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A new selective method for dimethylamine in water analysis by liquid chromatography using solid-phase microextraction and two-stage derivatization with *o*-phthalaldialdehyde and 9-fluorenylmethyl chloroformate

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Abstract

A new method is presented for the determination of DMA in water as its 9-fluorenylmethyl chloroformate (FMOC) derivative using solid-phase microextraction (SPME) and liquid chromatography. The method is based on the employment of SPME fibres coated with carbowax-templated resin (CW-TR) for analyte extraction and derivatization. The fibres were successively immersed in the samples, in a solution of *o*-phthalaldialdehyde and *N*-acethyl-L-cysteine (OPA–NAC) and finally, in a solution of FMOC. OPA–NAC reacted on the fibre with possible primary aliphatic amines present in the samples, particularly with PA which is a direct interferent in the determination of DMA with FMOC. In such a way, the formation of PA–FMOC during the second stage was prevented, and thus the method was selective for DMA. The proposed procedure was applied to the determination of DMA in the 1.0–10.0 μg/mL range. The method provided suitable linearity, accuracy and reproducibility, and limits of detection and quantification of 0.3 and 1.0 μg/mL, respectively. The applicability of the method for the determination of DMA in different types of water is shown.

Keywords: Dimethylamine; Water; Solid-phase microextraction; On fibre two-stage derivatization

1. Introduction

Today, the potential of solid-phase microextraction (SPME) for sample preparation is widely recognized. SPME integrates sampling, extraction, preconcentration and sample introduction into a single process, resulting in high sample throughput. Moreover, it is a inexpensive, solvent-free and versatile technique that can by coupled to either gas chromatography (GC) or liquid chromatography (LC). SPME can also be combined with chemical derivatization to improve the extraction efficiency, or to make the analytes more amenable for chromatography and detection.

To date, derivatization has almost exclusively used in SPME-GC [1]. Two alternatives have been described to perform derivatization: addition of the reagent to the samples and subsequent extraction of the derivatives formed, and extraction of the analytes onto the fibres and subsequent derivatization of the analytes (on-fibre derivatization). Some of the applications in this area are the determination of fatty acids in aqueous or gaseous phases [2], the determination of amphetamines in urine [3], the determination of aliphatic amines in water and urine [4], and the quantification of aldehydes in water [5].

More recently, attempts have been made to combine chemical derivatization with SPME/LC. In this sense, Pawliszyn and co-workers described a procedure for the analysis of anatoxin-a in aqueous samples based on its derivatization

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on the surface of SPME fibres with the fluorogenic reagent 4-fluoro-7-nitro-2,1,3-benzoxadiazole [6]. The reagent was dropped or sprayed onto the fibre containing the extracted analyte. The fibre was then heated to 70 °C for 10 min, and finally, the fibre was inserted into the interface of the LC equipment. We have recently described a method for the analysis of methylamine in water based on its on-fibre derivatization with 9-fluorenylmethyl chloroformate (FMOC) [7]. Derivatization was accomplished by immersing the fibres with the extracted analyte into the reagent solution. The same reagent has been also used to derivatize amphetamines in an study aimed at comparing two extraction/derivatization strategies: SPME and subsequent on-fibre derivatization of the extracted amphetamines, and solution derivatization followed by SPME of the derivatives formed [8]. The latter approach was found to be the only option suitable for the analysis of urine samples. This was because the extraction of matrix components into the fibre coating prevented the extraction of the reagent. For the analysis of aqueous matrices, the SPME/on-fibre derivatization approach was preferable as the analysis did not modify sample composition.

The literature shows numerous LC assays which involve relatively complex derivatization procedures. This is the case of methods in which the analytes must be previously transformed into species capable of reacting with the selected derivatization agent. The simultaneous use of two reagents has been proposed to resolve samples containing a large number of target compounds. Other applications describe the use of an additional reaction aimed at eliminating from the reaction medium possible interferents before the derivatization of the compounds of interest.

