.

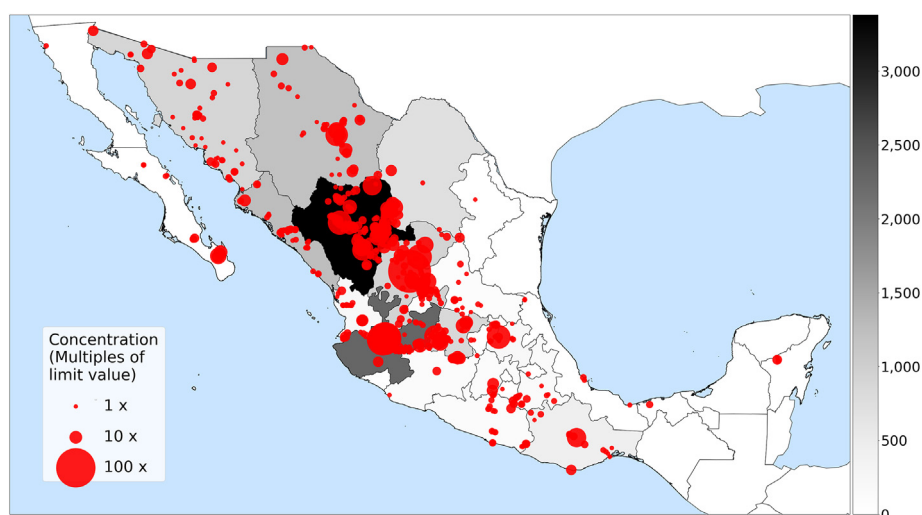
### 3.3. Population exposed to excess fluoride

We repeated the analysis above for F, adding data from all the towns within 5 km of a CONAGUA sampling site and averaging the F concentrations at those sites. We calculated how many people are exposed to a daily dosage above the reference value of  $0.06 \text{ mg}/(\text{kg} \cdot \text{day})$ , considered to be the limit above which significant health effects begin (Fig. 5, Table 3). About 3.05 million people in 2726 towns live within 5 km of a

**Table 1**

Average arsenic and fluoride concentrations of Mexican states, grouped according to their degree of aridity. Values for arsenic and fluoride are mean and (in parenthesis) standard deviation.

| Category  | Arsenic, $\mu\text{g/L}$ | Fluoride, $\mu\text{g/L}$ | States   |
|---|--------------------------|---------------------------|--|
| Arid<br>Precipitation $<700 \text{ mm/yr}$<br>Number of sites: 1341                       | 12.32<br>(13.17)         | 1160<br>(726)             | Aguascalientes, Baja California, Baja California Sur, Coahuila, Chihuahua, Durango, Guanajuato, Nuevo León, Querétaro, Sonora, Zacatecas |
| Intermediate<br>Precipitation = $700\text{--}1000 \text{ mm/yr}$<br>Number of sites: 1437 | 4.25<br>(5.82)           | 573<br>(369)              | Colima, Ciudad de México, Hidalgo, Jalisco, México, Michoacán, Morelos, San Luis Potosí, Sinaloa, Tamaulipas, Tlaxcala                   |
| Humid<br>Precipitation $>1000 \text{ mm}$<br>Number of sites: 1173                        | 3.02<br>(3.42)           | 361<br>(127)              | Campeche, Chiapas, Guerrero, Nayarit, Oaxaca, Puebla, Quintana Roo, Tabasco, Veracruz, Yucatán   |



**Fig. 4.** Arsenic contamination and associated cancer burden. States are colored gray according to the number of additional lifetime cancer cases predicted from exposure to arsenic above 10 µg/L. Red circles denote sampling sites with arsenic concentrations above this limit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sampling site that contained >1500 µg/L of F, and about 2.07 million people have an estimated exposure to F above 0.06 mg/(kg\*day).

#### 4. Discussion

The provision of safe drinking water is one of the most pressing challenges that Mexico must overcome to meet the 2030 Agenda for Sustainable Development, adopted by all United Nations Member States in 2015. An earlier work had found As and F in 170 municipalities in 23 of the 32 states in Mexico (Comisión de Hábitat Medio Ambiente y Sostenibilidad (CHMAS), 2018); the states with the greatest number of affected municipalities were Chihuahua (28), Zacatecas (27), Durango (25), San Luis Potosí (17), Jalisco (15) and Sonora (9). In total, 71.2% of the municipalities in the country were found to be affected by these contaminants.

The present study reinforces those earlier findings, and allows us to visually identify the areas around the country that are most affected. Our results match earlier estimates of the burden of disease associated with As in drinking water. Inorganic As is also present in food, which is believed to be responsible for 9129 to 119,176 additional cases of bladder cancer; 11,844 to 121,442 cases of lung cancer; and 10,729 to 110,015 cases of skin cancer worldwide (Limón-Pacheco et al., 2018; Oberoi et al., 2014).

Dependence on groundwater in these areas is expected to escalate in the near future due to increasing water demand, aquifer depletion, and pollution of surface water sources. Alleviating exposure to As and F requires treatment of both drinking water and the water used to irrigate food crops (Bhowmick et al., 2018). Contaminant removal systems can

be either centralized or domestic, though centralized systems are the ideal option because it is easier to monitor their functionality and properly dispose of the contaminated waste they generate. Technologies for As and F removal include oxidation, coagulation-precipitation, electrocoagulation, adsorption, ion exchange, and membrane-based techniques such as reverse osmosis (Bazrafshan and Ownagh, 2012; Lourdes et al., 2000; Shams et al., 2010).

