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Arsenic occurrence in Europe: emphasis in Greece and description of the applied full-scale treatment plants

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ABSTRACT

The presence of arsenic in groundwater comprises a worldwide problem and is recognized as a human health threat. The present work summarizes the arsenic contamination in Europe, where many countries are affected by elevated arsenic concentrations (i.e. Greece, Hungary, Romania, Croatia, Serbia, Turkey, and Spain). In particular, in Greece, several groundwater resources contain arsenic at increased concentrations, which render these water sources as non-potable. Arsenic-affected regions in Greece are classified mainly in three categories, namely, the geothermal-affected waters, the alluvial deposits of rivers and aquifers, and those influenced by mineralization and are typically close to mining activities. In Greece, arsenic concentrations in geothermal waters vary from 30 to 4,500 μg/L, in the regions close to alluvial deposits from 15 to 100 μg/L and in areas affected by mining activities from 20 to 60 μg/L. Arsenic-removal plants have been installed in several towns in Greece. The applied removal technologies are mainly based on chemical precipitation with $FeClSO_4$ or adsorption onto iron oxide materials, such as Bayoxide, granular ferric hydroxide or AquAsZero. In the cases where As(III) is present, biological oxidation or ozonation is applied to convert As(III) to the less mobile forms of As(V). Specific arsenic removal capacity at an equilibrium concentration equal to the regulation limit of 10 μg/L was between 1.7 and 4.2 mg As/g of adsorbent for adsorption processes and between 18 and 59 mg As/g Fe for chemical precipitation.

Keywords: Arsenic; Europe; Greece; Removal

1. Introduction

As the world population increases beyond six billion, one of the most fundamental resources for human survival, clean water, is decreasing. The rising demands for sanitary water often cannot be met by surface water supplies. This has led to increased dependence on groundwater resources in many parts of the world, which has caused new health issues, mainly because of the presence of arsenic in the

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groundwater sources [1]. Exposure to arsenic can result in a variety of health problems in humans, including various forms of cancer (e.g. skin, lung, and bladder), cardiovascular and peripheral vascular disease, and diabetes [2].

Arsenic concentration in surface waters is relatively low (typically 0.1–2 μg/L). Due to the prevailing oxidizing conditions, arsenic is mainly adsorbed to particulate matter, which considerably lowers the arsenic concentration in the water body. The highest concentrations, resulting from natural processes, can be found in groundwater. Arsenic has been detected in several groundwater sources around the world in variable concentrations. In particular, in Southeast Asia more than 50 million people are exposed to arsenic concentrations over 50 μg/L [3,4]. This problem has been termed as “the largest poisoning of a population in history.” Arsenic contamination of groundwater has been reported for other areas of the world as well, such as in South America (i.e. Argentina and Bolivia), in the United States, and Europe [1].

This paper represents a short overview of arsenic concentrations found in groundwater within European countries, with emphasis given in Greece. Greece has several documented hotspots containing elevated arsenic concentrations in groundwater as well as other geogenic groundwater contaminants, such as uranium and boron. With respect to the origin of arsenic, we classified the arsenic-affected regions in three main categories. Finally, we summarized the treatment units installed in Greece to eliminate the arsenic problem in the affected areas. To the best of our knowledge, until to date, there is no paper in the literature summarizing the cases of arsenic contamination found in Europe and especially in Greece, while providing essential information on the treatment technologies applied for the elimination of this problem at full-scale treatment units.

2. Arsenic occurrence in European groundwater resources

Some large areas of Europe are affected by high arsenic concentrations. Most notable are the cases of Hungary and Romania, at the Pannonian Basin, where around 600,000 people are exposed to danger due to high arsenic concentrations in the groundwater sources [1]. Other regions, where high arsenic concentrations have been measured are the Kutahya Plain in Turkey and the region of Chalkidiki in Central Macedonia in Northern Greece [5,6]. However, arsenic has also been detected in several European groundwater resources. A summary of the recent findings are shown in Table 1.

