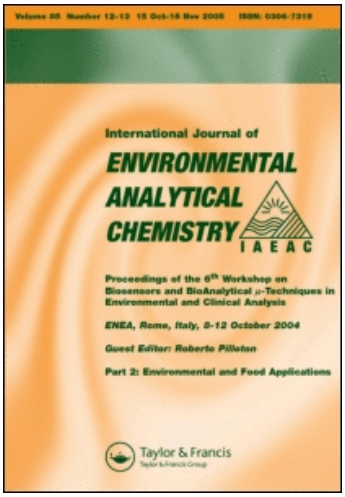

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Analytical Methods for Mercury Speciation in Environmental and Biological Samples - An Overview

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ANALYTICAL METHODS FOR MERCURY SPECIATION IN ENVIRONMENTAL AND BIOLOGICAL SAMPLES – AN OVERVIEW

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A comparison of the methods described for mercury speciation is presented. These methods are classified into three groups: 1) methods without chromatographic separations, 2) methods based on gas chromatography, and 3) methods based on the use of liquid chromatography. The most important methods used in each of these groups are described and compared.

Keywords: Mercury; organomercury compounds; atomic spectroscopy; chromatography

INTRODUCTION

Mercury is a highly toxic element that may reach the environment from natural geological deposits or from man-made industrial sources. The last ten years have seen an enormous progress in the development of highly-sensitive methods for mercury speciation in environmental and bio-medical research.

The distribution of mercury species in the marine environment is particularly interesting for analytical chemists. In the sea, mercury species often undergo biomethylation [1] to form highly-toxic free methyl mercury compounds that easily enter into marine food chains [2]. They accumulate in certain marine filter feeder organisms and appear later in the fatty tissues of the fishes that feed on them.

The development of methods to analyse trace amounts of mercury species in the different environmental compartments requires enormous efforts to avoid

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biased results [3]. Some years ago analysts had no reference materials to validate their results. It was particularly difficult to clearly differentiate between the approaches of those analytical procedures that identify and quantify different species from those by which the analyst defines the type of species to be measured. In many cases, a method proved too insensitive to analyse a species in biological and environmental samples.

Most of the methods are aimed to distinguish between inorganic mercury and methyl mercury, however, sometimes other compounds may be present, like ethyl mercury (EtHg), dimethyl mercury (DiMeHg) and phenyl mercury (PhHg). Most of the established methods have been applied to natural waters and to biological samples, particularly marine organisms.

In general, the analytical methods used for speciation of mercury species in environmental or biological samples include different sequential steps: a) extraction and preconcentration (if necessary) to attain a final concentration greater than the determination limit, b) separation of inorganic and organic species, and c) determination of the species. The extraction step is critical and must be performed carefully.

Preservation of aqueous samples

There is a consensus on the positive effects of low pH and high ionic strength to prevent mercury oxidation and complexation [4] as well as cation deposition on the container walls and conversion into the inorganic form [5]. Mineral acids are the most common reagents added to water in order to maintain unaltered mercury species [6,7]. Some preservative agents are added sometimes if the analysis must be delayed; preservative substances proposed include humic acids [8] and potassium dichromate [7]in acid media; freezing in liquid nitrogen has been recommended also [9]. On the other hand, the effect of container materials has been described in the literature; PTFE containers are preferred to PVC or glass made flasks [3]. Usually, the containers are cleaned previously with nitric acid.

Extraction of mercury compounds from gaseous and solid samples

The procedure most frequently used for air samples involves pumping air through a series of columns each containing a species selective sorbent: a glass wool for the removal of particulate forms; Ag-coated glass beads for the removal of Hg^0, and Chromosorb-W columns for the retention of Hg(II) and CH_3Hg^+species [10]. Other trapping systems have been developed with satisfactory results [11–14].

The extraction of solid samples (biota and sediments) requires a great deal of care in order to preserve the original speciation. The treatments commonly used are:

a) Acid treatment witn halogenated acids combined with solvent extraction. In general, hydrochloric acid and benzene are currently used [15]. Iodoacetic acid has been proposed [51], as well as NaCl [16,17] and KBr [18–20] in mineral acid medium. Due to the carcinogenicity of benzene, toluene [21,22], chloroform[18,23] and dichloromethane [9,24] have been proposed.

b) Alkaline digestion with KOH-methanol [9,25,26] and NaOH-cysteine [37] and extraction, although, in this case, high levels of organic matter, sulphides and diverse metal ions are also coextracted [27].

c) Treatment with a solution of tetramethylammonium hydroxide (TMAH) [40]. TMAH is an alkaline tissue solubilizer, and has been use for the speciation of tin and mercury. Although the resulting samples digest is not clear and colourless, the ease of sample preparation using this method can be a distinct advantage over other conventional procedures.

