REVISTA OBSERVATORIO DE LA ECONOMIA LATINOAMERICANA
Curitiba, v.21, n.10, p. 16061-16078. 2023.

ISSN: 1696-8352

Advances and challenges to implementing the project: arsenic in Uruguayan groundwater and associated health risk

Avanços e desafios para a implementação do projeto: arsênio nas águas subterrâneas Uruguaias e risco associado à saúde

DOI: 10.55905/oelv21n10-084

Recebimento dos originais: 08/09/2023
Aceitação para publicação: 10/10/2023

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ABSTRACT
In Uruguay there is a great concern about the arsenic (As) geogenic contamination in groundwater as an environmental health problem, after rather high levels of As in groundwater in some areas (above 20 µg L-1) were reported and considering that the majority of the rural population consumes drinking water from private aquifer wells. On

the other hand, Uruguay has an abundance of wells and quality groundwater, and it is vital to protect Uruguayan aquifers as important resources. In this respect we proposed and started to implement a multidisciplinary and interinstitutional program: AsURU Project ("Arsenic in Uruguayan groundwater and associated health risk") (website: facebook.com/AsURUArsenico) to address this environmental and health problem. Although there are no systematic studies on the quality of groundwater or on the incidence of health effects associated with As exposure, a medical geology study is being performed by the research team led by Nelly Mañay, Ph.D., to assess health risks retrospectively by correlating reported cancer incidence with As groundwater levels, distributed by different localities. The AsURU Project has developed a working plan that consists of: (a) identification of main Uruguayan aquifers with relevant As data, (b) subdivision in study areas, (c) establishment of geographically delimited pilot areas following specific criteria: i) areas which have drinking water supplied by aquifers with As levels above WHO guidelines; ii) should represent a significative extension of the concerned aquifer; iii) have enough information about the aquifer hydraulic parameters and iv) availability of quantitative data about population´s exposure timeline to groundwater and shreds of evidence about health adverse effects (d) hydrogeologic, hydrodynamic and hydrogeochemical study to identify the origin of As and space-time mobility, (e) evaluation of As exposure through groundwater in the pilot areas, (e) recommend solutions like good construction and other remedial actions. The AsURU Project has seen great progress and also encountered challenges. Its main achievements in line with the proposed action plan have been: (1) the identification of pilot areas with As concentration levels in groundwater above 20 µg L-1 (max. permitted level as per Uruguayan regulations); and even with As level concentrations above 10 µg L-1 (max. permitted level as per WHO regulations). (2) the formation of three work groups to develop three related projects: the Groundwater Resource Group, the Chemistry and Medical Geology Group, and the North Littoral Group; (3) meetings were held, including the "I Symposium of Arsenic in Uruguayan Groundwater" in 2018, with the participation of various state-owned companies and the "II Symposium of Arsenic in Uruguayan Groundwater" in 2022. However, there is still no specific financial support for developing the program in its totality.

Keywords: arsenic concentration, groundwater, pilot área.

