

# Investigating Arsenic Contents in Surface and Drinking Water by Voltammetry and the Method of Standard Additions

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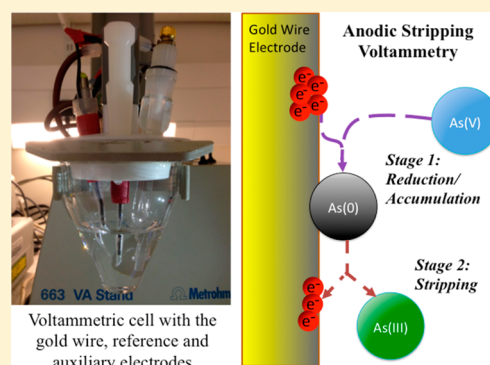
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## **S** Supporting Information

**ABSTRACT:** Testing water samples for arsenic contamination has become an important water quality issue worldwide. Arsenic usually occurs in very small concentrations, and a sensitive analytical method is needed. We present here a 1-day laboratory module developed to introduce Earth Sciences and/or Chemistry student undergraduates to key aspects of this topical issue. In this practical session, students were first introduced to the worldwide problems of arsenic contamination in groundwaters as a motivation of the experimental work. The latter consisted of quantification of arsenic levels in surface and drinking water at trace levels (nM, ppb) using the electroanalytical technique of anodic stripping voltammetry and the method of standard addition. Results were discussed with respect to water quality guidelines and geology. The complexity of data interpretation in this exercise can be tailored to a range of abilities and subject areas suited to the students and the course.

**KEYWORDS:** *Second-Year Undergraduate, Analytical Chemistry, Interdisciplinary/Multidisciplinary, Hands-On Learning/Manipulative, Problems Solving/Decision Making, Quantitative Analysis, Instrumental Methods, Geochemistry, Laboratory Equipment/Apparatus*



## ■ INTRODUCTION

Surprisingly, arsenic (As) and its compounds have been used for centuries as therapeutic drugs for a wide variety of symptoms.<sup>1</sup> Unfortunately, today arsenic is most notorious for its toxicity and for contaminating groundwater in many parts of the world.<sup>2</sup> In 2002, it was estimated that over 137 million people in more than 70 countries were living around As “hot spots”<sup>3,4</sup> (Figure 1). Bangladesh is one of the most affected countries where millions of people are confronted daily with highly contaminated drinking water.<sup>5</sup> Prolonged arsenic intake through drinking water or ingestion of contaminated food increases the risk of cancers, cardiovascular diseases, and neuropathy.<sup>6,7</sup> Arsenic contamination is occurring through anthropogenic activities such as mining or natural processes such as the dissolution of minerals and rocks containing As, hence reflecting the local geology.

In groundwater, inorganic forms of As predominate and consist of arsenate, the pentavalent form, and arsenite, the trivalent form (Figure 2). Arsenate is thermodynamically more stable in well-oxygenated environments, whereas arsenite is predominant in reducing conditions. These two forms have different mobilities in the environment and different toxicities to living organisms. Although exposure to all forms of arsenic has adverse effects on human health, inorganic As is more toxic than organic As, and arsenite is considered more toxic than