2.1. *Arsenic contamination in groundwater resources of the Pannonian Basin*

The most notable cases of arsenic contamination in Europe are those linked to the Pannonian Basin (Hungary, Romania, Croatia and Serbia). Published estimates suggest that more than one million people are exposed to levels greater than the EU maximum admissible concentration of 10 μg/L in their drinking

Table 1
Maximum arsenic concentrations in European countries/regions

Country/region	μg As/L	Groundwater use	Ref.
Belgium/Antwerp and Limberg	50	Drinking water	[7]
Czech Republic/Mokrsko	1,690	Drinking water	[8]
Croatia	610	Drinking water	[9]
Denmark	30	Drinking water	[10]
Finland	1,040	Drinking water	[11]
France	40	Drinking water	[12]
Germany/Northern Bavaria and Wiesbaden	150	Drinking water	[13,14]
Hungary	800	Drinking water	[15,16]
Iceland	310	No drinking water	[17]
Italy/Volcanic areas of Ischia, Vesuvius, Etna, Stromboli	1,558	Drinking water in the case of Etna	[18]
Romania/Transylvania and Western Plain	200	Drinking water	[19]
Serbia/Vojvodina	150	Drinking water	[20]
Spain/Duero Basin, Ambles Valey in Avila, Caldes de Malavella	615	Drinking water in Duero Basin	[21,22]
Sweden	300		[23]
Switzerland/Ticino, Wallis	370	Wallis drinking water	[24]
Turkey/Kutahya Plain	10,700	Drinking water	[5]

water, making it the largest area so affected in Europe [9,15,16,19,20]. In the following paragraphs the most notable cases are briefly discussed.

2.1.1. Hungary

In Hungary, about 5×10^5 people are affected by drinking water with As concentrations exceeding 10 μg/L. Arsenic concentration reaches values up to 225 μg/L, which in most cases is controlled by reductive dissolution of iron oxy-hydroxides. The arsenic problem is mainly located in the southern part of the Great Hungarian Plain—Csongrfid County—which encompasses 4,263 km^2. The affected population is distributed in five towns and 54 villages [15,16].

2.1.2. Romania

In Romania, arsenic is mainly found in the area of Romanian Western Plains, where investigation of 40 wells revealed enhanced As concentrations ranging from 13 to 200 μg/L [19]. Approximately, 5×10^4 people are exposed to elevated arsenic concentrations because the Romanian Western Plains are in the area of the Pannonian Basin, which is linked with Hungary. It can be assumed that the reason for the elevated As concentrations in groundwater of Western Romania is reductive dissolution of iron oxy-hydroxides, which is also evident from the negative Eh values (−50 to −200 mV) [16,19].

2.1.3. Croatia

In Eastern Croatia, arsenic concentrations up to 610 μg/L were determined in groundwater, which potentially influence the drinking water of around 2×10^5 people [9]. The region also belongs to the Pannonian Basin, where reductive dissolution of iron oxy-hydroxides causes high arsenic concentrations in groundwater [16].

2.1.4. Serbia

In the region of Voijvodina in Northern Serbia, arsenic concentrations in groundwater of more than 150 μg/L were measured [20]. This area belongs to the South Pannonian Basin. Groundwater are characterized by the presence of dissolved iron, manganese, and ammonia, strong indicators of reducing conditions, which cause arsenic mobilization into the aquifers. More than 6×10^5 people (40% of the population of South Pannonian Basin) obtain drinking water with arsenic concentration higher than 10 μg/L [20].

3. Arsenic occurrence in Greek groundwater sources

Elevated arsenic concentrations in groundwater have been reported for many areas in Greece [6,25–34]. The regions where arsenic is found in Greek groundwater sources are classified in three major categories. The geothermal regions, such as in Chalkidiki and in Aridaia region of Northern Greece, the rivers' alluvial deposits such as those in the basins of Aksios, Nestos and Strymon rivers, and aquifers, influenced by mineralization, resulting in arsenic mobilization over the centuries. A minor case is that of the lake Volvi, which pollutes the nearby aquifers [32]. Table 2 contains the most remarkable arsenic cases found in Greek groundwater under the described classifications, visualized on the map of Greece as arsenic spots in Fig. 1. It must be noted that arsenic concentration in all bottled waters was found well below the drinking water regulation limit.

3.1. Arsenic release as a result from geothermal activities

Arsenic in groundwater and soils, influenced by geothermal activity, is intensively observed in Northern Greece (Table 2), and in particular, at Western Chalkidiki area. This area has very limited water resources, and subsequently As-polluted groundwater have minimized the water supply potential for people, animals, and agriculture. As pollution of groundwater is really severe in Western Chalkidiki near the town of Nea Triglia, in this area, As concentrations up to

Fig. 1. Major arsenic cases highlighted on the map of Greece as *arsenic spots*.