Analytical methods for mercury speciation

Non-cromatographic methods

These generally make use of cold-vapour atomic absorption spectrometry (CVAAS) that measures the mercury peak at 253.7 nm. This is the usual method to determine total mercury and exhibits very high sensitivity, although cold vapour atomic fluorescence (CVAFS) or inductively coupled plasma-mass spectrometry (ICP-MS) have also been used.

For mercury speciation an additional step is needed to selectively reduce at least some mercury species. Basically, the differential behavior of Hg(II) and CH_3Hg^+ versus reducing agents can be used to achieve a simple speciation scheme (i.e. while Hg(II) is reduced to Hg^0 by $SnCl_2$, the methylmercury species is not). However, selective reduction cannot differentiate between the different organic species of mercury, but only between inorganic and organic mercury.

Within these methods there are several forms of quantitation: A) those that determine organic mercury by difference; B) those that determine directly inorganic or organic mercury; and C) those that selectively fix the mercury compounds prior to analysis (this generally applies to air samples or gaseous samples).

A) Methods to determine organic mercury by difference

A1. Inorganic mercury is determined by reduction with $SnCl_2$ under determined conditions, while total mercury is usually determined by decomposing beforehand all the organomercury species to inorganic mercury which, together with that present previously, is reduced by Sn (II) and determined by the cold-vapour technique. The content of organomercury is determined by difference [28]. Oxidative digestion [29-31] or even ultra-violet light radiation [32,33] may be used to decompose the organomercury species.

A2. The selective reduction of inorganic mercury and total mercury proposed by Magos [34] is widely used. This yields cold mercury vapour that is detected and measured by AAS. Selective reduction uses $SnCl_2$ and $SnCl_2$+ $CdCl_2$ to determine, respectively, inorganic and total mercury. The differences between total mercury and inorganic mercury correspond to the content of MeHg in the sample. This procedure can be successfully applied to biological samples or other types of samples where the concentration of mercury may be toxicologically important. However, the Magos selective-reduction method is not sensitive enough to measure satisfactorily the concentrations of inorganic and organic mercury in natural waters such as rainwater, fogs, ice or snow, and lake, river and ground waters.

A3. A procedure introduced to improve the Magos method, particularly its sensitivity, selectively reduces mercury with BrCl + $SnCl_2$ and uses bromine monochloride as an oxidising agent for methyl mercury before the $SnCl_2$ reducing agent is added to determine total mercury. The use of $SnCl_2$ alone determines only inorganic mercury and the reduced mercury is amalgamated and thermally desorbed to be detected by AAS or AES [35,36].

Although these procedures are generally simple they have the drawback of the lack of specificity and accuracy.

B) Methods that determine directly inorganic and organic mercury

B1. The first procedure uses selective reduction by $SnCl_2$ and sodium borohydride (Na BH_4). The stannous chloride is used first to reduce only the inorganic mercury in the sample. Next, sodium borohydride is injected into the reduction cell to reduce the organomercury [37]. The resultant elemental mercury in each step is swept into nitrogen and conveyed to a test cell where its absorbance is measured at 253.7 nm. The detection limit ranges from 0.003 to 0.005 ng/ml.

B2. More information about speciation is given by the method described by Goulden [38]. He uses an autoanalyser of segments of flow into which aliquotes of the solution to be tested are injected, so that it can react with three different reagents (I, II, III). Reagent I is aqueous EDTA and together with hydroxylamine in an alkaline solution reduces inorganic mercury to mercury. Reagent mixture II

is EDTA + $SnCl_2$ that reduces inorganic mercury and aryl compunds like phenyl mercury (II) to mercury. Reagent mixture III is $CdCl_2$ + $SnCl_2$ and this reduces to elemental mercury all mercury compounds, MeHg included.

All these reactions take place in adequately-heated reaction coils and air is passed through them to take away the elemental mercury. The air separates the liquid which is then extracted by a phase separator. Subsequently, the gas stream with its mercury vapour content is conveyed to the combustion furnace in which all organic material is destroyed. The gas stream is then dried with sulphuric acid in the cooled, packed column and passed to the mercury monitor to measure its atomic absorbance. The reported detection limit for total mercury is 0.001 ng/ml.