In the present work we have evaluated the possibility of using SPME to effect two-stages derivatizations with two different reagents. The secondary amine dimethylamine (DMA) has been selected as a model compound. As many other short-chain aliphatic amines, DMA is a compound of environmental interest due to its toxicity, reactivity and likely occurrence as a result of its wide industrial use. Moreover, DMA may react with nitrosating agents giving the carcinogenic compound *N*-nitrosodimethylamine. For these reasons, there is an increasing demand of analytical methods for monitoring DMA in environmental waters.

Most LC methods specifically developed for the analysis of DMA entail derivatization with FMOC [9,10]. However, in the course of our studies on the derivatization of aliphatic amines we have observed that DMA–FMOC derivative tends to coelute with the derivative originated by propylamine (PA). This interference was not taken into consideration in previously reported methods. Overlapping of DMA and PA occurs under a variety LC conditions [11]. To overcome this problem, in the present study we propose a sequential derivatization with *o*-phtaldialdehyde and *N*-acetyl-L-cisteyne (OPA–NAC), and then with FMOC (see Fig. 1). Since OPA–NAC is only reactive towards primary amines, in the first stage PA is transformed into PA–OPA–NAC. In the second stage FMOC only reacts with DMA. Since the PA–OPA–NAC and DMA–FMOC derivatives present very different features, they can be satisfactory resolved under typical reversed phase conditions. On the basis of the results obtained a new method is presented for the selective determination of DMA in water.

2. Experimental

2.1. Apparatus and chromatographic conditions

The chromatographic system consisted of a quaternary pump (Hewlett-Packard 1050 Series, Palo Alto, CA, USA), a SPME–HPLC interface (Supelco, Bellefonte, PA, USA) and a fluorescence detector (Hewlett-Packard, 1050 series). The detector was coupled to a data system (Hewlett-Packard, HPLC Chem Station) for data acquisition and calculation. For measurement of the FMOC derivatives the excitation and emission wavelengths were 264 and 313 nm, respectively. The OPA–NAC derivatives were monitored at excitation and emission wavelengths of 330 and 440 nm, respectively.

2.2. Reagents and solutions

All the reagents were of analytical grade. Dimethylamine, methylamine, ethylamine, propylamine, *n*-butylamine, *n*-pentylamine and diethylamine were obtained from Sigma (St. Louis, MO, USA). 9-Fluorenylmethyl chloroformate (FMOC) and *N*-acetyl-L-cysteine (NAC) were purchased from Aldrich (Stenheim, Germany). *o*-Phthaldialdehyde (OPA) was obtained from Fluka (Buchs, Switzerland). Sodium hydroxide, boric acid and hydrochloric acid were purchased from Panreac (Barcelona, Spain). Acetonitrile was of HPLC grade (Scharlau, Barcelona, Spain).

Stock standard solutions of DMA and the other amines (10 000 μg/mL) were prepared in water. Working solutions of these compounds were prepared by dilution of the stock solutions with water. All solutions were stored in the dark at 2 °C.

2.3. Columns and mobile phases

A LiChrospher 100 RP18, 125 mm × 4 mm i.d. column (Merck, Darmstadt, Germany) was the analytical column. In the optimized procedure a precolumn and a high-pressure six-port valve (Hewlett-Packard) were inserted between the SPME–HPLC interface and the analytical column in order to effect peak compression [7]. The precolumn (20 mm × 2.1 mm i.d.) was dry-packed with a Hypersil C_{18}, 30 μm, stationary phase. At the beginning of each chromatographic the precolumn and the analytical column were disconnected, so the eluent (water) was sent to waste. At 0.5 min, the valve was rotated, and the percentage of acetonitrile in the mobile-phase was progressively increased,

