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ORIGINAL PAPER

Assessment of natural arsenic in groundwater in Cordoba Province, Argentina

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Abstract Groundwater in the central part of Argentina contains arsenic concentrations that, in most cases, exceed the value suggested by international regulations. In this region, Quaternary loessical sediments with a very high volcanic glass fraction lixiviate arsenic and fluoride after weathering. The objectives of this study are to analyze the spatial distribution of arsenic in different hydrogeological regions, to define the naturally expected concentration in an aquifer by means of hydrogeochemistry studies, and to identify emergent health evidences related to cancer mortality in the study area. The correlation between arsenic and fluoride concentrations in groundwater is analyzed at each county in the Cordoba Province. Two dimensionless geoindicators are proposed to identify risk zones and to rapidly visualize the groundwater quality related to the presence of arsenic and fluoride. A surface-mapping system is used to identify the spatial variability of concentrations and for suggesting geoindicators. The results show that the Chaco-Pampean plain hydrogeologic region is the most affected area, with arsenic and fluoride concentrations in groundwater being generally higher than the values suggested by the World Health Organization (WHO) for drinking water. Mortality related to kidney, lung, liver, and skin cancer in this area could be associated to the ingestion of arsenic-contaminated water. Generated maps provide a base for the assessment of the risk associated to the natural occurrence of arsenic and fluoride in the region.

Keywords Arsenic · Cancer · Correlation · Fluoride · Groundwater · Loess · Sampling well · Spatial variability

Introduction

Arsenic (As) is a natural chemical element sometimes found in soils, rocks, and surface and subsurface waters. The mean As concentration in the Earth is in the 1.5–2.0 mg/kg range. Some particular rocks and sediments contain very high contents of As. Groundwater, frequently used as a source of fresh water, becomes contaminated when As molecules migrate from the solid matrix of porous media to the pore fluid.

Groundwater chemistry reveals different possible interaction mechanisms with the environment (e.g., air, soil, etc.), human activities (e.g., land use, industrial activity, etc.), and different contamination sources. The groundwater chemical composition

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restricts any of its potential uses and may influence the ecosystem.

Inorganic arsenic is a documented human carcinogen substance (Bergoglio 1963; Wu et al. 1989). Several epidemiological studies relate the ingestion of arsenic-contaminated water with several health problems, including different cancer types and skin alterations, such as hyperkeratosis and hyperpigmentation (Smith et al. 1992; Hopenhayn-Rich et al. 1998; Abernathy et al. 1999).

Argentina is one of the countries that have an important area with natural arsenic in groundwater (Nicolli et al. 1989; Smedley et al. 2005). The origin of arsenic is related to the mineralogical composition of loessical Quaternary sediments that contain significant volcanic glass fractions (Teruggi 1957; Karlsson 1993; Smedley et al. 2005). These aeolian sediments cover more than 600,000 km^2 of the central part of Argentina (Francisca 2007).

In contact with water, the volcanic glass fraction of loess lixiviates arsenic, fluoride (F), vanadium, and several other chemical elements that, at given concentration levels, can affect human health (Nicolli et al. 1989; Guo and Tseng 2000; Bates et al. 2004).

In Cordoba Province, located at the center of Argentina, the groundwater is frequently used as a source of drinking water in rural areas and for agriculture and industrial activities. However, the groundwater usually contains high salt content, arsenic, fluoride, vanadium, molybdenum, and other trace elements that restrict the use of this natural resource (Nicolli et al. 1989).

The use of recently developed geoindicators allows us to evaluate environmental processes and to monitor specific earth and environmental properties (Rapport and Friend 1979). The groundwater quality is considered as a geoindicator by means of the measurement and analysis of its properties (Berger 1997). The measure of different groundwater attributes (such as salinity, redox potential, chemical concentrations, radioactivity, and pollution) (Berger 1997) can be used for understanding environmental changes, to define the background concentrations in geochemical analysis, and to identify risk areas (Giedraitiene et al. 2002). These environmental indicators are very helpful in providing information about natural resources and evaluating the intensity and the direction of possible geoenvironmental changes (e.g., to distinguish natural from anthropogenic sources of contamination).