Education and community engagement are key factors for ensuring successful interventions. There is a need for community members to understand the risks of high As and F exposure, as well as the sources of contamination. It is also important to know the As contents in irrigation water used on crops such as rice, which can uptake and translocate As to the edible parts (Althobiti et al., 2018; Santra et al., 2013). Despite the challenges inherent to drinking water sources with variable content of toxic As and F, exposure to these elements has been significantly reduced through mitigation measures such as accessing alternative water sources and using centralized and non-centralized water treatment processes (e.g. reverse osmosis, nanofiltration, etc.) (Kimambo et al., 2019; Walker et al., 2008; World Health Organization (WHO), 2017).

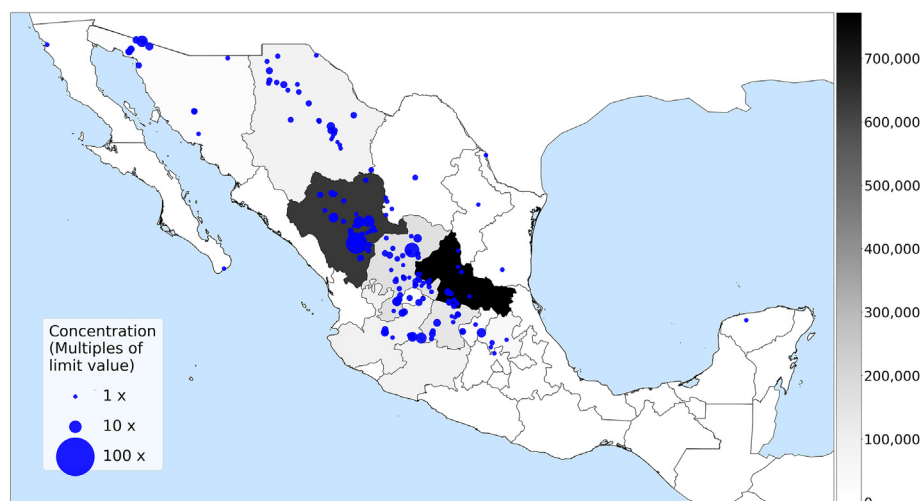
#### 5. Conclusion

As and F are major contaminants of drinking water that represent a significant challenge to public health worldwide. In this study, we analyzed the results of one of the largest surveys of drinking water contamination in Mexico. We used measurements from 3951 sites around the country to plot the geographic distribution of F and As, and to estimate the population exposed to unhealthy levels of these substances. Our results confirm earlier surveys of this problem, highlighting the burden on the north-central arid states of Durango, San Luis Potosí, Chihuahua, Zacatecas, and Jalisco. Fluoride contamination is significantly correlated with aridity (Pearson correlation =  $-0.45$ ,  $p = 0.0105$ ), and there is a significant difference in fluoride concentrations between arid and humid states (Welch's  $t$ -test,  $p = 0.004$ ).

About 56% of the country's population lives in a town within 5 km of one of our sampling sites (66 million people in 2010). From this total population, we estimated that 3.05 million people are exposed to F above the reference limit of 0.06 mg/(kg \* day), 8.81 million people are exposed to >10 µg/L of As, and an additional 13,070 lifetime cases of cancer are expected from this As exposure. These numbers are an underestimate of the country's overall burden of disease, and don't account for the synergistic health effects of exposure to both

**Table 2**  
States ranked by their arsenic-associated lifetime cancer burden.

| Rank | State      | Population exposed to As above 10 µg/L | Additional lifetime cancer cases caused by As |
|------|------------|--|---|
| 1    | Durango    | 1,174,741                              | 3381  |
| 2    | Jalisco    | 619,058                                | 2283  |
| 3    | Sinaloa    | 1,166,453                              | 1247  |
| 4    | Chihuahua  | 1,626,153                              | 1198  |
| 5    | Zacatecas  | 391,704                                | 912   |
| 6    | Sonora     | 1,069,452                              | 894   |
| 7    | Guanajuato | 618,884                                | 854   |
| 8    | Coahuila   | 712,095                                | 722   |
| 9    | Oaxaca     | 523,865                                | 444   |
| 10   | Hidalgo    | 32,923                                 | 204   |



**Fig. 5.** Fluoride contamination. States are colored according to the number of people exposed to fluoride above the safe daily dosage of 0.06 mg/(kg \* day). Blue circles denote sampling sites with fluoride concentrations above the limit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Table 3**

States ranked by their population exposed to excessive fluoride.

| Rank | State           | Population exposed to F above 0.06 mg/(kg * day) |
|------|-----------------|--|
| 1    | San Luis Potosí | 772,124  |
| 2    | Durango         | 634,753  |
| 3    | Zacatecas       | 165,624  |
| 4    | Guanajuato      | 133,703  |
| 5    | Jalisco         | 91,556   |
| 6    | Michoacán       | 86,878   |
| 7    | Chihuahua       | 81,933   |
| 8    | Hidalgo         | 45,725   |
| 9    | Sonora          | 22,428   |
| 10   | Querétaro       | 18,861   |

contaminants. Nevertheless, they show the magnitude of the public health problem presented by As and F in Mexico.

### CRediT authorship contribution statement

**M.T. Alarcón Herrera:** Conceptualization, Investigation, Writing - review & editing, Supervision, Project administration. **Daniel A. Martin-Alarcon:** Conceptualization, Methodology, Software, Validation, Formal analysis, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Mélida Gutiérrez:** Conceptualization, Formal analysis, Writing - original draft, Writing - review & editing, Visualization. **Liliana Reynoso Cuevas:** Writing - original draft. **Alejandra Martín:** Writing - original draft. **Mario A. Olmos Márquez:** Writing - original draft. **Jochen Bundschuh:** Writing - review & editing.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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