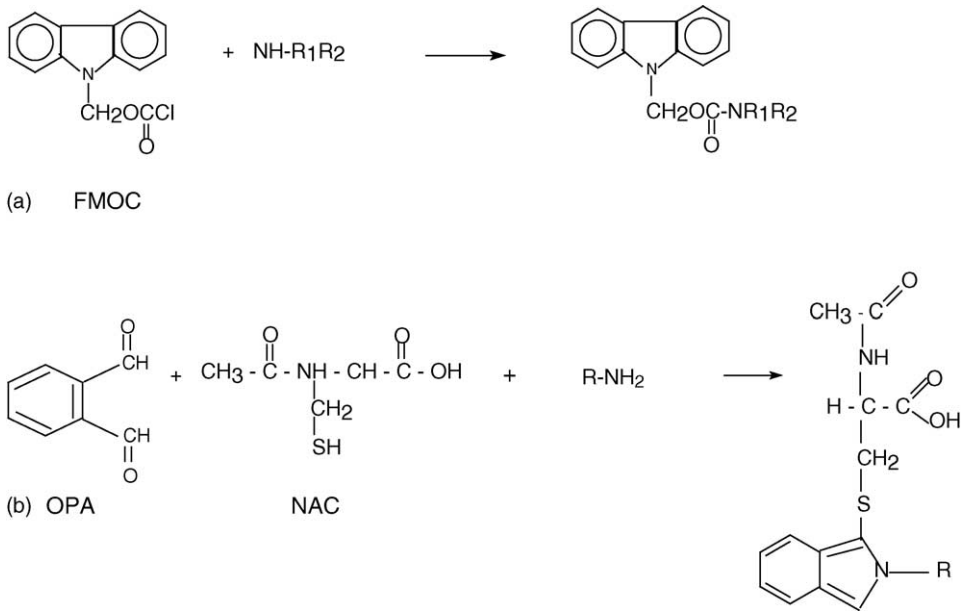

Fig. 1. Reaction schemes for the derivatization of amines (a) with FMOC (primary and secondary amines), and (b) with OPA–NAC (only primary amines).

so the DMA–FMOC derivative retained in the precolumn was transferred to the analytical column. The acetonitrile content in the mobile-phase was increased from 0% at 0 min to 40% at 2.5 min, to 70% at 9.5 min and then to 100% at 17.5 min. The mobile-phase flow rate was 0.95 mL/min.

All solvents were filtered through 0.45 μm nylon membranes (Teknokroma, Barcelona, Spain) and degassed with helium before use.

2.4. SPME parameters

A SPME assembly with replaceable extraction fibre was used for extraction of DMA and the derivatization reagents. Carbowax-templated resin (CW-TR, 50 μm) were obtained from Supelco. The fibres were immersed into a glass vial containing the samples (25 m) in order to extract DMA. The adsorption time was 15 min. After on-fibre derivatization, the fibres were placed into the SPME–HPLC interface device, and the DMA–FMOC derivatives were desorbed from the fibre under static mode: the fibre was soaked with 200 μL of acetonitrile for 5 min (desorption time). Next, the valve of the interface was activated so DMA–FMOC was sent to the precolumn, and the chromatographic run was started. At the end of each run, the fibres were cleaned and conditioned by immersing them into 10.0 mL water for 2.0 min, and then, into 10.0 mL of acetonitrile for other 2.0 min.

2.5. Derivatization procedures

2.5.1. On-fibre derivatization with FMOC

Fibres with the extracted DMA (or other short-chain aliphatic amines tested) were immersed into a 2 mL glass vial containing the FMOC derivatization solution for a defined period of time (reaction time). This derivatization solution consisted in a mixture of 0.5 mL of 0.05 M borate buffer (pH 10.0) and 1.0 mL of FMOC. The FMOC solutions were prepared daily by dissolving the pure compound in acetonitrile. Different concentration of FMOC in the 0.25–25.0 mM range were tested. The borate buffer was prepared by dissolving boric acid in water, and then the pH was adjusted to 10.0 by adding 0.5 M NaOH. After the reaction time, the fibres were removed from the vial, and placed into the SPME–HPLC interface.

Each sample was derivatized in triplicate and all assays were carried out at ambient temperature.

2.5.2. On-fibre derivatization with OPA–NAC and with FMOC

Fibres with the extracted DMA (or other short-chain aliphatic amines tested) were immersed into a 2 mL glass vial containing the OPA–NAC derivatization solution. Unless otherwise stated, the derivatization time was 5 min. The derivatization solution consisted of 0.1 mL of 0.05 M borate buffer (pH 10.0) and 0.9 mL of OPA–NAC. The OPA–NAC solution was prepared daily by dissolving the pure compounds (at the same concentration) into the minimum volume of methanol (≈1.4 mL). The resulting solution was further diluted to 100 mL with water. Different concentrations of OPA–NAC in the 3.7–15.0 mM range were assayed. The borate buffer was prepared as described in the above section.