In this work, several groundwater quality maps are developed. The spatial distribution of arsenic and fluoride concentrations in groundwater is determined from these new maps and two proposed geoindicators. The mean and variance of As and F concentrations are correlated to each other and with cancer occurrence for each county in the Cordoba Province. The purpose of the work is to show that mortality associated to kidney, lung, liver, and skin cancer depends on the ingestion of arsenic-contaminated water. The relevance of the spatial distribution of the groundwater quality determined by the natural occurrence of arsenic and fluoride on health risks is analyzed. The results show positive correlations between epidemiological evidences and the rural population fraction that use groundwater as a source of drinking water.

Sediments and hydrogeochemistry

The study area covers 165,321 km^2 and comprises Cordoba Province in the center of Argentina (Fig. 1). Quaternary loessical soil deposits, known as Pampean Loess, cover 600,000 km^2 of northeastern Argentina. Very fine sand, silt, and clay particles lifted by the wind and transported by aeolian action are the main fractions of these sediments (Sayago et al. 2001; Francisca 2007). The origin of loess in this area is associated to the volcanic activity of the Andean Cordillera (Tricart 1969; Kröhling 1999). The thicknesses of these loessical sediments usually range from a few meters up to a maximum of 65 m (Teruggi 1957). The loess spatial distribution is related to its origin and a multistage transport mechanism involving fluvial, glacial, nival, and aeolian processes (Zárate 2003).

Table 1 shows the principal physical properties of Argentinean loess, according to Teruggi (1957), Francisca et al. (2002), Rocca et al. (2006), and Francisca (2007). Normally, the silt fraction prevails over the clay and sand fractions. There is a very high content of volcanic glass in the sand and silt fractions of the studied loess, in addition to quartz, feldspar, and biotite (some other minerals are frequently encountered in fractions of less than 1%). The clay fraction contains mainly montmorillonite and some illite, quartz, and feldspar (Teruggi 1957). The montmorillonite fraction comes from the weathering of volcanic glass.

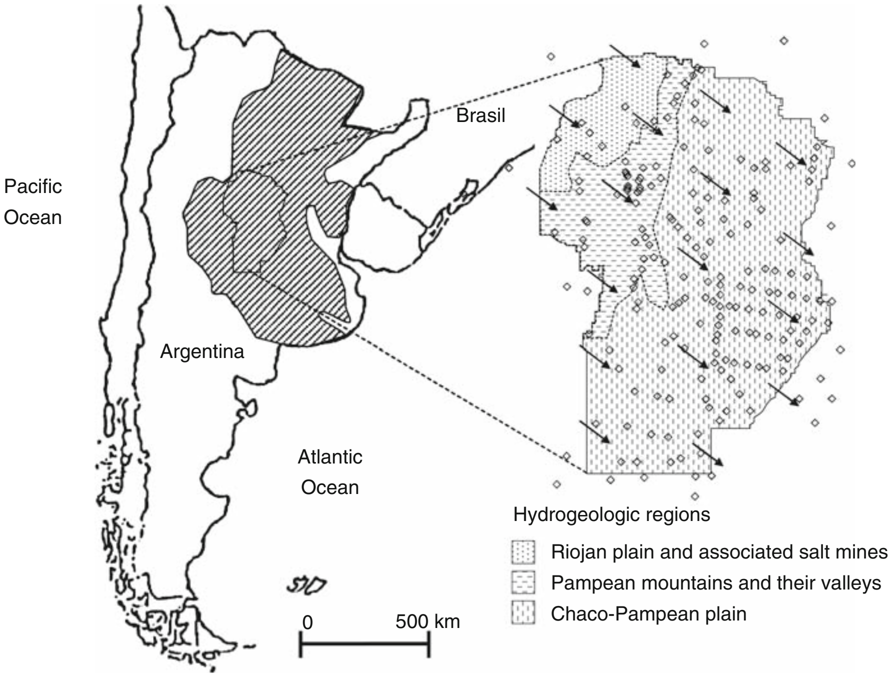

Fig. 1 Geographic distribution of the Pampean Loess in Argentina (modified from Francisca 2007). The *arrows* indicate groundwater flow direction and the *diamonds* represent sampling wells

Table 1 Principal physical properties of the Pampean Loess in Argentina (data compiled from Teruggi 1957; Francisca et al. 2002; Rocca et al. 2006; Francisca 2007)