RESUMO

No Uruguai há uma grande preocupação sobre a contaminação geogênica do arsênio (As) em águas subterrâneas como um problema de saúde ambiental, depois que níveis bastante elevados de As em águas subterrâneas em algumas áreas (acima de 20 µg L-1) foram relatados e considerando que a maioria da população rural consome água potável de poços de aquíferos privados. Por outro lado, o Uruguai tem uma abundância de poços e águas subterrâneas de qualidade, e é vital proteger os aquíferos uruguaios como recursos importantes. A este respeito, propusemos e começamos a implementar um programa multidisciplinar e interinstitucional: AsURU Project ("Arsênio nas águas subterrâneas uruguaias e risco de saúde associado") (site: facebook.com/AsURUArsenico) para abordar este problema ambiental e de saúde. Embora não haja estudos sistemáticos sobre a qualidade das águas subterrâneas ou sobre a incidência dos efeitos na saúde associados à exposição ao As, um estudo geológico médico está sendo realizado pela equipe de pesquisa liderada por Nelly Mañay, Ph.D., para avaliar os riscos para a saúde retrospectivamente correlacionando a incidência relatada de câncer com os níveis de água subterrânea do As, distribuídos por diferentes localidades. O projeto AsURU desenvolveu um plano de trabalho que consiste em: (a) identificação dos principais aquíferos uruguaios com dados As relevantes, (b) subdivisão em áreas de estudo, (c) estabelecimento de áreas piloto geograficamente delimitadas seguindo critérios específicos: i) áreas que têm água potável fornecida por aquíferos com níveis As acima das diretrizes da OMS; ii) deve representar uma extensão significativa do aquífero em questão; iii) ter informações suficientes sobre os parâmetros hidráulicos dos aquíferos e iv) disponibilidade de dados quantitativos sobre o cronograma de exposição da população às águas subterrâneas e fragmentos de evidências sobre efeitos adversos à saúde (d) hidrogeológico, hidrodinâmico e hidrodinâmico estudo químico para identificar a origem de As e mobilidade espaço-tempo, (e) avaliação da exposição de As através das águas subterrâneas nas áreas piloto, (e) recomendação de soluções como boa construção e outras ações corretivas. O Projeto AsURU tem visto um grande progresso e também encontrou desafios. As suas principais realizações em conformidade com o plano de ação proposto foram: (1) a identificação de áreas-piloto com níveis de concentração de As em águas subterrâneas superiores a 20 µg L-1 (nível máximo permitido de acordo com a regulamentação uruguaia); e mesmo com concentrações de As superiores a 10 µg L-1 (nível máximo permitido de acordo com a regulamentação da OMS). (2) a formação de três grupos de trabalho para desenvolver três projetos relacionados: o Grupo de Recursos de Águas Subterrâneas, o Grupo de Geologia Médica e Química, e o Grupo Litoral Norte; (3) foram realizadas reuniões, incluindo o "I Symposium of Arsenic in Uruguayan Groundwater" em 2018, com a participação de várias empresas estatais e o "II Symposium of Arsenic in Uruguayan Groundwater" em 2022. No entanto, ainda não existe apoio financeiro específico para o desenvolvimento do programa na sua totalidade.

Palavras-chave: concentração de arsênio, águas subterrâneas, área piloto.