B3. A very simple method to determine methyl mercury in fish samples is based on the extraction of organically-bound MeHg as bromide by chloroform and then determining it directly by CVAAS before reducing the extracted MeHg with a solution of $NaBH_4$. The inorganic mercury is determined in the residual aqueous phase by reduction with $NaBH_4$. Tests of many samples have shown that the sum of the values of MeHg and inorganic mercury is the same as that of mercury obtained by CVAAS of aliquotes of the same samples after acid digestion. The detection limit reported for Me Hg is 25 ng/g of dry sample [18].

B4. A non-chromatographic procedure used to speciate mercury is based on determinations by flow injection CVAFS. The procedure uses a microcolumn filled with sulphydryl cotton that retains only the methyl mercury -the inorganic mercury is reduced by $SnCl_2$ and is determined by atomic fluorescence. The MeHg is then eluted from the column by hydrochloric acid, oxidised by Br^--BrO^{-3} solution and reduced by $SnCl_2$ before determining mercury as above. The reported detection limit for MeHg is 0.006 ng/ml [39].

In addition to these selective reduction procedures, a method that employs electrothermic vaporisation has been recently described. The sample is dissolved in a solution and two aliquotes are removed. The first sample is heated strongly and the total mercury expelled is removed by an argon flow and measured by ICP-MS. The methyl mercury in the second sample is extracted as MeHgI and removed by an argon flow, the inorganic mercury that remains is then determined by the same technique to calculate the MeHg by difference [40].

In general, these procedures show interesting possibilities, although literature is scarce. The sensivities when CVAAS is used are excellent. However, the selectivity in general is not satisfactory, specially in comparison with those methods that uses chromatographic separations.

C) Methods with preliminary selective fixation of mercuric compounds

The most representative is that developed by Schroeder and Jackson for atmospheric samples [41]. The air, or any other gas, is passed through several different

collectors that recover different mercuric compounds: Chrom W (in HCl) for $HgCl_2$, Tenax GC for CH_3HgCl, Carbosieve B for $(CH_3)_2Hg$, and gold (wire) for Hg^0. A flow of argon-nitrogen is then passed through the collectors and the outflow is subjected to an appropriate thermal treatment to desorb the mercury and to convey the different compounds previously retained to a pyroliser where they are decomposed. The mercury formed is retained by a gold filament that is heated to 450° C to expel the mercury which is then removed to be measured by either atomic absorption or atomic fluorescence. However, there is no reliable information about the practical use of this method.

Gas chromatography methods

Gas chromatography (GC) is probably the most frequently used technique to measure organomercuric compounds. The most important characteristics of these procedures were reviewed some years ago [42]. Most of the stationary phases used to separate mercury compounds are polar like poly (ethyleneglycol) succinate [PEGS], butanediol succinate [HI-EFF-4BP], diethylelyglycol adipate [DEGA], diethyleneglycol[DEGS], and poly(ethyleneglycol)[CARBOWAX 20 M].

The principal advantage of GC methods to speciate mercury compounds is that the values they give are direct measurements. In addition, in many cases, individual organomercury compounds can be measured. The principal disadvantage is that it is usually necessary to carry out derivatisation of the inorganic and organic mercury because the low volatility and rather unstable nature of these compounds in the chromatographic columns complicates and prolongs the measurement process. The three derivatisations most frequently used are: 1) butylation with Grignard's reagent (butylmagnesium chloride in tetrahydrofurane;2) formation of hydrides with $NaBH_4$; and, 3) ethylation with $NaBEt_4$.

There are three principal detection systems mostly used with gas chromatography: A) electron capture devices (ECD), B) atomic spectroscopy (AS) detectors and C) coupling of GC to mass spectrometry (MS).

A. Electron Capture Devices

There are two major problems inherent in the use of GC-ECD for mercury speciation. The first results from the use of a non-selective detector, which responds to the halide moiety in the organo-mercury halide species, but also to any other electron-capturing species in the injected sample. Therefore, an extensive, three-stage extraction, clean-up procedure is necessary to reduce potential interferences, although this is time-consuming, labour intensive and typically results

in <90% extraction efficiencies [43–45]. This procedure will not allow inorganic mercury to be determined unless a reagent is subsequently added to form, for example, the methyl derivative. The second problem lies in the difficulty in successfully chromatographing the organomercurial halides. In the columns currently used (5 % DEGS-PS on Supelcoport), the retention times of the organomercury compounds increase with time, whereas the peak areas decrease due to the disturbance of the stationary phase [46]. In spite of this, the methods were used extensively until they began to be combined with GC and AS.