After the reaction time, the fibres were removed from the vial containing the OPA–NAC reagent, and subjected to derivatization with FMOC. Derivatization with FMOC was carried out according to the procedure described in the previous section.

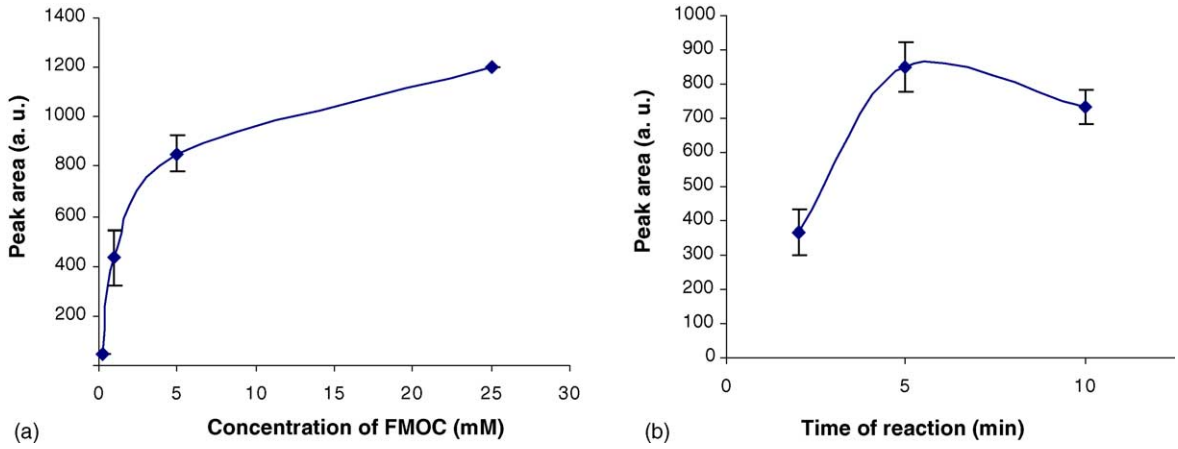

Fig. 2. Optimization of conditions in the reaction between DMA and FMOC: (a) effect of the concentration of reagent, for a time of reaction of 5 min; (b) effect of time of reaction, for a concentration of FMOC of 5 mM. Concentration of DMA, 5 μg/mL. For other experimental details, see text.

Each sample was derivatized in triplicate and all assays were carried out at ambient temperature.

2.6. Analysis of real water

Tap water, ground water, irrigation water and river water were spiked with DMA reproducing different concentrations within the tested range, and processed by the OPA–NAC/FMOC method under optimized conditions. In previous test it was verified that such samples did not contain DMA. Waste water were collected and acidified to pH 2.0 with concentrated hydrochloric acid for preservation. Before analysis, the pH was adjusted to pH 10.0 with 0.5 M NaOH, and samples filtered through 0.45 μm membranes (Teknokroma). Samples were then processed by the proposed OPA–NAC/FMOC method.

Each sample was derivatized in triplicate and all assays were carried out at ambient temperature.

3. Results and discussion

3.1. On-fibre derivatization of DMA with FMOC

Conditions selected for the SPME of DMA were as follows: sample volume of 25.0 mL, adsorption time of 15.0 min, and desorption time of 5.0 min. Under such conditions, the responses of the analyte were found to depend mainly on the reaction parameters. The elution conditions were adjusted to achieve a complete resolution between DMA–FMOC and unreacted FMOC (which was also significantly extracted onto the fibres).

The first parameter related with the derivatization of DMA that we studied was the concentration of FMOC. Fibres with the extracted DMA were immersed into a mixture of 0.5 mL of 0.05 M borate buffer (pH 10.0) and 1.0 mL of FMOC at a concentration ranging between 0.25 and 25.0 mM. The presence of the buffer is necessary for the amine to be in the proper form (unprotonated) for reaction [12]. In this study, the concentration of DMA was 5 μg/mL and the reaction time was 5 min. After then, the fibres were placed into the SPME–HPLC interface for desorption and chromatography of the derivative formed.