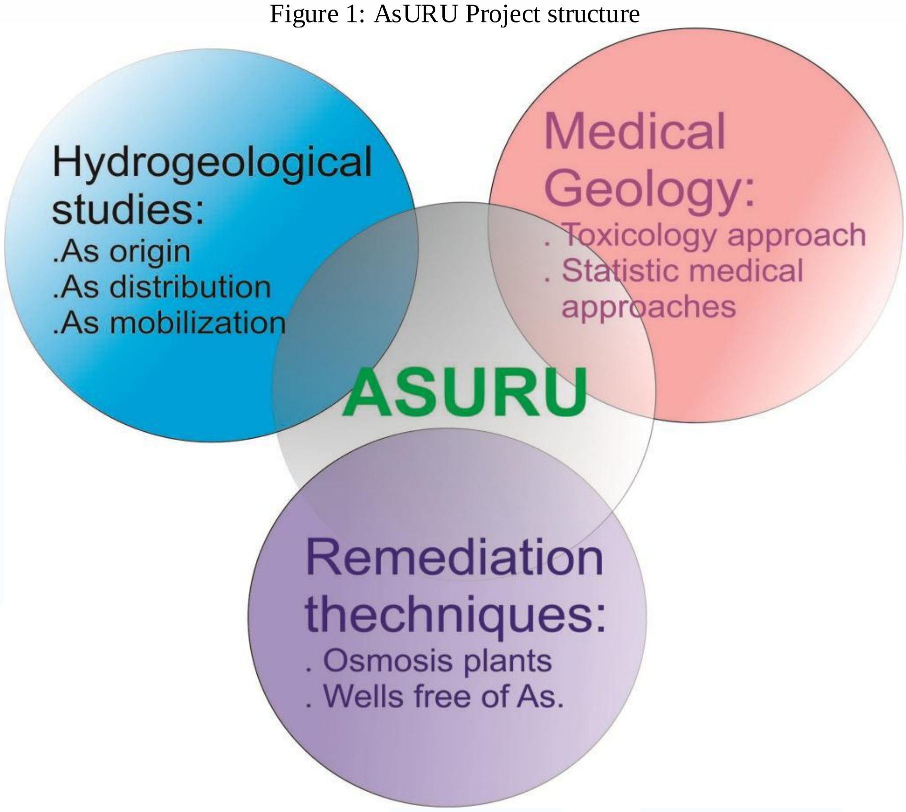
1 INTRODUCTION

Arsenic (As) is a natural element which in its inorganic form can be toxic for human beings. One of the main sources of As exposure is groundwater for drinking use. According to our national drinking water regulations and background, there are no systematic studies on the quality of groundwater or on the incidence of health effects, regarding arsenic (As) exposure in Uruguay. However, after rather high levels of As in groundwater (above 20 µg L^{-1}) were reported in some areas and considering that most of Uruguay´s rural population consumes drinking water from private aquifer wells, an initial medical geology study has been performed, trying to assess health risks retrospectively by correlating the incidence of melanoma in each locality with higher As in groundwater levels (Mañay et al., 2019). Other medical geology studies have also been carried out (Machado et al. 2020) including one regarding the As correlation with fluoride, sulfate, iron, and manganese (Machado et al. 2019). In Uruguay, there is a great concern about the arsenic (As) geogenic contamination in groundwater as an environmental health problem. On the other hand, Uruguay has an abundance of wells and quality groundwater, and it is vital to protect Uruguayan aquifers as important resources.

In an attempt to find integral solutions to this environmental problem, we have presented and are currently coordinating the project "Arsenic in Uruguayan groundwater and associated health risk" (AsURU) (Collazo et al. 2019, 2020 and Pamoukaghlián, 2021) with the incorporation of new researchers and the participation of delegates of several public bodies in the project´s regular meetings. AsURU has three main components: (a) Hydrogeological studies, (b) Medical Geology and (c) Remediation techniques (Figure 1).

This multidisciplinary and interinstitutional project has not received public funds. Still, it has been an excellent framework for researchers from geosciences, biosciences, and stakeholders to come together to address Arsenic-related environmental health issues in Uruguay (*website: facebook.com/AsURUArsenico*).

Figure 1: AsURU Project structure



Source: the authors

1.1 STUDY AREA

As the study area, the AsURU Project considers *a priori* the whole country, but the main aquifers which represent the most important water resources and where high levels of arsenic (As) were detected are specially taken into account. This includes the southwestern area where the Raigon Aquifer is located; a west littoral area where the Mercedes Aquifer is located and the Northern Littoral area, where the Salto Aquifer is an important water resource. For each of these aquifers, a pilot area has been selected (Figure 1).

1.2 GEOLOGICAL AND HYDROGEOLOGICAL FRAMEWORK

In Uruguay, there are two main aquifers affected by the arsenic problem. One is the Raigon Aquifer System and the other is the Littoral Aquifer System. Another aquifer that has been taken into account is the Salto Aquifer. Pilot areas are located in these three aquifers.

The Raigon Aquifer is geologically conformed by the Raigon Formation defined by Goso and Bossi (1966). This unit outcrops in the littoral of Colonia, San José, and Canelones departments and it is made of whitish sandstones of variable granulometry with green claystone lenses and beds, conglomerates, and calcareous concretions. Rhythmic stratification, lenses, parallel and cross-stratification are the main sedimentary structures, and it corresponds to a transitional continental fluvial environment (Preciozzi et al. (1985) and Spoturno et al. (1993). Its age is Medium Pliocene to Medium Pleistocene after Perea y Martínez (2004). It is underlain by the Camacho Formation and overlain by the Libertad Formation. The Raigon Aquifer behaves like a confined aquifer in its southeastern region and in other instances as a phreatic aquifer. According to Heinzen et al. (2003) the hydrodynamic parameters are: average transmissivity 264 m^2/d, average storage coefficient 6,5 x 10^{-3} and specific flow rates estimated at about 6 m^3/h/m. The average flow rates of the exploited wells are about 10-20 m^3/h, after Montaño et al. (2005). Hydrogeochemical characterization allows us to classify groundwater as sodium bicarbonate water, without any anomalies relating to majority or minority elements. However, nitrate pollution is a problem, and there are As values above Uruguay's state-mandated limits and above the recommended WHO (2022) limit, up to 40 µg L^{-1}. The Raigon Aquifer is studied in the selected pilot areas, in Conchillas and Libertad. It is important to mention that in the Conchillas area, groundwater is pumped out of the sedimentary Raigon Aquifer and from the fractured aquifer conformed by the crystalline basement. This crystalline basement corresponds to the granite gneiss Florida strip, between the Arroyo Grande and San José belts (Bossi et al. 1998). This belt is intruded by post-tectonic granites such as the Piedra Alta granodiorite o the Conchillas granite-gneiss. There are commonly decametric mica-schists xenoliths in the granites and gneisses (Oyhantçabal et al. 2011).

