

Extraction of arsenic species from airborne particulate filters— Application to an industrial area of Greece

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Abstract

In the present study, the extraction of the arsenic species arsenite (As(III)), arsenate (As(V)), monomethylarsonic (MMA) and dimethylarsinic acid (DMA) from airborne particulate filters was investigated and optimized. For this purpose, total suspended particulate matter as well as size fractionated aerosol samples were collected from the industrial area of Aspropyrgos, Greece, in glass fibre and polycarbonated filters, respectively. Among H_3PO_4 and HCl, tested in various concentrations, concentrated HCl was found to be the most effective extractant for arsenic from both polycarbonated and glass fibre filters, without provoking any arsenic species transformation. However, the quantitative extraction of arsenic species from glass fibre filters required the subsequent washing of the filters with ultrapure water after their leaching with concentrated HCl. The developed procedure was applied to airborne particulate filters for arsenic speciation in Aspropyrgos' atmosphere. The results showed an enrichment of As in the fine ($\text{PM}_{2.5}$) compared with the coarse ($\text{PM}_{10-2.5}$) fraction of airborne particulates, while As(V) was found to be the predominant arsenic species in all samples. Finally, As concentration in the PM_{10} fraction, for the investigated area and time period from December 2004 to June 2006, was below the target value of 6 ng As m^{-3} , referred in the Directive 2004/107 of European Union.

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1. Introduction

The investigation of toxic species in airborne particulate matter has been greatly associated with atmospheric pollution research. Various studies have concluded that atmospheric particles with an aerodynamic diameter less than $10 \mu\text{m}$ (PM_{10}) and, mainly less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) are associated with a variety of toxic effects to human health [1], which are highly dependant on the aerosols' chemical composition [2]. Arsenic (As), occurring in the atmosphere as gaseous compounds and particulate phase [2], is a carcinogenic environmental toxin [2–4], contributed mainly by human activities, such as mining, operation of indus-

trial units and use of fuels [5,6]. The harmful effect of As to human health [6–8] led recently European Union to involve it in the Directive 2004/107/EC, which established a target value of 6 ng As m^{-3} in PM_{10} [9]. However, the toxicity and the physicochemical properties of arsenic are highly dependant on its chemical form; inorganic arsenic species, arsenite (As(III)) and arsenate (As(V)) exhibit high toxicity having been classified as carcinogenic agents [5,6,8], while their methylated derivatives, monomethylarsonic (MMA) and dimethylarsinic acids (DMA) are less toxic [5]. Therefore, arsenic speciation in aerosols is a significant tool for the evaluation of risk in population's health caused by respirable particles as well as for the identification of possible emission sources [6]. However, the common practice is the determination of the whole arsenic in total suspended particulates or fractionated airborne particulate matter [2,10,11], while arsenic speciation studies have been scarcely applied even in small-scale environmental studies [12,13]. Thus, limited

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leaching procedures have been developed for the quantitative extraction of the individual As species from airborne particulate filters.

The development of an effective procedure for the extraction of the 4 major in the environment arsenic species, As(III), As(V), MMA and DMA from airborne particulate filters was set as a major goal of the present study. For this purpose, the industrial area of Aspropyrgos–Elefsis, part of the western region of Attica called “Thriassion Plain”, was selected as a case study as it combines urban with industrial pollution. Indeed, high arsenic concentration is expected in Thriassion Plain’s atmosphere, as it is heavily polluted by a wide range of industries, including refineries, cement, steel and other smaller industrial units, while the presence of 2 national roads exacerbates its atmospheric pollution [14]. It should be mentioned that this case may have another aspect of interest referring to the possible effects of respirable particles to human’s health with respect to the rapid increase of Thriassion Plain’s population in the last decade [14]. For the present study, total suspended as well as fractionated airborne particulates were collected from Aspropyrgos’ atmosphere by means of a Gent stacked sampler in glass fibre and polycarbonated filters, respectively. The extraction of the arsenic species As(III), As(V), MMA and DMA was investigated and optimized using HCl and H₃PO₄ in a wide concentration range from 0.1 to 9.5 M. The detection of the individual arsenic species was carried out using hydride generation inductively coupled plasma-optical emission spectrometry (HG-ICP-OES). The developed procedure was applied for As speciation in aerosol samples collected from Aspropyrgos’ atmosphere in December of 2004 and 2005 as well as June of 2005 and 2006, with a sampling rate of 4 campaigns/month.