As observed in Fig. 2a, the peak areas obtained for DMA–FMOC increased by increasing the concentration of FMOC within the tested range, although little improvement was observed with the highest concentrations assayed. Moreover, a large excess of reagent resulted in overlapping between the peak of DMA–FMOC and the peak of unreacted FMOC. As a compromise, a concentration of FMOC 5.0 mM (giving a final concentration on the reaction mixture of 2.5 mM) was selected for further experiments.

The effect of time of reaction was investigated in the 2.0–10.0 min range. In this study the concentration of FMOC used to prepare the derivatization solution was 5.0 mM, and the other conditions were those indicated above. The optimum time of reaction was found to be 5.0 min (see Fig. 2b). Consequently, this was the time of reaction selected in further work.

The chromatograms obtained for a blank and for an standard solution of DMA under the selected conditions are depicted in Fig. 3. It can be observed that DMA–FMOC (retention time, t_r = 9.9 min) is well separated from unreacted FMOC (t_r = 7.3 min). Another peak related to the FMOC (most probably corresponding to a condensation product) was detected at 11.4 min.

The proposed conditions were also applied to analyze standard solutions of other short-chain aliphatic amines. The compounds tested were methylamine, ethylamine, PA, butylamine, pentylamine and diethylamine. Besides DMA, all the amines tested were significantly extracted onto the fibres and reacted FMOC. However, their respective FMOC derivatives could be resolved from DMA–FMOC, with the only exception of the derivative formed by PA, which also eluted at 9.9 min (see Fig. 4). Attempts were made to resolve the peaks originated by DMA and PA. Several mobile phase composi-

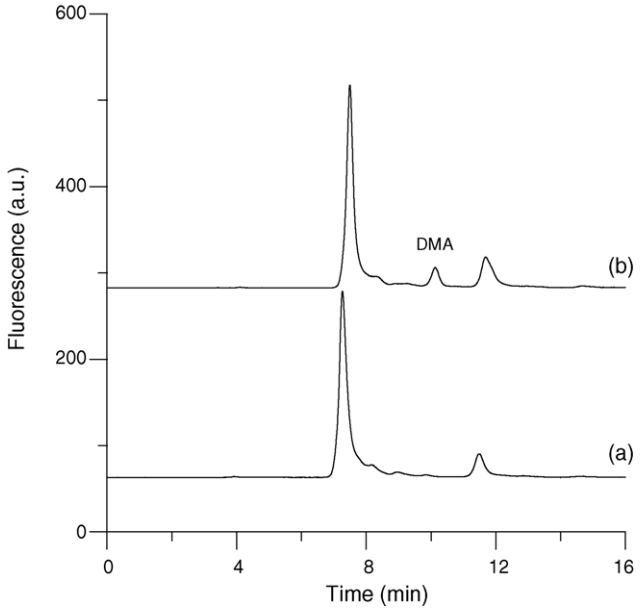

Fig. 3. Chromatograms obtained for (a) a blank (water) and (b) a standard solution of DMA (5 μg/mL) by the on-fibre derivatization method with FMOC. Peaks at 7.1 and 11.6 corresponded to unreacted FMOC and a condensation product, respectively. Final concentration of FMOC in the reaction mixture 3.3 mM; reaction time, 5 min. For other experimental details, see text.

tions and elution programs were tested, but in all conditions assayed the PA–FMOC eluted at almost identical times than DMA–FMOC.