Recently, the Libertad pilot area was extended to the Libertad-Kiyú area to better study the As anomalies in the Raigon Aquifer. Panzl (2022) encountered As concentrations in Kiyú between 12 and 20 µg L^{-1}, with As III being the predominant type.

The Littoral Aquifer System is made up of the Cretaceous Aquifers of the Paysandú Group, particularly the Mercedes Formation which spans through 25,000km^2

across Salto, Paysandú, Río Negro, Colonia, Flores, and Durazno departments (Goso and Perea, 2004). It overlies in discordance the Paleoproterozoic crystalline basement and the Cretaceous Arapey Formation. The Mercedes Formation was defined by Lambert (1939) and its age was assigned as Superior Cretaceous by Serra (1945). Goso et al. (1999) describe predominant clastic lithologies (conglomerates and sandstones) calcareous, ferriferous, and silicified lithofacies often encountered and minor pelitic lithologies. The same author divides the Mercedes Formation into two members: (1) the Yapeyú Member, made of whitish sandstones, well sorted, quartz feldspar with calcareous cement, with parallel and cross lamination, and normal gradation; shales with parallel lamination and often with carbonatic interlaminations. (2) the Chileno Member is made of whitish and yellowish sandstones and conglomerates. Conglomerates are polymictic with quartz, basalt, granite, gneiss, quartzite, and schist mega clasts. Subordinate sandstones are medium to coarse-grained, grey colored with calcareous or silicious cement.

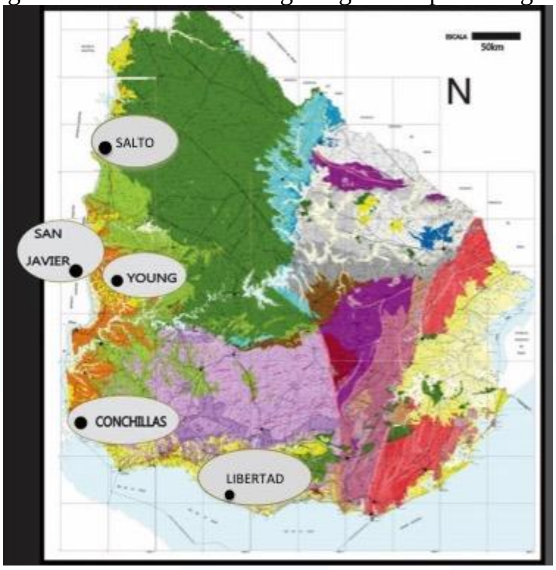
Goso et al. (1999) and Goso y Perea (2004) interpret a fluvial paleoenvironment, with narrow channels and variable flow conditions. The Mercedes Aquifer is alternatively phreatic or confined. Average permeability is medium to high and varies depending on whether the aquifer is behaving as phreatic or confined (Heinzen et al. 2003). Specific flow for the Mercedes Aquifer toward the southern portion of the Río Negro River is around 0,6 m/h/m, maximum 3,8 m/h/m, and minimum 0,05 m/h/m. Toward the northern area of the Río Negro higher values are observed: an average of 1,03 m/h/m, a maximum of 6,5 m/h/m and a minimum of 0,05 m/h/m, according to Bonjour (2004). Transmissivity is 36 m/d average with a minimum of 0,667 and maximum of 189 m/d, and storage coefficients indicate that the aquifer is confined in this area (Bonjour, 2004). Hydro-geochemistry characterization allows us to classify groundwater as calcium bicarbonate water, generally of good chemical quality without any significant anomalies relating to majority and minority elements, according to Heinzen et al. (2003). However, several cases of As values surpassing state-mandated limits have been encountered. In order to study the Littoral Aquifer System, two pilot areas were selected in Young and San Javier (Figure 2).