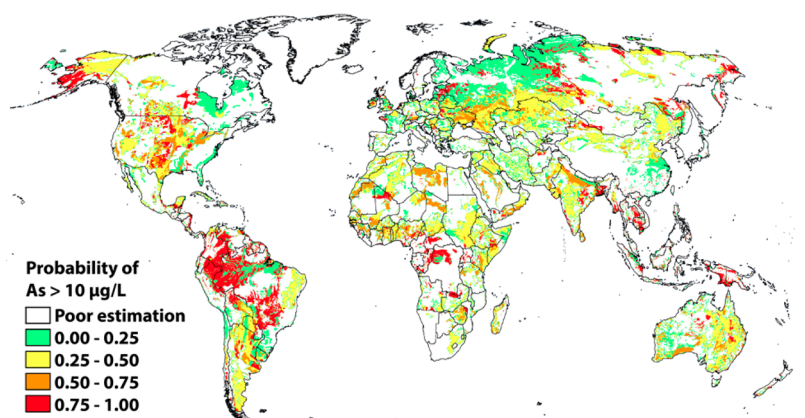
arsenate.<sup>9</sup> The individual determination of arsenic species is thus important but their separate reporting is not legally required. In view of the law, the legal limit of arsenic in water is currently 10 ppb in many countries around the world, although some have lower or higher limits, such as 5 ppb in Denmark<sup>10</sup> or 50 ppb in Bangladesh.<sup>5</sup> According to the World Health Organization (WHO), a limit of 10 ppb is currently what can be best achieved due to practical limitations of both removal of arsenic and analytical detection problems; this limit is not based on health considerations.<sup>11</sup> If that was the case, using the usual one in a million incremental cancer risks estimates for a carcinogenic substance, the limit of As in drinking water should be at 0.004 ppb.<sup>12</sup> Consequently, the 10 ppb limit is currently considered by WHO as a “guideline value” and is likely to decrease in the future, which will add further pressure on water companies to achieve both the required arsenic removal and the monitoring of lower levels.<sup>11</sup>

Either the analytical detection<sup>13</sup> or the removal<sup>14</sup> of arsenic in drinking water are current topics that are appealing to students due to the importance and current relevance of this issue. This laboratory-based experiment focuses on the

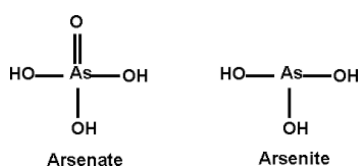
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**Figure 1.** A global map highlighting the spatial extent of areas with groundwater with geogenic (i.e., natural) arsenic concentrations of greater than 10 ppb (10  $\mu\text{g/L}$ ). Modified from Amini et al. (2008).<sup>8</sup>



**Figure 2.** Chemical structures of pentavalent arsenate and trivalent arsenite.

detection of arsenic in water samples and on the correlation of these arsenic levels with the geological environment from where the waters are sourced. Although arsenic is routinely detected in water by spectroscopic techniques such as ICP-MS, ICP-OES, or HG-AFS, the electrochemical method of anodic stripping voltammetry (ASV) is used here. The learning objectives of the experiment are 4-fold:

- (1) To introduce the worldwide issues of arsenic contamination in groundwater.
- (2) To present the basics of analytical chemistry at trace levels through the technique of stripping voltammetry.
- (3) To understand the standard addition method and its limitations and understand that any results carry an associated uncertainty given by the standard deviation and confidence intervals.
- (4) To critically assess experimental results with the local geology of the water sources as well as with health and safety limits.

This experimental work can be suited to students with different backgrounds, e.g., by highlighting the geological context to Chemistry students while focusing more on analytical techniques for Earth and Environmental students.

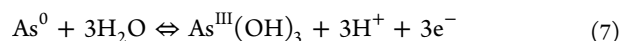
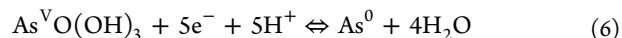
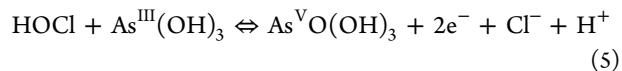
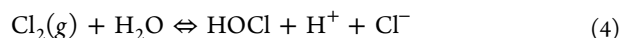
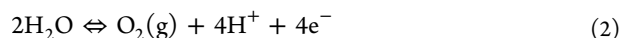
## ■ BACKGROUND