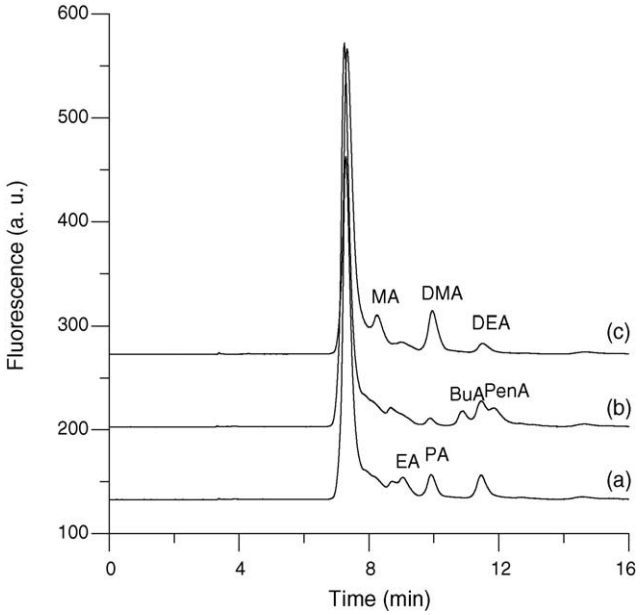

Fig. 4. Chromatograms obtained for standard solutions containing mixtures of the tested short-chain aliphatic amines: (a) ethylamine (EA) and PA; (b) *n*-butylamine (BuA) and *n*-pentylamine (PenA); (c) methylamine (MA), DMA and diethylamine (DEA). Concentration of each amine in the samples, 5.0 μg/mL. Final concentration of FMOC in the reaction mixture 3.3 mM; reaction time, 5 min. For other experimental details, see text.

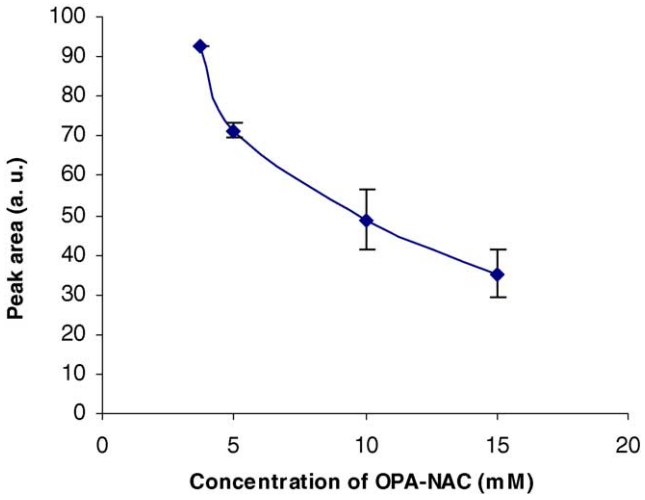

Fig. 5. Effect of the concentration of OPA–NAC on the responses obtained for PA. Concentration of PA, 5 μg/mL; time of reaction, 5 min. For other experimental details, see text.

3.2. On-fibre derivatization of PA with OPA–NAC

OPA is one of the reagent most widely used for labelling compounds containing primary amino groups. This is because in the presence of a thiol (such as NAC) OPA forms highly fluorescent isoindoles (see Fig. 1b). The utility of this reagent for the determination of primary aliphatic amines has been previously demonstrated [13]. The simultaneous use of OPA and FMOC for resolution of samples containing a large number of target compounds has been previously described [14,15]. In the present study we have evaluated the possibility of using OPA–NAC to transform possible PA into its corresponding isoindole derivative, thus preventing the formation of PA–FMOC during the derivatization of DMA.

PA was extracted from samples under conditions used for DMA and next, the fibres were immersed into a solution containing OPA–NAC. As in the derivatization with FMOC, a basic medium was also necessary. For this reason, the derivatization solution was a mixture of barate buffer of pH 10.0 and OPA–NAC. After a defined time of reaction, the fibres were removed from the derivatization solution of OPA–NAC, and next, immersed into the derivatization solution of FMOC. Conditions for derivatization with FMOC were those proposed for DMA.

In order to achieve maximum conversion of PA, different solutions of OPA–NAC at concentrations in the range 3.7–15.0 mM were assayed (resulting in final concentrations in the derivatization mixture of 3.33–13.5 mM). The concentration of PA in the samples was 5.0 μg/mL, and the reaction time was 5.0 min The results obtained are depicted in Fig. 5. As expected, the peak corresponding to PA–FMOC in the resulting chromatogram decreased as the concentration of OPA–NAC increased. For a concentration of OPA–NAC of 15.0 mM, the peak of PA–FMOC was not detected. This is illustrated in Fig. 6, which compares the chromatograms

obtained for PA after derivatization with FMOC and after the optimized two-stages reaction procedure with OPA–NAC and FMOC. It is interesting to note that for the same concentration of OPA–NAC, the peak of PA–FMOC was detected when using times for reaction with FMOC longer than 5.0 min. This suggested that PA–FMOC is more stable than the compound formed with OPA–NAC.