Table 2
Arsenic concentration in Greek groundwater

Area	Concentration (μg As/L)	Environmental conditions	Comments
Chalkidiki/Agia Paraskeui	4,500	Geothermal	Spa/[As(III)]*
Chalkidiki/Petralona	1,500–2,000	Geothermal	Irrigation wells
Chalkidiki/Triglia	200–400	Geothermal	Irrigation wells/[As(V)]*
Nigrita thermal waters	1,000–1,200	Geothermal	Spa
Kavala/Eleytheres	800	Geothermal	Spa
Loutraki Aridaias	350–450	Geothermal	Spa
Thermopyles	200–300	Geothermal	Spa
Epirus/Kavasila	120–160	Geothermal	Spa
Island of Kos/Kefalos	5–35	Geothermal	Drinking water/[As(III)]*
Aridaia Basin	5–30	Geothermal	Drinking waters
Aksios delta/Chalastra	35–40	Alluvial deposits	Drinking water/[As(III)]*
Aksios delta/Kumina	35–45	Alluvial deposits	Drinking water/[As(III)]*
Aksios delta/Malgara	20–27	Alluvial deposits	Drinking water/[As(III)]*
Aksios delta/Platy	20–30	Alluvial deposits	Drinking water/[As(III)]*
Nestos delta/Keramoti	15–20	Alluvial deposits	Drinking water [As(III)]*
Eastern Thessaly/Agia and Mpourmpoulithra	20–35	Mineralization	Underground water
	40–60	Mineralization	Spring water [As(V)]*
Kavala/Nikisiani	25–30	Mineralization	Drinking/spring water
Polykastro Kilkis	20–40	Mineralization	Drinking/spring water

Notes: Arsenic concentrations in this table have been measured by our group several times over the last decade. The measurements have been performed by atomic absorption spectrometry coupled with graphite furnace. Arsenic speciation has been performed by passing the samples through appropriate ion-exchange resins prior to measurement.
*Dominant arsenic species.

2,000 μg/L were recorded. The sources of As, released to the surface by geothermal fluids, may be related to igneous rocks underlying thick sedimentary formations. This assumption is also supported by As in correlation with K, Mn, and Fe, which indicates geogenic origin, while the presence of hydrothermal conditions resulted in enhancements of their mobility [35].

Noteworthy to report are the high arsenic concentrations in the thermal sea waters of Aghia Paraskevi (Kassandra peninsula, Western Chalkidiki), attributed to the dissolution of orpiment (As2S3), with arsenic content close to 61 wt.% [35]. Arsenic release due to geothermal activities is also found in areas of volcanic activity in South Aegean active volcanic arc [35]. This arc starts from the island of Methana and passing nearby the island of Milos and Santorini is finishing at the island of Kos. In particular, measurements of As concentrations in different types of sediments in the islands of Methana, Milos, Santorini, and Kos revealed elevated concentrations of arsenic in sediments, in concentrations as high as 20.5 mg/kg at Methana island, 108 mg/kg at Milos island, 356 mg/kg at Yali island, 493 mg/kg at Santorini island, and 56 mg/kg at Kos island at the location of Kefalos, where an arsenic removal treatment plant has been installed.

3.2. *Arsenic release in the areas of alluvial deposits due to reducing conditions*

Arsenic concentrations in groundwater into alluvial deposits are high in the areas of Aksios, Strymonas, and Nestos rivers in Northern Greece (Table 2). The groundwater characteristics are dominated by reducing conditions with negative redox potential values, high iron and manganese concentrations, and ammonia concentrations. The mechanism of arsenic release in these waters is, therefore, controlled by the so-called reductive dissolution of iron and manganese oxides [6]. More specifically, the adsorbed arsenic onto iron and manganese oxides of sediments along with iron and manganese is mobilized into the aqueous phase, polluting the groundwater aquifer, when reducing conditions prevail. This mechanism is generally accepted in sedimentary environments as a key process for the release of As into groundwater. While in the geothermal groundwater of Chalkidiki region, pentavalent arsenic dominates, in the alluvial deposits of Aksios area, arsenic is mainly present as trivalent arsenic mostly as a result of prevalent reducing conditions [6].