The Salto Aquifer spans 15 to 20 km from Salto City to Bella Union City, it is formed by sandstones and conglomerates of the Salto Formation, defined by Caorsi and Goñi (1958). According to Veroslavsky and Montaño (2004), the Salto Formation is made of sandstones and conglomerates, orange, reddish, grey, and brownish, interbedded with green shales affected by important silicification events. This unit is less than 30 m thick in its type area (Preciozzi et al. 1985). Its age is the Late Cenozoic, according to De Santa Ana and Veroslavsky (2002). Aquifer transmissivity is 50 m^2/d (Montaño et al. 1995) and considering a medium thickness of 20 m, the permeability is 2,5 m/d. The estimated aquifer flow is 1750 m^3/d (Montaño, 2004). Hydrogeochemical characterization allows us to classify the groundwater as calcium bicarbonate water. There are no significant anomalies in respect to physical properties or majority and minority ions, but rather high As levels have been encountered. The pilot area selected to study this aquifer is Salto.

2 METHODOLOGY

The AsURU Project has developed a working plan that consists of: (a) identification of main Uruguayan aquifers with relevant As data, (b) subdivision in study areas, (c) establishment of geographically delimited pilot areas following specific criteria: i) areas which have drinking water supplied by aquifers with As levels above WHO guidelines; ii) should represent a significative extension of the concerned aquifer; iii) have enough information about the aquifer hydraulic parameters and iv) availability of quantitative data about population´s exposure timeline to groundwater and evidences about health adverse effects (d) hydrogeologic, hydrodynamic and hydrogeochemical study to identify the origin of As and space-time mobility, (e) evaluation of As exposure through groundwater in the pilot areas and health risk assessment, (e) recommend solutions like well construction and other remedial actions.

Figure 2: Pilot areas on the geological map of Uruguay



Source: Modified from Bossi et al. (1998).

3 RESULTS AND DISCUSSION

Arsenic levels in groundwater samples were anomalous in several cases, and lightly superior to the admissible maximum values, according to the existing regulations in Uruguay (20 µg L^{-1}) (UNIT 2010). This difference is in fact more substantial when we compare Uruguayan aquifers' As concentration values with the WHO norms (10 µg L^{-1}) (WHO 2022). Arsenic concentrations can be observed in Figure 3 (after Collazo et al. 2019) and groundwater Arsenic distribution is shown in Figure 4 (after Wu et al. 2021). This led us to establish five pilot zones: (1) Conchillas with average values of As > 30 µg L^{-1} in groundwater samples; (2) Young with As > 10 µg L^{-1} in groundwater samples; (3) San Javier with As > 10 µg L^{-1}, where the drinking water supplier, the state-owned company O.S.E, is implementing remediation using osmosis; (4) Libertad with As between 10 µg $L^{-1}$and 30 µg L^{-1}; (5) Salto (Northern Littoral) with As between 40 µg L^{-1} and 50 µg L^{-1} values. Solving the problem and keeping As at admissible rates in groundwater is imperative for the country to ensure the health and well-being of its population.

The Libertad pilot area was extended, becoming the Libertad-Kiyú pilot area. In Kiyú a research project (Arsenic mitigation in groundwater, supported by CTAgua, the *Water Technological Center*, under the responsibility of Gianella Fachin, Ph.D., and

Paula Collazo, Ph.D., was developed and water levels with As concentrations above 20 µg L^{-1} were identified, reaching maximum values of 40 µg L^{-1} . The objective of this project was to evaluate the efficiency of a remediation mechanism that uses ferric chloride to help retain As and lower its concentration in the water. The project has taken place in a rural school (Figure 5 A, C) with the construction of a new well where rock sampling and water sampling have been consistently carried out, encountering a correlation between the high As levels in water with rock As concentrations for the different depths (results are available at CTAgua 2022 and were presented by Collazo et al. 2022).