Property	Unit	Typical value
Dry unit weight	kN/m^3	12.2–14.5
Wet unit weight	kN/m^3	14.9–16.8
Liquid limit (LL)	%	20.8–32.2
Plastic index (PI)	%	0–8.0
Sand fraction	%	5–15
Silt fraction	%	40–75
Clay fraction	%	7–25
Volcanic glass (in sand and silt fraction)	%	6–50.5
pH (at 2.5 water to soil particle ratio)	–	8.5–9.63
Color	–	Light yellow

Figure 2 shows a picture of the volcanic glass fraction frequently encountered in the loessical sediments in the study area. The volcanic glass content may vary significantly, depending mainly on location and past transport mechanisms. Typically, loess deposits close to the mountains in the western part of Cordoba Province have volcanic glass content lower than 4%, while in the pampas, it can usually reach values between 14.3 and 38.7% (Karlsson 1993).

Shallow aquifers, encountered at a few meters bellow the surface, are unconfined in the entire study area (usually from 0.5 to 20 m below the surface). The groundwater flow direction is from W–NW to E–SE according to the potentiometric surface (Fig. 1). Deeper aquifers are usually semiconfined and interconnected with each other, forming a multilayer system. The permeability, porosity, and mineralogy

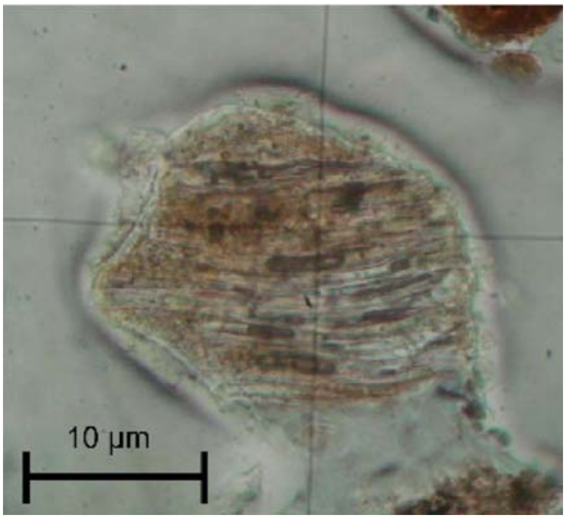

Fig. 2 Picture of the volcanic glass typically encountered in Argentinean loess

of soils vary laterally and in depth, affecting the groundwater chemical properties. Three clear hydrogeologic regions were identified from the analysis of groundwater in the study area (Santa Cruz and Silva Busso 1999): (a) Riojan plain and associated salt mines, (b) Pampean mountains and their valleys, and (c) Chaco-Pampean plain. Figure 1 shows the extension of these hydrogeologic regions.

The presence of arsenic in groundwater is associated to the mineralogy and sediment texture (Nicolli et al. 1989; Smedley and Kinniburgh 2002; Paoloni et al. 2005). Arsenic and fluoride naturally appear from the dissolution and weathering of the volcanic glass fraction of loess (García et al. 2006). Arsenic concentrations in the solid matrix of Argentinean loess fluctuate between 4.3 and 8.3 mg/kg for the sand and silt fraction, while the clay fraction usually contains concentrations from 8.9 to 29.8 mg/kg (Smedley et al. 2005; Bhattacharya et al. 2006).

Table 2 presents the mean values of the total arsenic and fluoride concentrations in groundwater for each county in Cordoba Province (from unprocessed data collected from official reports from 518 monitoring wells) (Obras Sanitarias de la Nación 1942).

Monitoring wells in the *Riojan plain and associated salt mines* zone showed arsenic concentrations between 0 and 0.04 mg/l and fluoride concentrations between 0.5 and 2.2 mg/l. Observed arsenic and fluoride concentrations in the *Pampean mountains and their valleys* ranged from 0 to 0.32 mg/l and 0.2

Table 2 Mean concentrations of arsenic and fluoride in groundwater in Cordoba Province