4. Applied treatment technologies in full-scale plants in Greece

Arsenic removal from groundwater is mainly accomplished by chemical coagulation using ferric or aluminum salts, adsorption on specific adsorption media based on iron or aluminum oxides, lime softening, reverse osmosis, and use of zero valent iron [36]. When arsenic is present in trivalent form (i.e. arsenite), then a pre-oxidation step such as the use of biological oxidation, ozonation, chlorination or manganese oxides must be incorporated in the treatment process [36]. Oxidation of As(III) to As(V) enhances the overall removal efficiency, because uncharged As(III) is not efficiently removed by the application of all treatment methods [37,38].

In Greece, treatment measures have been taken, in order to provide the population with drinking water containing arsenic concentration less than 10 μg/L. The treatment technologies, which have been applied, were greatly dependent on the environmental conditions and arsenic speciation in the affected groundwater. Information has been drawn from the works of Tresintsi et al. [39] and Katsoyiannis et al. [40]. In the deltaic area of Aksios, arsenic is mainly present in the trivalent form, due to the reducing conditions prevailing in the groundwater. Treatment units have been constructed and placed in operation in the cities of Malgara, Kumina, and Vrachia. These units consist of biological oxidation for the removal of iron, manganese, and ammonia by practicing oxygen, resulting also in As(III) oxidation. Then, As(V) is removed by chemical precipitation with $FeClSO_4$ for the cases of Kumina and Malgara [41] and by adsorption on granular ferric hydroxide (GFH) for the case of Vrachia. Similar to Vrachia treatment plant, biological oxidation followed by the adsorption on GFH was applied in Mitrousi in Serres region (Strymon river basin) with initial arsenic concentration ranging between 10 and 20 μg/L, mostly As(III).

In the village of Triglia, iron, manganese, and ammonia are in negligible concentrations, due to relatively oxidative conditions prevailing in the local groundwater. Thus, oxidation of arsenic was performed by ozonation and arsenic is effectively removed by chemical precipitation with $FeClSO_4$. Arsenic is also removed by chemical precipitation with $FeClSO_4$ from the drinking water of Daidalos in Kos Island after the biological treatment of iron, nitrite, and As(III) oxidation.

In contrast, in the area of mountain Pagaion, close to ancient Macedonian's mines, arsenic content from the local spring water, serving as drinking water for the municipality of Nikisiani, is directly removed by adsorption onto AquAszero [39], because arsenic is mainly in the pentavalent form and oxidation is not required. Similarly, no oxidation step is applied at the treatment unit of Mpourmpoulithra spring water (Melivoia). In this case, arsenic was effectively adsorbed onto Bayoxide. However, Melivoia treatment plant has already been discontinued, because it failed to remove antimony despite the allegations of the Bayoxide producer. Table 3 presents a summary of water characteristics and treatment units, installed in Greece.

The results of Fig. 2 show the specific arsenic removal (Q_{10}) achieved in full-scale treatment plants at equilibrium concentration equal to drinking water regulation limit of 10 μg/L. The highest adsorption capacity was observed at the treatment plant of Triglia (59 mg As/g Fe), where chemical precipitation was applied at relatively low pH value of 7.1. In addition, Fig. 2 shows that the application of chemical precipitation results in almost an order of magnitude with higher Q_{10} values than the commercial adsorbents, such as GFH or Bayoxide. Another important observation is that the treatment pH significantly affects the efficiency of chemical precipitation as reflected by the values of specific arsenic removal. The Q_{10} value of 59 mg As/g Fe at treatment pH 7.1 determined in Triglia plant, proved almost double of that determined in Daidalos (26 mg As/g Fe) and Kymina plant (31 mg As/g Fe) achieved at treatment pH 7.7. In general, we observed that increasing the pH by 0.5 units, the q value was decreased by approximately 50%, being in agreement with relevant previous studies [36,41]. Conclusively, the adsorption capacity of solid adsorbents is generally lower than that of the freshly precipitated Fe(III) solution (Fig. 2). This significantly higher specific arsenic removal of *in situ*-formed (hydrous) ferric hydroxides is attributed to their extensive "surface area" and the short-chain polymers of $Fe(OH)_y^{z+}$, which favor surface complexes with arsenate oxy-anions [39].