### Voltammetric Detection of Arsenic: Principles and Mechanisms

Anodic stripping voltammetry (ASV) is a versatile, low cost, and portable electroanalytical technique allowing the detection of total inorganic arsenic at a polycrystalline gold wire electrode<sup>15–17</sup> in acidified solutions ( $\sim 0.1$  M HCl). Because of the different sensitivities between arsenate and arsenite, arsenic must either be oxidized to the former or reduced to the latter prior to its determination. The oxidation is easier as well as faster and requires no addition of reagent. It is indirectly achieved in the voltammetric cell by imposing a low reductive

potential (e.g.,  $-1$  V) at the gold electrode.<sup>15</sup> At such potential, protons are reduced, and  $\text{H}_2$  gas is formed (eq 1), which is seen by the evolution of bubbles at the gold electrode. This reduction current must however be matched by an oxidation current at the auxiliary electrode; this current is mostly coming from the oxidation of water to oxygen (eq 2, as seen by  $\text{O}_2$  bubbles at the auxiliary electrode) and the oxidation of chloride to chlorine gas (eq 3). The latter reacts with water to form hypochlorous acid (eq 4). Both are strong oxidants, and they rapidly oxidize any arsenite present in the solution to arsenate (e.g., eq 5). The oxidation process is fast, taking only few seconds at pH 1. After that treatment, all inorganic arsenic is thus present as arsenate. The students were encouraged to look out for bubbles generated at both the auxiliary and working electrodes to directly visualize these reduction (eq 1) and oxidation (eq 2 and 3) processes.

The ASV measurement first consists of the reduction of arsenate to metallic arsenic  $\text{As}^0$  at the gold surface during the deposition step (eq 6) followed by the stripping step where  $\text{As}^0$  is oxidized back to the solution as arsenite (eq 7), the latter being oxidized chemically back to arsenate (eq 5; note that this reaction is written for acidic conditions). The peak current generated during the stripping step is directly correlated to the concentration of As in the solution. These equations were explained to the students during the experimental work.

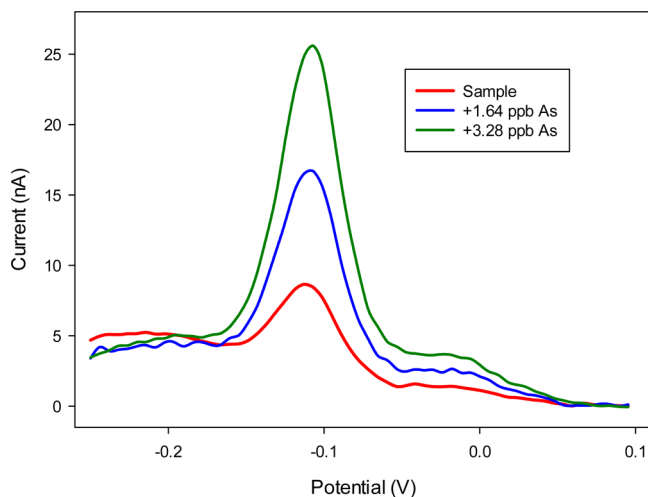


### Standard Addition Method

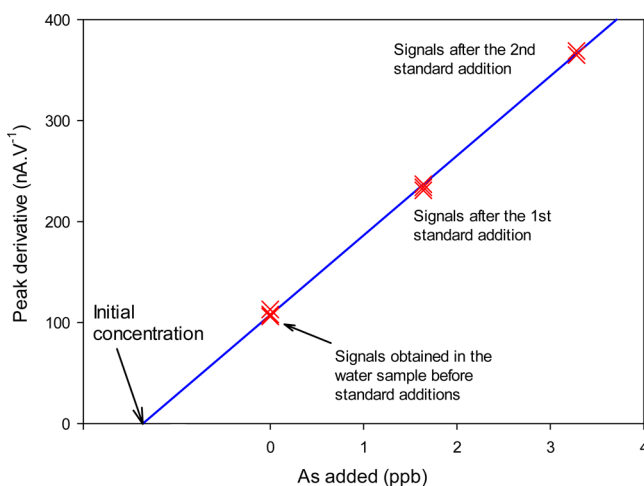
Arsenic concentrations were measured using the method of standard additions that inherently take into account any matrix interferences that could affect the sensitivity for arsenic. Students were explained the method and used an [Excel sheet](#)

(provided in the [Supporting Information](#)) to calculate the original concentration as given by the  $x$ -intercept, the standard deviation  $\sigma_x$  of the intercept,<sup>18</sup> and the 95% confidence intervals.