County	Sup. (km^2)	Population (hab)[a]	Rural population[a] (%)	As[b] (mg/l)	F[b] (mg/l)	*n*
Calamuchita	4,642	45,418	27.0	0.04	0.56	7
Capital	562	1,284,582	1.0	S/D	S/D	–
Colón	2,588	171,067	10.0	0.02	0.64	15
Cruz del Eje	6,653	52,172	27.6	0.03	1.30	6
General Roca	12,659	33,323	29.4	0.25	5.34	35
General San Martín	5,006	116,107	13.9	0.16	1.26	38
Ischilín	5,123	30,105	20.9	0.03	0.66	5
Juárez Celman	8,902	55,348	22.3	0.22	2.45	47
Marcos Juárez	9,490	99,761	9.6	0.21	1.94	49
Minas	3,730	4,881	100.0	0	0.90	3
Pocho	3,207	5,132	100.0	0.07	1.50	2
Presidente Roque S. Peña	8,228	34,647	19.0	0.1	2.52	31
Punilla	2,592	155,124	3.4	0.02	1.48	20
Río Cuarto	18,394	229,728	10.6	0.14	3.51	60
Río Primero	6,753	42,429	45.8	0.04	0.62	9
Río Seco	6,754	12,635	53.6	0.03	1.55	13
Río Segundo	4,970	95,803	17.3	0.09	1.15	34
San Alberto	3,327	32,395	41.4	0.04	0.81	7
San Javier	1,652	48,951	29.9	0.02	0.42	6
San Justo	13,677	190,182	20.1	0.17	1.40	39
Santa María	3,427	86,083	28.2	0.03	0.95	4
Sobremonte	3,307	4,531	54.4	0.06	2.80	2
Tercero Arriba	5,187	107,460	12.7	0.06	1.00	30
Totoral	3,145	16,479	56.9	0.04	0.55	2
Tulumba	10,164	12,211	73.2	0.05	1.13	11
Unión	11,182	100,247	21.5	0.21	1.81	43

Rural population includes dispersed people and villages with <2,000 people, [a] from Instituto Nacional de Estadísticas y Censos (2001), [b] from Obras Sanitarias de la Nación (1942)

n = number of wells being sampled

to 7.8 mg/l, respectively. On the other hand, the higher contents of arsenic and fluoride were observed in the *Chaco-Pampean plain*. In this hydrogeologic region, the arsenic concentrations ranged from 0 to 1.8 mg/l, while in the case of fluoride, they ranged from 0 to 13.5 mg/l.

Materials and methods

Two groundwater quality geoindicators are defined from the arsenic and fluoride concentrations:

$$N1_i = \alpha \frac{\mathrm{As}_i}{0.01\ \mathrm{mg/l}} + \beta \frac{\mathrm{F}_i}{1.5\ \mathrm{mg/l}} \quad (1)$$

$$N2_i = \gamma \frac{\mathrm{As}_i}{E[\mathrm{As}]} + \delta \frac{\mathrm{F}_i}{E[\mathrm{F}]} \quad (2)$$

where α, β, γ, and δ are weighting factors adopted according to local requirements and risk factors associated to each contaminant, As_i and F_i are the measured arsenic and fluoride concentrations, $E[\mathrm{As}]$ the mean arsenic concentration, $E[\mathrm{F}]$ the mean fluoride concentration, and 0.01 mg/l and 1.5 mg/l are the World Health Organization (WHO) suggested values for naturally occurring arsenic and fluoride in drinking water, respectively (WHO 2006). Geoindicators $N1$ and $N2$ relate the observed concentrations with the allowable concentrations suggested by the WHO and with the average concentrations observed in previously defined hydrogeologic regions, respectively. Both indicators are dimensionless numbers that allow a rapid visualization of the regional groundwater quality related to the presence of these two chemical elements, regardless of the observed local chemical concentrations.

On the other hand, the correlation between the total arsenic and fluoride concentrations measured in groundwater is determined as:

$$\rho_{\mathrm{As,\,F}} = \frac{Cov[\mathrm{As,\ F}]}{\sigma[\mathrm{As}]\sigma[\mathrm{F}]} \quad (3)$$

$$Cov[\mathrm{As,\ F}] = E[(\mathrm{As}_i - E[\mathrm{As}])(\mathrm{F}_i - E[\mathrm{F}])] \quad (4)$$

where $\rho_{\mathrm{As,\ F}}$ is the coefficient of correlation, Cov [,] the covariance, E[ ] the mean value, and σ[ ] is the standard deviation.

Maps showing the spatial distribution of arsenic and fluoride concentrations were generated by using a surface mapping system. The spatial variation of the previously defined geoindicators was also obtained. The data was interpolated by considering the spatial continuity of the available points by means of the Kriging method (see Krige 1978; de Marsily 1986; Marengo et al. 2008). The location of monitoring wells can be seen in Fig. 1. The number of wells being sampled in each county is shown in Table 2. There is an average of three wells monitored at each location. Note that several points located beyond the province border are included in order to avoid boundary effects on the obtained maps.