A1. In the now clasical method of Westöö [47] that determines methyl mercury, but not inorganic mercury, benzene or toluene are used to extract the methyl mercury as the chloride (or bromide) from a sample treated with HCl (or HBr). The extract is then back-extracted by a thiol type compound (cysteine, glutathione) dissolved in water in which the methyl mercury is extracted and a subsequent extraction is carried out with toluene or benzene. Finally, the methyl mercury is measured by chromatography. The limit of detection is 0.1 ng/ml. To avoid the formation of emulsions during the extraction processes a cysteine-impregnated paper is used that functions like the cysteine aqueous solution. The inorganic mercury is calculated by difference after total mercury is measured by AS [48].

A2. A variant of the previous method consists in the extraction of the organic mercury as chloride into benzene by the process previously described, and then to measure it chromatographically with an ECD. The inorganic mercury that remains in the sample is methylated with trimethyl tin and the derivative is extracted in benzene and reextracted by a thiosulphate solution to determine inmediately this mercury by atomic absorption in a graphite oven [49].

A3. It has been suggested that methyl mercury could be determined by selective retention it in a microcolumn of cotton wool impregnated with a mixture of mercaptoacetic acetic-acid anhydride-acetic acid-sulphuric acid. The adsorbed methyl mercury is later extracted with benzene and then measured by GC-ECD. Total mercury is determined by AS [50].

A4. Recently, a method has been described for the extraction-determination of methyl mercury in marine sediments, based on the quantitative microwave-assisted extraction of this compound using hydrochloric acid and tolune as solvents, and subsequent determination by GC-ECD. The detection limit was 8 ng/g of sediment [51].

B. MIP-AES detectors or other atomic techniques

Most of the developed MIP-AES procedures use microwave induced plasma and they offer certain advantages.

B1. The general procedure for GC separation and MIP-AES detection involves an adequate extraction and previous treatment of the compounds according to the

Westöö method. The procedure shows great sensitivity, with a quantification limit between 1.7 to 3.0 ng/ml. A great number of biological and environmental samples have been analysed in this way [16, 52–55].

B2. In one particularly interesting procedure, initially applied to blood samples, mercury (organic and inorganic) is extracted in toluene as complexes of diethyldithiocarbamates before butylating with Grignard's reagent in a test-tube. Once the excess reagent is decomposed, an aliquote is injected into the cromatograph using a non-polar column. The detector uses MIP-AES. The reported detection limit is 0.4 ng/ml [56].

B3. The authors of the above MIP-AES procedure modified it by extracting the methyl mercury and inorganic mercury complexes as diethyldithiocarbamates by retention in a dithiocarbamate resin packed in a miniature 60 µl column in a closed and semi-automated flow injection system. The quantitively enriched mercury species on the resin are completely eluted with an acidic thiourea solution, extracted into toluene, butylated and measured with the same GC-MIP-AES detector [57]. Because of the pre-concentration, the relative detection limits are rather good: 0.005 pg/ml for methyl mercury and 0.15 pg/ml for inorganic mercury. The procedure is applied satisfactorily to a wide variety of biological and environmental samples [58], and has been applied also to the determination of mercury in waters with high concentrations of humic acids; the detection limits found are 0.04 and 0.28 pg/ml for MeHg and inorganic Hg, respectively [59]. Recently, the same research group have developed diverse modifications of these procedure to improve the applicability. On the other hand, an on-line amalgamation trap for the collection of mercury species separated previously by GC before its determination with MIP-AES has been devised [60]. For direct measurement of the column eluate, the detection limits for mercury species in natural gas condensate is elevated because of background interference from carbon compounds passing to the plasma at the same time; carbon compounds give rise to emission that spectrally interferes with the signal from the mercury detector and can overload the plasma discharge, reducing the excitation capability. With an amalgamation trap (a gold wire in a heater coil), mercury can be selectively collected from the column eluate and subsequently passed to the plasma in a flow of pure helium. With this device, the detection limit of the derivatized (butylated) monomethyl and inorganic mercury is 0.56 ng/ml.