The reagent cost of treatment plants implemented precipitation-sand filtration, including that of Triglia, was estimated to be lower than 0.01 €/m^3 contributing in around 10% to operational cost [39]. After that the operational cost as a sum of reagent, energy, labor, and depreciation cost of Daidalos, Kymina, and Malgara treatment plants, where the backwash water

Table 3
Arsenic removal from Greek groundwater by full-scale treatment plants

Municipality	Flow rate (m^3/h)	Pre-oxidation	Treatment[a]	Water (pH)	Total As (μg/L) (inlet)	As(III) (μg/L) (inlet)
Malgara	75	Biological	Chemical precipitation	7.9	18	13
Kumina	100	Biological	Chemical precipitation	7.7	44	26
Vrachia	30	Biological	GFH/adsorption	7.3	7–11	4–6
Mitrousi	50	Biological	GFH/adsorption	7.9	18	11
Triglia	100	Ozone	Chemical precipitation	7.1	208	21
Nikisiani	50	No treatment	AquAsZero/adsorption	7.3	27	4
Daidalos	20	Biological	Chemical precipitation	7.7	33	25
Melivoia	60	No treatment	Bayoxide/adsorption	7.6	41	4

[a] Iron sediments of all treatment plants which implemented chemical precipitation were separated by sand filtration.

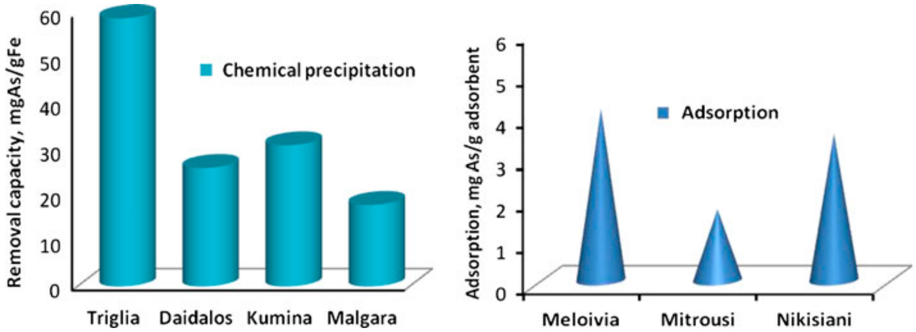

Fig. 2. Arsenic removal capacity (Q_{10}) of the treatment plants at equilibrium concentration of 10 μg/L. Note the different scale of Y axis. Adsorption process in Nikisiani is in progress and adsorption capacity is referred in equilibrium concentration 3 μg/L.

is directly disposed to the sewage, was similar and estimated to be around 0.09 ± 0.02 €/m^3. In contrast, Triglia's plant presented a higher operational cost of 0.14 ± 0.02 €/m^3, which attributed to handling of backwash water including thickening and dewatering of the sludge. The total operational cost at Mitrousi and Melivoia water treatment plants practicing arsenic adsorption was estimated to be around 0.16 ± 0.02 €/m^3. The cost of adsorbents count is around 0.09 ± 0.02 €/m^3, contributing thus, with more than 50% in operational cost. The lower initial arsenic concentration of Mitrousi water counterbalances the lower adsorption capacity of GFH, while Melivoia's treatment plant (Bayoxide) taking the advantage of gravity for water filtration, operates at almost no energy cost. The current operational cost (at equilibrium concentration 3 μg/L) of Nikisiani water treatment plant was estimated to be around 0.08 ± 0.02 €/m^3, which expected to diminish by the half due to implementation of two adsorption columns in series and to high arsenic adsorption capacity of AquAsZero, as well as due to gravity filtration of the water.

5. Conclusions

The present work has summarized the major arsenic-affected areas in European groundwater resources and has focused in describing the situation in Greece, reviewing the several relevant publications. The most notable cases in Greece are located in Northern Greece, in Central Macedonia, particularly in the areas of Aksios basin and Chalkidiki geothermal region, in the basin of Aridaia and in Eastern Thessaly. The paper described the treatment technologies that have been applied at full-scale level for the treatment of drinking water in the affected areas. The applied methods are based on the application of an oxidation step, mainly biological, for the removal of iron, manganese and ammonia resulting also in oxidation of As(III) to As(V), followed by either chemical precipitation using $FeClSO_4$ or by adsorption onto GFH, Bayoxide and AquAsZero. The treatment plants were very efficient with regard to arsenic removal, providing drinking water with arsenic concentration of less than 10 μg/L. The only case where the treatment was discontinued was the case of Mpourmpoulithra in Eastern Thessaly. This was not attributed to ineffective arsenic removal but because of the simultaneous presence of Sb, since Sb adsorption onto Bayoxide (the applied sorbent) did not prove to be an effective process for antimony removal.

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