The spatial variation of arsenic in groundwater is compared with epidemiologic evidences reported by Hopenhayn-Rich et al. (1998) for the years from 1986 to 1991. The standardized mortality ratio (SMR) for lung, kidney, liver, and skin cancer is analyzed. Notice that only historic records of arsenic concentrations are considered, since epidemiologic evidences can be observed in people consuming contaminated groundwater after long periods (usually >20–30 years), even though other factors such as concentration, arsenic species type, climate, and human factors may be relevant (Bergoglio 1963; Smith et al. 2000).

Results and discussion

Figure 3 shows the spatial variation of maximum (a) and minimum (b) arsenic concentrations in groundwater, as obtained by using the Kriging interpolation method. The higher concentrations are observed in the northeastern, eastern, and southern region, while the lower values are detected in the northwestern part of the study area, with values as high as 1.8 mg/l (with a local maximum of 12 mg/l in 'La Francia' town, omitted in the map).

The zone significantly affected by the presence of arsenic corresponds to the Chaco-Pampean plain (Fig. 1). About 90% of the area has maximum concentrations higher than 0.01 mg/l (Fig. 3a), which suggest that at least one of the aquifers exceeds the value recommended by the WHO. In addition, regions with very high As concentration also display a higher difference between the maximum (Fig. 3a) and minimum (Fig. 3b) observed values. This tendency is attributed to the spatial variability (horizontal and vertical) of arsenic concentrations and to the presence of multilayer aquifers (Schulz et al. 2005).

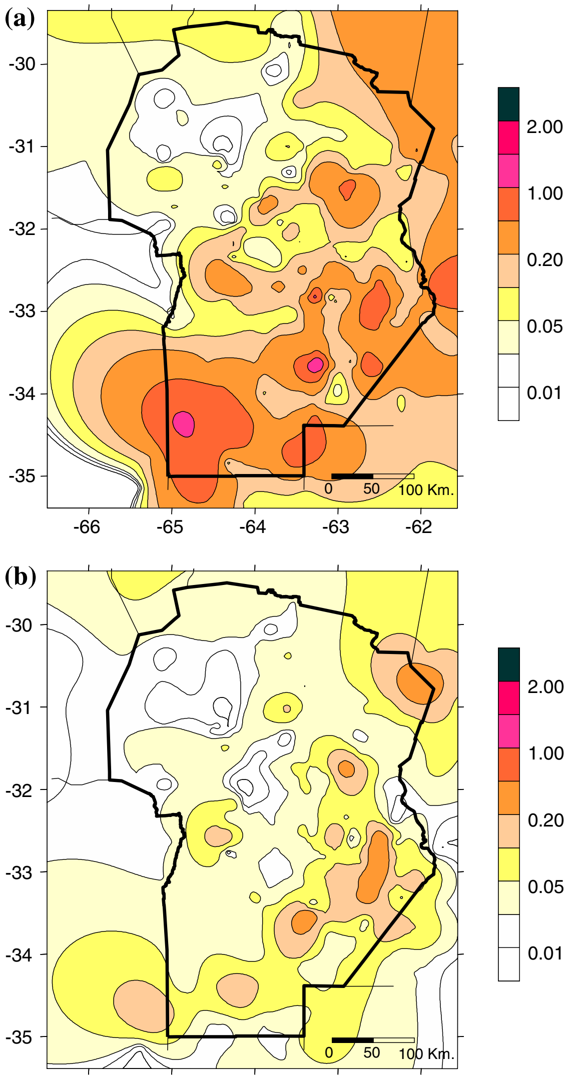

Fig. 3a, b Spatial distribution of arsenic in groundwater in mg/l: **a** maximum registered concentrations; **b** minimum registered concentrations

Figure 4 presents the spatial variation of maximum fluoride concentrations in groundwater. In this case, the higher concentrations are also observed in the southern and eastern regions. The measured values fall between 0 and 13.5 mg/l, with a mean of 2.36 mg/l. Note that the mean value of fluoride concentration in groundwater exceeds the limit suggested by the WHO ($F < 1.5$ mg/l).