Studies where As concentrations are determined by the method of standard addition on a gold wire electrode<sup>15,16</sup> have highlighted that the linear range is small (e.g., up to approximately 3 ppb for 30 s deposition), and the signal starts to saturate thereafter. It is thus important that additions are made within the linear range of the method. [Figure 3](#) gives an example of voltammetric arsenic signals that were obtained by the students together with a typical standard addition plot ([Figure 4](#)).



**Figure 3.** Typical background-subtracted ASV signals of arsenic in acidified water before (red curve) and after standard additions.



**Figure 4.** Example of a typical standard addition curve. The absolute value of the  $x$ -intercept is the concentration of the analyte in the sample.

## EXPERIMENTS

### Instruments, Chemicals, and Samples

Instruments consist of a potentiostat, a stand to hold electrode in a voltammetric cell, a computer, and three electrodes: gold wire electrode (working), auxiliary, and reference electrodes. Chemicals were of analytical grade. Mineral waters were bought

from a local shop, and surface waters were collected from water bodies in London. All details are given in the [instructor notes](#) ([Supporting Information](#)).

### Voltammetric Detection of Arsenic

Arsenic detection was achieved by anodic stripping voltammetry similarly to that previously reported.<sup>15</sup> All voltammetric details are given in the [instructor notes](#) and in the [student handout](#) ([Supporting Information](#)).

### Recovery Experiment for Testing Accuracy

To test the accuracy of the method, a recovery experiment was first conducted by adding a known concentration of arsenic in an acidified As-free water and quantifying that amount by the method of standard addition. The experimentally determined concentration was then compared to the expected one using eq 8.

$$\begin{aligned} \text{\%recovery} &= \frac{\text{concentration determined by standard addition}}{\text{expected concentration}} \times 100 \\ &= \frac{\text{concentration determined by standard addition}}{\text{expected concentration}} \times 100 \end{aligned} \quad (8)$$

### Data Treatment and Analysis

Peak derivatives of the nine measurements (three repeats for the acidified sample in addition to three repeats for each of the two standard additions) were entered into the [Excel sheet](#) (see [Supporting Information](#)), and the concentration, standard deviation, and confidence intervals were determined. Prior to the experiments, students were encouraged to use the Excel sheet with data of an imaginary calibration curve (given in the student handout) and visualize the linear range and effect of the instability of the signal on the final result. The importance of the number of measurements on the standard deviation and confidence intervals was also highlighted. This exercise, run prior to the experimental work, helped the students to critically assess their results and suggests potential solutions for obtaining optimum results in terms of accuracy and low standard deviation.

As part of the exercise, the students used the data obtained in the recovery experiment to determine the concentration, standard deviation, confidence intervals, and accuracy of the technique. If time allows, the same exercise can include more standard additions to highlight the loss of linear range at higher concentrations. In any case, the students were made aware that best standard addition procedures are achieved when the final peak is up to twice the original peak and still within the linear range.

## HAZARDS

The students conduct a Health and Safety assessment for the exercise and COSHH forms are discussed in detail. Arsenic stock solutions (typically in the  $\mu\text{g}/\text{mL}$  range) are prepared by the instructor, and only standard solutions diluted down to the ppb level are handled by the student. HCl-concentrated solutions are diluted twice to  $\sim 6$  M before use by the students.

## RESULTS

Here, we are presenting data sets obtained by two different groups of Earth Science second and third year undergraduate students testing different water samples and a spiked sample during the class terms of 2013–2014 and 2014–2015. The students conducted the analytical work in the laboratory in the

morning (3 h, including introduction) and the data reduction and problem sets in the afternoon (3 h) in groups of three. The waters included over-the-bench available potable water and natural surface waters from London, previously collected by the students in acid-cleaned bottles. Group 1 analyzed the water brands A, B, C, and D (carbonated), and group 2 looked at surface water collected from the Thames River and Serpentine lake in central London.