B4. Another method uses two gas chromatographs in series: the first uses a packed column, and the second, a capillary column. Both columns are connected by a capillary transfer line, heated at 150 °C, that ends in a needle that goes to the injector of the second cromatograph. Detection is carried out by MIP-AES. The larger sample capacity of the packed pre-column permits the selective transfer of the mercury analytes to a capillary gas chromatography system in which they can

be focussed and separated further on the analytical column. This process has a double advantage: it minimises the risks of extinguishing the plasma by an excesst of solvent that reaches the MIP; and reduces the fouling of the stationary phase of the detector that occurs when large-volume injections are used with other methods [61].

The benefits and limitations of this approach to the determination of mercury species are well-illustrated by its application to the detection and measurement of mercury species in natural waters when Grignard's derivatisation is used after solid-phase extraction on dithiocarbamate resin, followed by elution with acid thiourea, and complexometric extraction into hexane. The relative detection limit reported for this method (denoted as GC-GC-MIP-AES) for methyl mercury is 0.008 pg/ml for an injection of 50 µl. Diverse procedures based on the separation of organic and inorganioc Hg by GC and detection by CVAFS are described also [9,62].

B4. A described procedure that uses CVAFS to determine methyl mercury differentiates the samples with KOH-methanol. The mercury is extracted with methyl chloride passed to aqueous medium and there ethylated. Then, the GC-CVAFS technique is applied. Total mercury is measured by CVAAS and the inorganic mercury is calculated by difference [63].

B5. In recent years, GC has been proposed in combination with ICP-MS. Besides the conventional capillary GC [64,65], the use of a sylanized quartz tube packed with a chromatographic sorbent which has been cooled with liquid nitrogen and heated electrically [66] in order to desorb the analytes, has been the most popular. Both these approaches suffer from several drawbacks.

The use of capillary GC entails the need for a regular (rather bulky) chromatographic oven with temperature gradient programming. The sample should be injected in a volatile organic solvent which interferes with more volatile analytes (Me_2Hg and MeEtHg) and limits the amount of extract that can be analysed to 1 µl, thus negatively affecting the experimental detection limits. On the level of the interface the huge difference between the carrier (column) gas flow (1 ml/min) and the flow required to "punch" the plasma (1l/min) results in a sample dilution effect and the vulnerability of the interface to cold spots and dead volumes. This increases the complexity of the interface in terms of the precision of machining and the need for heating. The system has a high dead volume itself, and the inertness of the packing is limited, which may induce dismutation reactions which requiere silanization of the packing [67]. Uses of other multicapillary GC in speciation analysis have been described [68,69].

An automated, small and easily montable/demountable accesory based on a constant temperature multicapillary GC is proposed for time-resolved introduction of gaseous mercury species into an ICP-MS. The fast, narrow-band injection

was achieved by cryofocussing (-80 °C) of dimethyl, methylethyl and dimethylmercury in a capillary housed in a steel tube prior to desorption of the species (within 3–5 s) by rapid-pulse high intensity current. The compatibility of the operating variables with the ICP ionization conditions and negligible peak broadening on the column and in the ICP-MS interface allow the sensitive isotope-selective speciation of mercury (limit of detection 0.15 pg) in biological and sediment samples [70].

C. GC-MS coupling

The possibility of the use of GC-MS to mercury speciation has been developed in 1995 by Cai and Bayona [71]. The procedure described involves the use of a solid-phase microextraction of the mercury (Hg^{2+} and CH_3Hg^+) from water and biological samples. A previous liberation of mercury from biological samples is necessary, according the procedure reported by Fischer et al. [72] (which uses methanolic KOH sonication). The subsequent extraction was performed with sodium tetraethylborate and subsequent extraction with a silica fiber coated with poly(dimethylsiloxane). The mercury derivatives are desorbed in the splitless injection GC port and analysed by EIMS. Detection limits found are 0.0075 and 0.0035 ng/ml for methyl mercury and inorganic mercury, respectively.

HPLC methods

During the last few years many methods have been described that determine mercury species by HPLC and HPLC coupled with atomic spectroscopy. The main advantage of HPLC over GC is that there is no need to carry out prior derivatisation before separation, so the methods are generally much simpler and faster.

The methods used to speciate mercury with HPLC may be classified according to the detection system used, as follows: A) Voltametry, B) Visible UV spectroscopy, C) CVAAS, D) Plasmas (MIP-AES, ICP-AES, ICP-MS). Most of these methods use reverse-phase chromatography with C-18 (octyldecylsilane) chromatographic columns as the polar mobile phase (generally methanol).