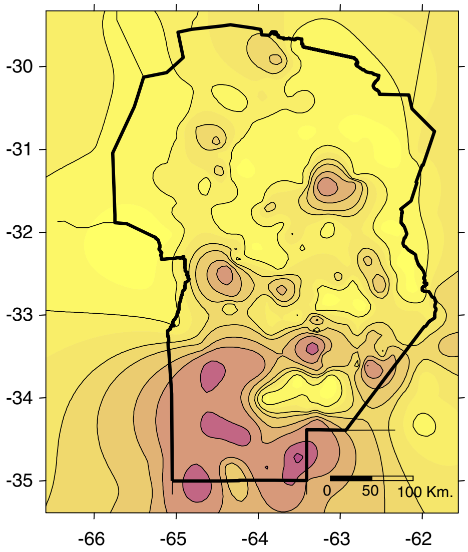

Fig. 4 Spatial distribution of the maximum registered concentrations of fluoride in groundwater in mg/l

Regional studies show that hydrogeologic zones substantially affected by high mean As concentrations in groundwater also contain high mean F concentrations (e.g., the Chaco-Pampean plain in Figs. 3a and 4). However, in some cases, the analysis of specific monitoring wells indicate that the presence of As is not always associated with high F concentrations.

Figure 5 shows the relation between the mean concentrations of As and F in groundwater for each county in Cordoba Province. There is a clear positive relationship between the As and F concentrations, since both variables increase in the same direction (this tendency emerges for the mean natural concentrations determined for each county). The obtained coefficient of correlation (Eq. 3) gives $\rho_{\mathrm{As,F}} = +0.68$, which means that As and F concentrations are somehow correlated to each other. This

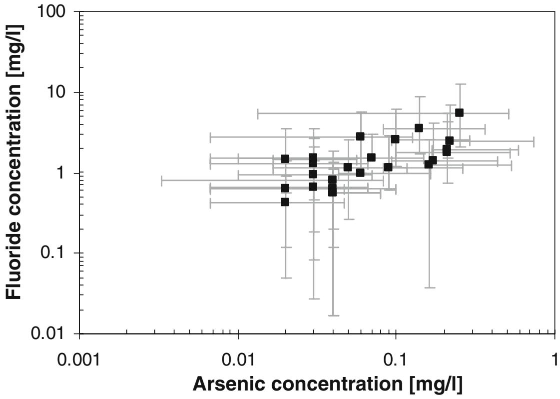

Fig. 5 Relationship between the observed fluoride and arsenic concentrations in groundwater

correlation is associated with the same origin of both chemical species related to the dissolution of the volcanic glass fraction of loess.

In spite of the global regional trend observed for the mean concentrations shown in Fig. 5, the local differences between As and F concentrations produce a noticeable variability, quantified by the error bars. The error bars show a mean ± one standard deviation interval ($E \pm \sigma$) for the two considered chemical species (64% confidence interval). An average of 20 measurements was considered in order to compute the variance of natural concentrations in order to quantify the variability in the observed trend.

Most of the data and the corresponding error bars presented in Fig. 5 have arsenic and fluoride concentrations higher than the maximum allowed concentrations of 0.01 and 1.5 mg/l, respectively. This tendency confirms the importance of subsurface water contamination in Cordoba.

Figure 6 shows the spatial variation of the groundwater quality geoindicators defined by Eqs. 1 and 2. The maps were developed by adopting the mean values of As_i and F_i for the monitoring wells at each site. The weighting factors α, β, γ, and δ were all assumed to be equal to 1. In addition, the following mean concentrations were defined: $E[\mathrm{As}] = 0.03$ and $E[\mathrm{F}] = 1.01$ for the Riojan plain and its associated salt mine zone, $E[\mathrm{As}] = 0.06$ and $E[\mathrm{F}] = 1.51$ for the Pampean mountains, and $E[\mathrm{As}] = 0.16$ and $E[\mathrm{F}] = 2.14$ for the Chaco-Pampean plain.

The main difference between the geoindicators $N1$ (Eq. 1) and $N2$ (Eq. 2) is that the first shows the combined effect of having concentrations higher than the maximum values suggested by the WHO, while $N2$ allows identifying the joint effect of the variability of reported concentrations, with respect to the natural expected value for the aquifers in previously defined hydrogeologic regions. In terms of applications, $N1$ gives information related to the potential use of the groundwater as a source of drinking water, while $N2$ provides details related to the variability of the aquifer useful for identifying anthropogenic sources of contamination.