Table 1 presents typical results obtained at the recovery experiment. Students were asked to calculate the original As

**Table 1. Recovery Experiment**

standard experiment	As recovered, $\pm\sigma$ , ppb <sup>a</sup>	95% confidence interval, ppb	recovery, %
peak derivative	1.88 $\pm$ 0.03	0.08	115
peak height	1.90 $\pm$ 0.07	0.17	116
peak area	2.07 $\pm$ 0.16	0.38	126

<sup>a</sup>Concentration of As found after two standard additions of 1.64 ppb As standard solution (3 blank measurements + 3 analyses for each addition); experiment conducted May 8, 2014. Representative student results.

concentration as well as the standard deviation (using the Excel file given in the Supporting Information) and the accuracy/recovery percentage (using eq 8) when using the peak height, the peak area, or the peak derivative to quantify the As peak intensities.

Both peak height and peak derivative here give similar As concentrations whereas that found from the peak area is significantly higher (Table 1). Recovery percentages were

found to vary significantly between 90 and 115% over the various groups that performed the experiment. Students were asked to think about the possible causes for such a response and to suggest ways of improving the measurement. Possible causes include standard additions outside the linear range, contamination in the background electrolyte, drifting of the sensor response during the procedure, and problems in choosing the peak baseline for both peak areas and peak height. Solutions to these issues include increasing the number of measurements per addition to ensure a stable signal, ensure minimum contamination by working cleanly with clean reagent, and use of the peak derivative to avoid any problems related to choice of the baseline. A stable and accurate experimental setup can be problematic to achieve and an error of 10–15% for these student systems is difficult to avoid. Nevertheless, the experimental work was a good formative exercise for students to appreciate some of the issues that analytical techniques may face.

Table 2 presents the results obtained by two different groups on two following days.

## DISCUSSION

### Do the Arsenic Concentrations Determined in the Waters Represent a Health Hazard?

After having determined the As concentrations and the errors associated with the determinations, the students discussed the measured concentrations with respect to the potential health impact. Limits on As concentrations in drinking water have been recommended by the WHO at 10 ppb.<sup>11</sup> All waters tested were well below the recommended limit (Table 2), and the

**Table 2. Arsenic Concentrations in Various Waters**

sample	day	As added standard, ppb	peak derivative ( $\times 10^{-7}$ A V <sup>-1</sup> ) <sup>a</sup>	As found, ppb	SD, ppb	RSD, %	95% confidence interval
standard (1.64 ppb)	1	0.00	1.36/1.36/1.36	1.88	0.03	1.6	0.08
		1.64	2.56/2.59/2.62				
		3.28	3.76/3.75/3.73				
	2	0.00	1.30/1.29/1.34	1.94	0.07	3.6	0.15
		1.64	2.53/2.46/2.46				
		3.28	3.49/3.57/3.60				
Thames river water	1	0.00	0.45/0.43/0.46	0.60	0.07	11.8	0.15
		1.64	1.65/1.55/1.51				
		3.28	2.91/2.79/2.71				
Serpentine lake water	1	0.00	1.06/1.13/1.08	1.36	0.04	2.9	0.09
		1.64	2.34/2.37/2.31				
		3.28	3.69/3.65/3.69				
water A	1	0.00	2.34/2.33/2.30	5.32	0.21	3.9	0.50
		1.64	2.92/2.96/2.98				
		3.28	3.78/3.73/3.70				
	2	0.00	2.47/2.45/2.50	5.99	0.33	5.5	0.78
		1.64	3.29/3.29/3.23				
		3.28	3.92/3.81/3.81				
water B	1	0.00	0.52/0.47/0.53	0.75	0.05	6.7	0.11
		1.64	1.47/1.50/1.53				
		3.28	2.60/2.64/2.65				
	2	0.00	0.51/0.50/0.49	0.69	0.04	5.8	0.09
		1.64	1.66/1.71/1.64				
		3.28	2.79/2.85/2.92				
water C	1, 2	0.00	no peak	BDL <sup>b</sup>	n.a.	n.a.	n.a.
water D	1, 2	0.00	no peak	BDL <sup>b</sup>	n.a.	n.a.	n.a.