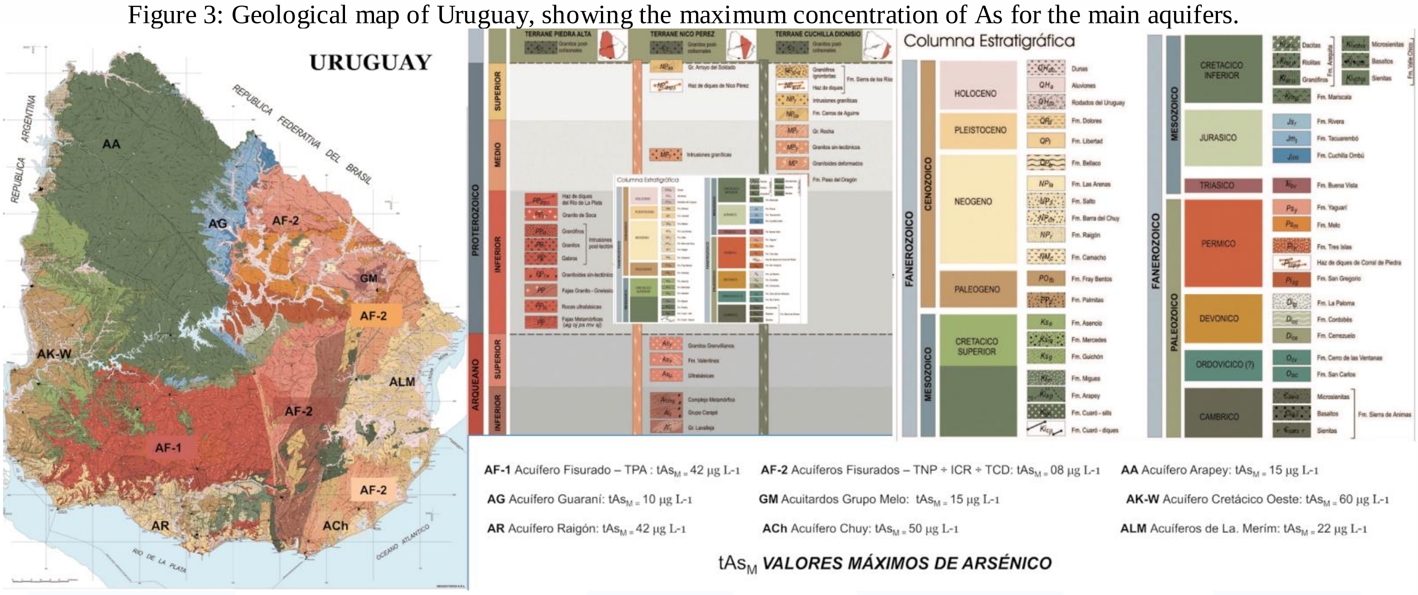
Following these studies, another project is being carried out in the Kiyú pilot area: “Arsenic in groundwater: sources, spatial variation and mobilization factors” supported by ANII (Uruguay's agency for research and innovation) and under the responsibility of Valery Bühl, Ph.D. This project has the goal of evaluating arsenic anomalies in the Raigon Aquifer and studying their correlation with the existing geological, geochemical, and microbiological conditions to identify the processes that cause its mobilization and generate a methodology to prevent the occurrence of this toxic element.

Furthermore, a Student CSIC Project (PAIE, “Student Research Academic Project”) and a grade thesis have been carried out in Kiyú (Panzl, 2022). The PAIE Project focused on the hydrodynamic behaviour of As, establishing the concentration variation during a pumping test, with positive results. Panzl (2022) proved this hydrodynamic correlation with a long pumping test of 72 hours with As III and As V sampling in different flow rate steps (Figure 5, B).