The time of retention of PA–OPA–NAC, established by registering the chromatograms at the proper detection conditions ($\lambda_{excitation}$ = 330 and $\lambda_{emission}$ = 440 nm) was 4.1 min. This peak was not detected under the detection conditions used to monitorize FMOC derivatives ($\lambda_{excitation}$ = 264 and $\lambda_{emission}$ = 313 nm).

In Fig. 6 is also depicted the chromatogram obtained for DMA by the OPA–NAC/FMOC method. The mean ratio

$$\frac{\text{peak area of DMA by the OPA-NAC/FMOC method}}{\text{peak area of DMA by the FMOC method}}$$

was 0.950 (n = 5, 5.0 μg/mL of DMA). This indicates that the amount of DMA in the fibres was not substantially altered by the derivatization with OPA–NAC, which can be explained by the high affinity of this amine for the CW-TR coating. Therefore, the inclusion of the derivatization step with OPA–NAC did not significantly affect the sensitivity obtained in the measurement of DMA–FMOC. This is an advantage of SPME mediated derivatizations over conventional solution derivatizations, which typically entail analyte dilution due to the addition of the reagents.

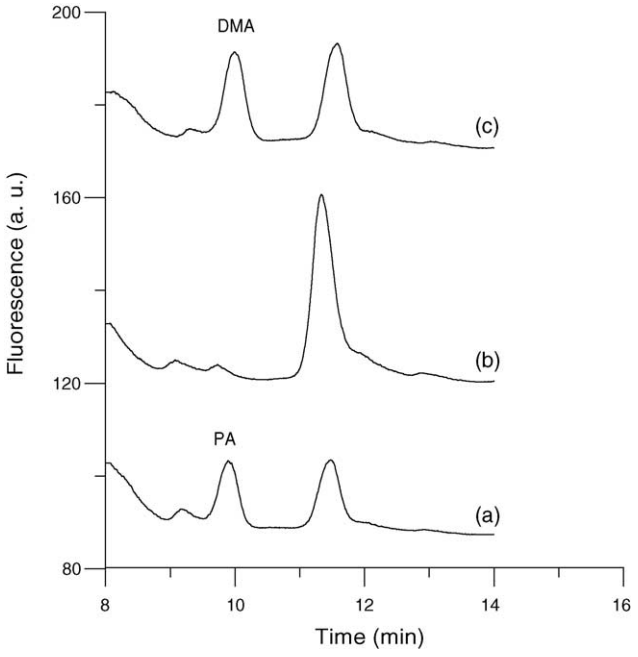

Fig. 6. Chromatograms obtained for PA (5.0 μg/mL) (a) by the FMOC method, and (b) by the OPA–NAC/FMOC; (c) chromatogram obtained for DMA (5.0 μg/mL) by the OPA–NAC/FMOC method. Reaction with OPA–NAC: final concentration of OPA–NAC, 2.5 mM; time of reaction, 5 min. Reaction with FMOC: final concentration of FMOC 3.3 mM; reaction time, 5 min. For other experimental details, see text.

Table 1
Accuracy for the determination of DMA by the optimized OPA–NAC/FMOC method (n = 3)

Sample	Added concentration (μg/mL)	Determined concentration (μg/mL)	Relative error (%)
Standard	1	1.1 ± 0.3	+12
	5.0	5.0 ± 0.8	+0.4
	10	10.4 ± 1.3	+4
	5.0 + 5 μg/mL of PA	4 ± 1	−20
	5.0 + 10 μg/mL of PA	4.9 ± 0.5	−2
Tap water	1.0	0.9 ± 0.3	−6
	5.0	5.17 ± 1.18	+3
Irrigation	1.0	1.0 ± 0.4	0
water	5.0	5.1 ± 0.4	+3
River	1.0	1.03 ± 0.4	+3
water	5.0	4.3 ± 0.6	−15
Ground	1.0	0.9 ± 0.2	−8
water	5.0	4.9 ± 1.9	−3