2. Experimental

2.1. Sample collection

Sampling of airborne particulate matter was carried out at an atmospheric monitoring station of the Association for the Development of the Thriassion Plain in Aspropyrgos, in the vicinity of 2 refineries and the national road Athens–Corinth. The airborne particulate sampling station along with the different possible atmospheric pollution sources are depicted in Fig. 1.

Total suspended particulate matter was collected in glass fibre filters using a high volume sampler with about 1600 l min⁻¹ sampling rate, employed for a period of 24 h. Size fractionated aerosol samples were collected by means of a Gent-type stacked filter unit [15–17], using a low volume (17 l min⁻¹) sampler. Both high and low volume samplers were installed at a height of 3 m above ground level. As preliminary studies had shown, the amount of fractionated aerosol samples within 24 h in that area was less than 1 mg and, therefore, the duration of each sampling period was prolonged to 48 h in order to collect enough airborne particulate sample for the analysis. The separation of aerosol particles into two size fractions was achieved by subsequential filtration through two Nucleopore track-etch polycarbonated filters (Whatman) with a diameter of 47 mm each, having

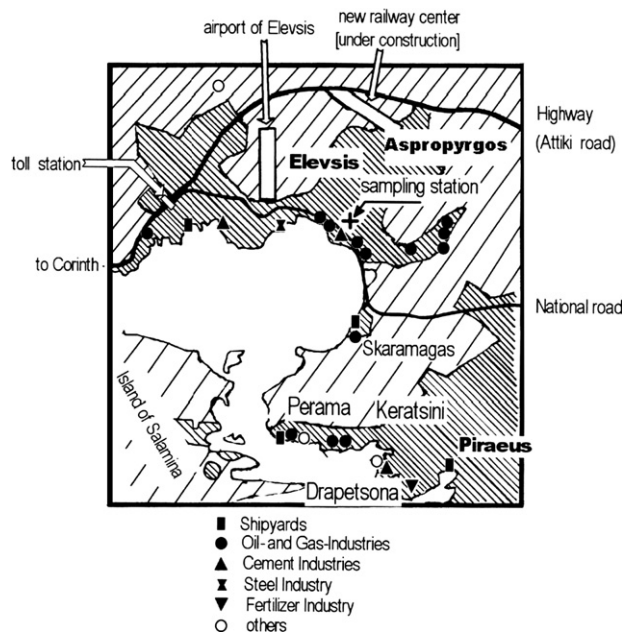


Fig. 1. Map of the industrial area of Aspropyrgos/Elefsis along with possible pollution sources and the sampling station used for this work.

different pore sizes. The first filter with 8 μm pore size (coarse) and the second with 0.4 μm (fine) were placed in a stacked filter cassette equipped with an upper-size inlet cut-off. The coarse filter collects particles with an aerodynamic diameter between 2 and 10 μm (PM_{10–2.5}) and the second particles < 2 μm (PM_{2.5}).

2.2. Instrumentation

The determination of the total As, after appropriate digestion procedure, as well as the determination of the individual As species As(III), As(V), MMA and DMA, after their separation, was carried out using HG-ICP-OES (Jobin Yvon, Model Ultrace JY 138, wavelength: 193.759 nm). The parameters were: Power: 1000 W, cooling gas flow (Ar): 12 l min⁻¹, sheath gas flow (Ar): 0.4 l min⁻¹ and sample uptake: 1 ml min⁻¹. A solution 1% of NaBH₄ in 1 M NaOH in high purity water (HPW) (Millipore, Simplicity) was necessary for the hydride generation, with an uptake of 0.5 ml min⁻¹. For the acidic digestion of the samples prior to the determination of their total As content, a PTFE autoclave (Perkin-Elmer 3) was used.

For the separation of As(V), MMA and DMA the anion and cation-exchanger AG 1-X8 and Dowex 50 WX8, respectively, were packed into glass columns in a resin bed volume of 15 ml each. The solutions were passed through ion-exchangers with a flow rate of 2 ml min⁻¹ using a peristaltic pump (Ismatec). The latter pump was also used in the same flow rate for the elution of the two resins.

2.3. Reagents

All reagents used were of analytical grade if otherwise is mentioned. Stock solutions in the range of 1000 mg As l⁻¹

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