A M.Sc. thesis to study As in groundwater in the littoral north of Uruguay is in course by Andrea Texo, and a Ph.D. thesis entitled “Arsenic speciation in groundwater study and its possible impact on the population” is being developed by Paulina Pizzorno, M.Sc., partial results have been published (e.g., Pizzorno et al. 2021; 2022), detailing innovative advances in the method of As species determination (Pizzorno et al. 2023).

Additionally, the Littoral North Group of AsURU made significant advances in the understanding of groundwater As distribution in Uruguay, modelled by a variety of machine learning, basic expert systems, and hybrid approaches Wu et al. (2021).

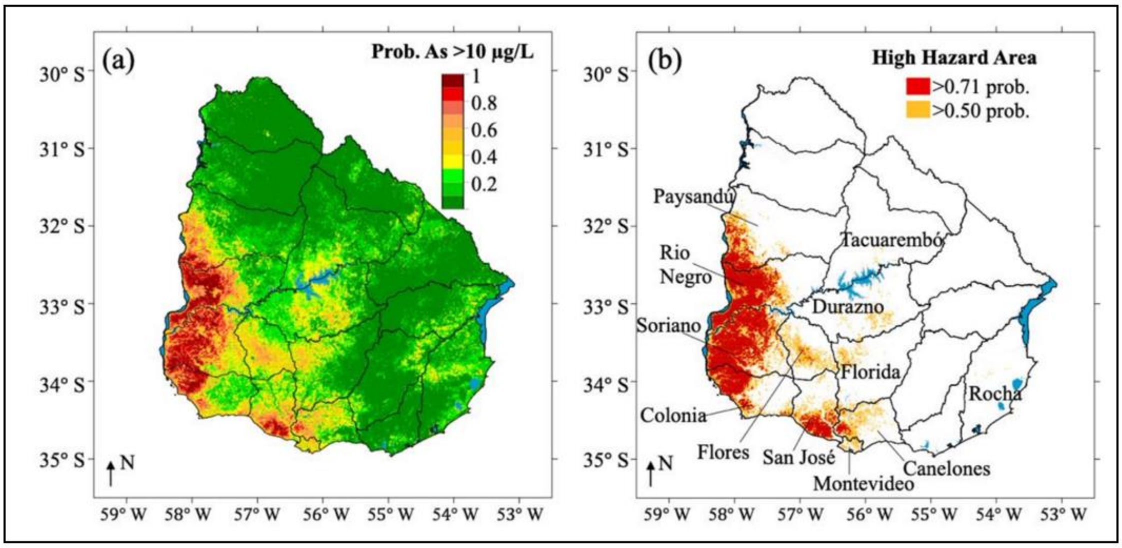
Figure 3: Geological map of Uruguay, showing the maximum concentration of As for the main aquifers.



Source: Extracted from Collazo et al. (2020), based on O.S.E. data source.

Another research took into account data of As total from 432 wells, it shows the distribution of groundwater arsenic in Uruguay modelled by a variety of machine learning, basic expert systems, and hybrid approaches (Wu et al, 2021). They demonstrated that shallow and deep aquifers of Uruguay had similar distributions of high-arsenic groundwater in the four departments of southwestern Uruguay: Paysandú, Río Negro, Soriano and Colonia, which mainly come from the Mercedes aquifer as it is shown in Figure 4 (Wu et al., 2021). Additionally, the high level of arsenic occurs more in shallow groundwater in the departments of San José (where the Raigón aquifer is located), Canelones, the northwest corner of Florida, western Durazno and Flores (Wu et al., 2021). Also when they compared the arsenic distribution in sedimentary and crystalline aquifers, high-arsenic groundwater appeared more in sedimentary (36%; n = 108 of 300) than crystalline (5%; n = 7 of 134) aquifers, they proposed that it could be related to different predominant mobilization processes.

Figure 4: Pure overall model (1A-ML-Pure) of Uruguay groundwater As concentrations: (**a**) map of probability of groundwater As concentrations exceeding 10 µg/L of shallow aquifers; (**b**) map of high groundwater As hazard areas of deep aquifers (defined here by a probability exceeding cutoff values of 0.50 or 0.71).



Source: Wu et al. (2021).