The defined geoindicator $N1$ displays very high spatial variability in an extended area with extremely high values in the Chaco-Pampean plain (Fig. 6). In most cases, $N1$ was higher than 1, which means that either concentrations of As, F, or both exceed the value suggested by the WHO. On the other hand, the spatial variability of $N2$ suggests that arsenic and/or fluoride concentrations may have high local values at each hydrogeologic region. This pattern is associated with the presence of a complex multilayer aquifer system, since no anthropogenic sources of contamination were identified.

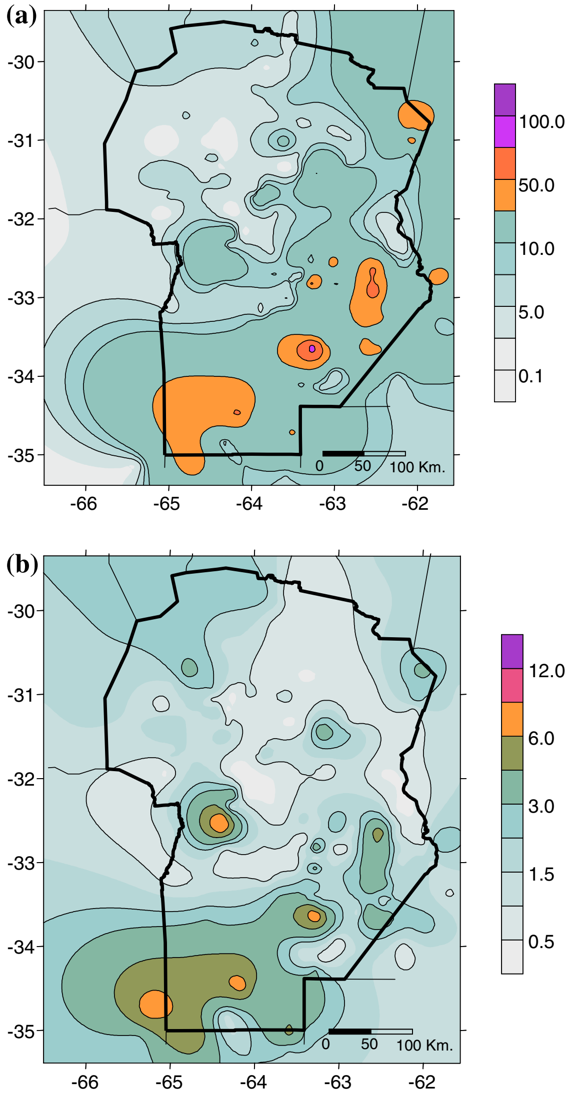

Fig. 6 **a** Geoindicator N1 (Eq. 1). **b** Geoindicator N2 (Eq. 2)

Figure 7 classifies all counties according to the percentage of monitoring wells where the As concentration in groundwater is lower than 0.01 mg/l. The zone where most of the wells contain concentrations higher than that allowed by the WHO almost coincide with that where the maximum concentrations were detected (Fig. 3a). These results suggest

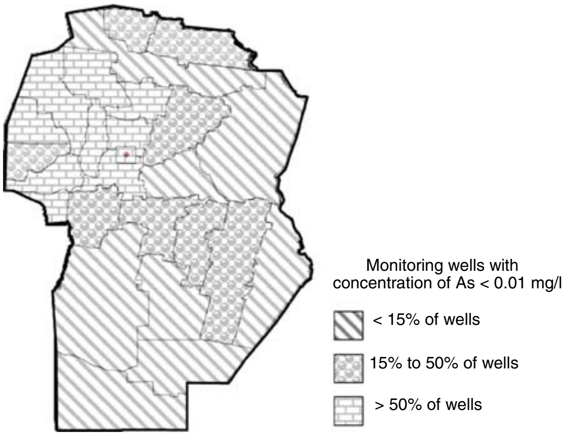

Fig. 7 Percentage of monitoring wells where the As concentration in groundwater is lower than 0.01 mg/l

that less than 15% of the monitoring wells may contain As concentrations lower than 0.01 mg/l in 52% of the surface. The more affected zone coincides with the Chaco-Pampean plain hydrogeologic region. On the other hand, more than 50% of wells may not be affected by arsenic contamination in the west and northwest of the study area. Finally, more than 50% of the study area is affected by arsenic contamination, with concentrations higher than 0.01 mg/l in more than 85% of monitoring wells.