<sup>a</sup>The peak derivative was used for the calculations. <sup>b</sup>BDL: below detection limit; concentration values were below the quantifiable limit (no peak was observed), and therefore data are not available (n.a.); SD: standard deviation.

students suggested they would not require additional As treatment if they were used for drinking water purposes. Two over-the-counter mineral waters, waters C and D (carbonated) had As concentrations below the detection limits of 0.5 ppb (determined using three times the standard deviation from the background). Water A had the highest As concentration with  $5.3 \pm 0.5$  ppb. This is still well below the guidelines set by the WHO but close to the 5 ppb limit now set by countries including Denmark and was flagged by the students as potentially a problematic water to drink.

### Investigating the Link between Arsenic Concentration in Drinking Water and the Geology from Water Source Regions

In the next step, the students were asked to test if the different As concentrations could be related to the lithologies from where the waters originate (the information was found by reading the company web pages or on the bottle etiquette).

From the samples analyzed during our laboratory course, water A had the highest As concentration. This water was the only one sourced from a volcanic region. The company web page indicated that the water is filtered through different layers of volcanic rocks, including a mixture of granites, basalts, ash, trachyandesites, and puzzolana before surfacing. Volcanic regions are associated with elevated As concentrations in waters as As becomes more soluble at the higher temperatures typically associated with hydrothermal systems.<sup>19</sup> Water B, with the lower As concentration in contrast, was sourced from an aquifer situated in an area of old, geologically inactive highly crystalline metamorphic geology, where there is an absence of (hydrothermal) heating. This means that As is not as soluble and so less is transported within the naturally filtered water, thus potentially explaining the low As concentrations seen. Waters C and D were mainly from Silurian limestone, silts, and sandstones; all these rock types are known for having very low As concentrations, which seems to be reflected in the analysis of the waters (no arsenic was detected).

The two natural surface waters taken in London featured low As concentrations, consistent with previous work suggesting that surface waters have low As concentrations of generally less than 10 ppb.<sup>20</sup> A possible mechanism explaining the low concentrations was proposed by the students following the lecture that focused on the review paper on As geochemistry in aquifers.<sup>21</sup> The higher oxygen concentrations found in surface waters result in the complete oxidation of the arsenic. Arsenate adsorbs more strongly onto iron minerals and therefore removes the As in the surface water.<sup>21,22</sup> A possible way to test this hypothesis would also be to analyze the solid particulates in the surface waters (using the material collected after filtration).

### CONCLUSIONS

In this laboratory experiment, the students were introduced to the topics of arsenic contamination and water quality and of analytical chemistry. Stripping voltammetry was used to detect trace level concentrations of As in natural and drinking water samples. Students learned about the standard addition method and became acquainted with notions of, e.g., linear range, recovery, and confidence intervals.

Results obtained by the students showed that As concentrations vary in different water samples. They compared the obtained concentrations against the health and safety limits set by WHO and for individual countries. All samples were

within the 10 ppb limit set by the World Health Organization,<sup>8</sup> but some were at the legal limits imposed in some countries.

Local geology of water sources were used to explain the differences in As concentrations across the water samples. Active volcanic regions with high temperature hydrothermal fluids give rise to relatively high As concentrations in the waters that filter through them, whereas sandstones generally contain very low As concentrations. In surface river systems and lakes, oxidation and adsorption of As(V) onto sinking particles could be the reason for their low As concentrations.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available on the ACS Publications website at DOI: 10.1021/acs.jchemed.6b00025.

Student handout for the determination of concentrations, standard deviations and confidence intervals upon standard additions (PDF, DOCX)

Excel sheet for the determination of concentrations, standard deviations and confidence intervals upon standard additions (XLSX)

Instructor notes for the determination of concentrations, standard deviations and confidence intervals upon standard additions (PDF, DOCX)

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#### Notes

The authors declare no competing financial interest.

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