3.3. Determination of DMA by the OPA–NAC/FMOC method

The reliability of the described method for the quantification of DMA in water was evaluated by processing standard samples containing the analyte in 1.0–10.0 μg/mL concentration range [16,17]. The linearity was tested by analysing samples at five different concentrations within the studied interval. The calibration equation obtained was $y = -(35 \pm 35) + (142 \pm 7)x$, with a mean correlation coefficient of R = 0.990 (n = 10). The method also provided suitable reproducibility, with intra-day and inter-day coefficients of variation, established for 5.0 μg/mL of DMA, of 7% (n = 3) and 16% (n = 6), respectively. The limit of quantification (LOQ) and the limit of detection (LOD) were 1.0 and 0.3 μg/mL, respectively.

The accuracy was evaluated by processing standards solutions containing DMA at different concentrations within the tested concentration range. As observed in Table 1, suitable accuracy was observed, with a relative errors ranging from +12% (for a concentration equivalent to the LOQ) to +0.4%. In this table are also listed the concentrations of DMA calculated for standard samples containing mixtures of DMA and PA. The results obtained confirmed that the presence of PA did not affect the quantification of DMA. Therefore, the proposed method can be considered selective for DMA.

3.4. Application to real water samples

Different types of samples were analyzed in order to determine DMA: tap water, irrigation water, river water and ground water. Samples were fortified at different levels of concentration within the tested concentration range. The concentration of DMA was established from the calibration equation obtained from standards of DMA. The results of this study (see also Table 1) indicate that concentrations calculated with proposed procedure were close to the real ones.

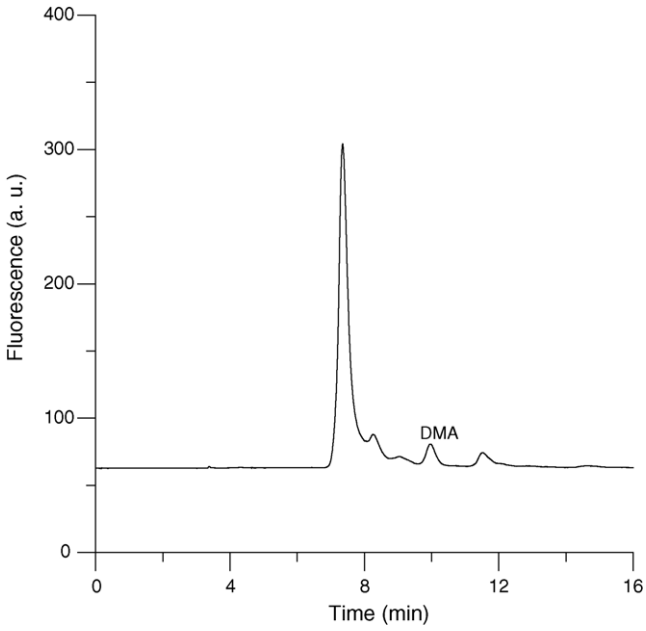

Fig. 7. Chromatogram obtained for a waste water sample by the optimized OPA–NAC/FMOC method.

Finally, the described method was applied to the analysis of waste water obtained from an industry dedicated to the production of fertilizers. As an illustrative example, in Fig. 7 is depicted the chromatogram obtained for one of the samples assayed. The concentration determined for DMA in such sample was 1.75 μg/mL.

4. Conclusions

The concept of SPME with on-fibre derivatization has been extended to derivatization procedures involving the sequential use of different reagents. In the present work, a two-stage derivatization method has been developed for DMA based on its derivatization with FMOC. The fibres with the extracted analyte were successively immersed into derivatization solutions of OPA–NAC and FMOC. During the first derivatization stage, possible primary aliphatic amines present in the samples were transformed into their OPA–NAC derivatives. Conditions were adjusted to transform quantitatively PA, which is a direct interferent in the determination of DMA with FMOC. In such a way, the formation of PA–FMOC during the second stage is avoided, and thus the method can be considered selective for DMA.

Compared with conventional solution derivatization, the incorporation of the derivatization process into SPME fibres not only simplified the experimental effort, but also avoided analyte dilution caused by the addition of the reagents. This may be particularly important in derivatization procedures requiring two or more different reagents to achieve the desired selectivity.

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