Figure 5: Fieldwork in Kiyú: A) Rural School with groundwater As problems. B) Kiyú well sampling. C) Rural school well.

Source: the authors

4 CONCLUSIONS AND RECOMMENDATIONS

The AsURU Project has seen great progress and also encountered challenges. Its main achievements in line with the proposed action plan have been: (1) the identification of pilot areas with As concentration levels in groundwater above 20 µg L^{-1} (max. permitted level as per Uruguayan regulations, UNIT 2010 – expected to change in 2023 to adopt the WHO norms); and with As level concentrations above 10 µg L^{-1} (max. permitted level as per WHO norms). (2) the formation of three work groups to develop three different projects: a) the Hydric Groundwater Resource Group, led by Paula Collazo, Ph.D. and Karina Pamoukaghlián, Ph.D.; b) the Chemistry and Medical Geology Group, led by Nelly Mañay, Ph.D., with the collaboration of Valery Bühl, Ph.D., and Paulina Pizzorno, M.Sc.; (c) the Littoral North Group led by Elena Alvareda, Ph.D. that they are also developing a methodology to study arsenic in rice and the impact of flooded cultivated soils with groundwater with Arsenic total levels above the recommended values. (3) meetings were held, including the First Meeting of the AsURU Project at CeReGAS (Regional Center for Groundwater Management), the "I Symposium of Arsenic in Uruguayan Groundwater" in 2018, with the participation of various state-owned companies (the national water company, O.S.E.; the national environmental department, DINAMA; the national water department DINAGUA; the mining and geology department DiNaMiGe), a mixed private-public company for rural housing, MEVIR, and the national university UdelaR (Chemistry, Science, Medicine and Engineering Faculties), and the "II Symposium of Arsenic in Uruguayan Groundwater," which took place in 2022, as part of the X Uruguayan Geological Congress. Various conferences and lectures were presented (see Bühl et al. 2022 and Collazo et al. 2022).

As mentioned before, some projects have been performed and are being performed nowadays. These are "subprojects" of the AsURU Project, which is in fact a research program with the capability to articulate and look for external financial support. The Technological Water Center (*CTAgua, Centro Tecnológico del Agua*) supported the "Groundwater arsenic mitigation" project implemented under the responsibility of Paula Collazo, Ph.D., and Gianella Facchin, Ph.D., during the Covid-19 pandemic, with great effort, in 2020 and 2021.

The national research and innovation agency, ANII, is supporting the “Arsenic in groundwater: sources, spacial variation and mobilization factors” project under the responsibility of Valery Bühl, Ph.D.

The CSIC students' project (PAIE), grade theses and postgraduate theses have been carried out in this context in different parts of the country, including: Kiyú pilot area (PAIE, grade and postgraduate theses), Punta Espinillo fractured aquifer (crystalline basement) in Montevideo, San Pedro fractured and sedimentary aquifers and others (crystalline basement and Raigón Aquifer).

Work team coordinators and collaborators have participated in the first and second "Symposium of Arsenic in Uruguayan Groundwater” events, in the 8$^{th.}$ International Congress on Arsenic in the Environment, and they are in charge of the Session "Toxic metal and metalloids pollution in surface and groundwater hydric resources" recently presented during the SETAC Latin America 15th Biennial Meeting (SETAC, Society of Environmental Toxicology and Chemistry) in 2023.

Notwithstanding these achievements, the project has also met with several difficulties and challenges. Although government entities have participated and expressed their concern about the problem and their interest in the project, it is not so easy to get the relevant government entities to get fully invested and help this or any other project achieved its goals. Moreover, the fact that there are really scarce resources is a great difficulty, and we will have to prioritize to tackle the most pressing issues.

ACKNOWLEDGEMENTS

We are grateful to CENUR-Litoral Norte (regional UdeLaR university center); to ANII, National Agency for Research and Innovation, for supporting the ANII FMV_1_2017_1_135656 and the ANII FMV_3_2022_1_172829 projects; and to CTAgua for supporting the “Groundwater arsenic mitigation” project. We are also grateful to the state water company, O.S.E., for the provided information.

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