A. Voltametric detection systems use a gold-amalgamate electrode and a C-18 chromatographic column to separate and measure mercury, MeHg, EtHg and PhHg. The reported detection limits range from 1 to 2 ng/ml [73].

B. UV-visible light spectrophotometry determines several organo-mercury compounds (MeHg, EtHg, PhHg, methoxy ethyl Hg, etc.) with a C-18 chromatographic column and gradient elution. Because of the low sensitivity of the detec-

tor, the detection limits are poor; from 7 to 95 ng/ml, but this system is simple and inexpensive [74].

3. Cold vapour atomic spectroscopy is a widely used technique because of its great sensitivity; it detects very low levels of mercury in a large variety of samples. Most of the applications use a C-18 column, cysteine is the eluent most frequently employed because of its complexing capacity. The separated mercury complexes are usually treated with tin chloride in sodium hydroxide solution to reduce them to Hg^0 that is then conveyed by an argon flow gas to the measuring cell where the mercury vapour is measured at 253.7 nm [75].

One variant of CVAS that has given good results in tests uses a vesicular mobile phase, produced by didodecyldimethylammonium bromide (DDAB). This procedure is applied to seawater and urine. The reported detection limits varies from 0.1 to 0.2 ng/ml [76].

D. In the last few years, methods that couple HPLC with plasma (MIP, ICP, ICP-MS) have been developed. However, the fact that the plasmas may sometimes be easily extinguished by liquid aerosols greatly limit their use.

On the other hand, coupling HPLC with MIP-AES appears to offer potential. The procedure described uses an automatic arrangement that incorporates a flow system to introduce the reactants. A vesicular type mobile phase produced by didodecyldimethylammonium bromide (DDAB) and ultrasound is used in a C-18 chromatographic column. The separated mercury compounds are reduced to $Hg^0$with $NaBH_4$ that is introduced into the surfatron microwave generator of the MIP. The reported detection limits varies from 0.15 ng/ml for inorganic mercury, and 0.35 ng/ml for MeHg [77].

Better potential for the future seems to be offered by ICP-MS; its sensitiviy is higher than ICP-AES and can also use a C-18 or another type of chromatographic column. It can determine inorganic mercury, MeHg, EtHg, and PhHg. Furthermore, because the compounds are measured directly, it is not necessary to reduce the mercury compounds to Hg^0 and, consequently, the procedure is much simpler. The reported detection limits when applied to urine samples range from 3 to 9 ng/ml [78,79].

CONCLUSION

The methods described until today for mercury speciation show, in general, several analytical steps, although it is quite obvious that the ideal analytical method for speciation of toxic elements would be directly in-situ analysis of the sought species in the desired sample; in the near future it is possible that electrochemical

and optical (bio)-sensors could be viable approaches for metal speciation. At present, however, in-situ direct species-specific techniques are seldom useful to solve speciation problems in real-fife situations because a lack of sensitivity. For this reason, previous sample pretreatments, separations, etc., before final detection are usually needed [80].

The methods that measure mercury without separating the compounds are fast, and simple, but they have the inconvenient that, in many cases, some of the mercury species (usually organic mercury) must be calculated by difference. Some of the procedures described appear to have potential, but there is little or no information about practical applications.

Those methods based on GC with EC or AS detection are very effective and sensitive, but suffer from the need to form volatile and stable derivatives before the chromatographic separation process and this complicates and slows the measurements. The increasing availability of ICP-MS in recent years has resulted in a number of studies on the GC introduction of organometallic compounds into the ICP plasma with a view to trace element environmental speciation analysis; coupling of gas or liquid chromatographies with ICP-MS (with some preconcentration process if necessary) seems the most promising solutions. On the other hand, methods that use liquid chromatography do not need derivatisation of the analytes and today these are the fastest and simplest procedures that, when combined with atomic spectroscopy, are adequately sensitive. Possibly, in the near future, coupling HPLC with ICP-MS, perhaps with some preconcentration process should the analysis require, might prove to be the most appropriate in spite of the high cost of the instruments.

However, we should bear in mind that some present-day analytical methods reach pg/ml detection limits and appear to be more than adequate to solve many of the analytical problems associated with most biological and environmental samples that need speciation of the most common mercury compounds that they might contain.

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