Figure 8 presents the relationship between the SMR for kidney, lung, liver and skin cancer, and rural population for all counties. The SMR for males and females and the 95% confidence interval indicated by the error bars were obtained from Hopenhayn-Rich et al. (1998). They performed a comprehensive analysis of exposure groups and confirmed that there is a good correlation between the risk of lung and kidney cancer and the evidence of arsenic ingestion, while no clear association was determined for the mortality from liver and skin cancer. However, tendencies observed in Fig. 8 clearly indicate that lung, kidney, skin, and liver cancer all show some positive correlation with the rural population fraction. This tendency emerges when the data is related to the rural population that uses well water as a drinking water source. Hence, mortality related to kidney, lung, liver, and skin cancer in Cordoba Province is associated to the ingestion of arsenic-contaminated water, even though other risk factors, such as

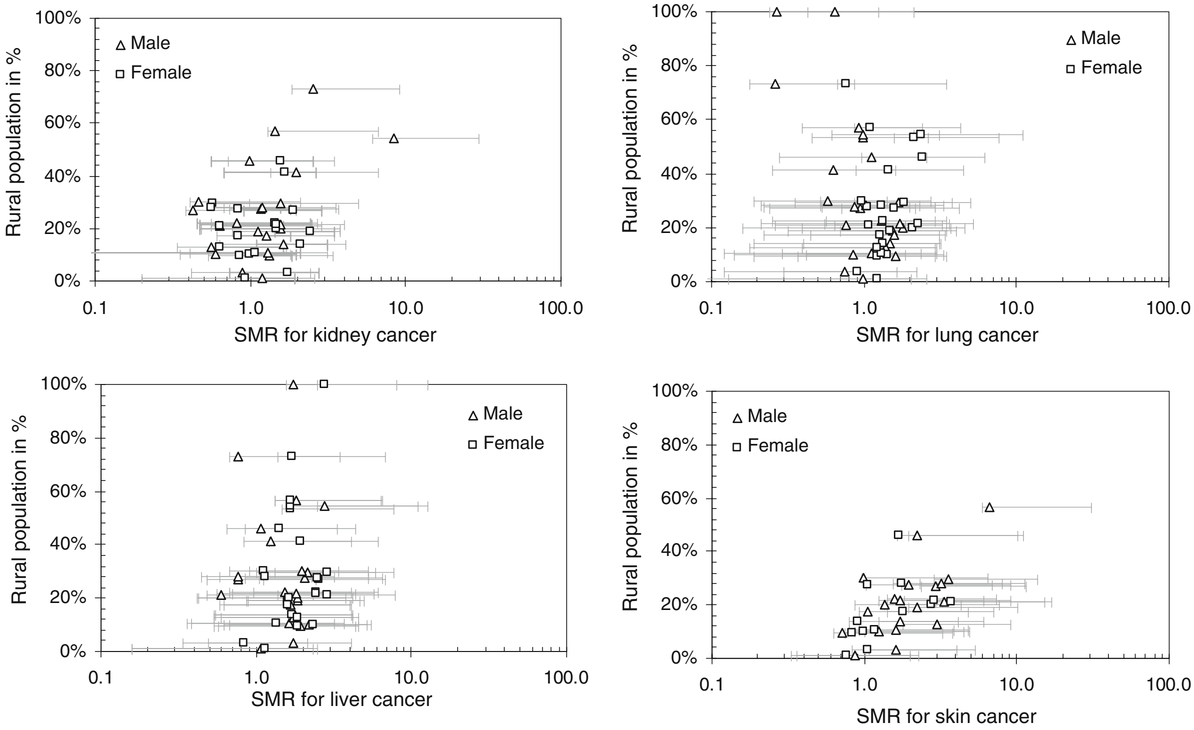

Fig. 8 Relationship between rural population and standardized mortality ratio (SMR) for kidney, lung, liver, and skin cancer in Cordoba Province (SMRs from Hopenhayn-Rich et al. 1998)

smoking, pollution, and ultraviolet ray exposure may be relevant. Additionally, synergetic effects and multisystem disease could also be responsible for the positive correlations observed in Fig. 8.

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