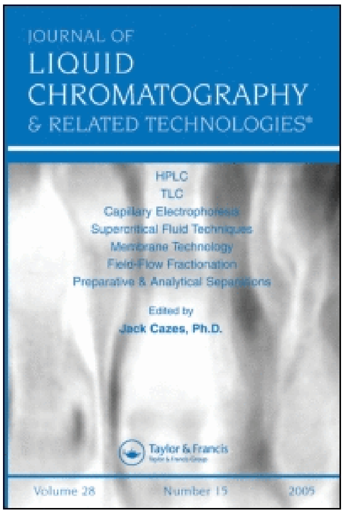

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MICROWAVE-ASSISTED EXTRACTION OF POLYCHLORINATED BIPHENYLS AND POLYCHLORINATED DIBENZODIOXINS FROM FLY ASH

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ABSTRACT

The effects of extraction parameters on the microwave-assisted extraction (MAE) of polychlorinated biphenyls (PCBs) and polychlorinated dibenzodioxins (PCDDs) from fly ash were compared. The effects of toluene-isopropyl alcohol (IPA) mixture compositions, solvent volume, temperature, extraction time (rise and hold time), and microwave power on the variation of recoveries were studied. The MAE carried out at 100°C for 35.3 min using 5 mL of toluene-IPA (90 : 10, v/v) was more effective in isolating PCBs and PCDDs from fly ash.

Multi-layer column chromatography on neutral and acidic silica gel with *n*-hexane was used for cleaning up the extracts. High performance liquid chromatography (HPLC) with ultraviolet (UV) detection was employed to identify and quantify the analytes.

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INTRODUCTION

Microwave-assisted extraction (MAE) has attracted growing interest in recent years, as it allows rapid extractions of analytes from solid matrices and it offers extraction efficiency comparable to that provided by traditional techniques. Numerous applications of this recent technique deal with extracting pollutants from environmental samples. For instance, polychlorinated biphenyls (PCBs) were efficiently extracted from sewage sludge using MAE (1). Satisfactory extractions could be achieved within 10 min, while recoveries can compare favorably with Soxhlet values. MAE has also been applied in determining PCBs in water samples (2). PCBs were first trapped onto C18 membrane disks, before being eluted by applying a microwave field, allowing rapid determination of such compounds in water samples.

An MAE procedure was developed for extracting PCDDs (polychlorinated dibenzodioxins) and PCDFs (polychlorinated dibenzofurans) from sewage sludges (3). Recoveries ranged from 30 to 70% for the 17 isomers investigated. Increasing the power from 45 W to 90 W has no effect on the recoveries. The results obtained using MAE were congruent with concentrations found using Soxhlet extraction. Shu et al. (4) conducted an MAE of polycyclic aromatic hydrocarbons (PAHs) from sediment and river bed soil. Both the MAE and the Soxhlet extraction method yielded comparable results.

Lopez-Avila et al. (5) reported the use of microwave irradiation in the presence of solvents to extract different organic compounds (organochlorine pesticides, polychlorinated biphenyls, organophosphorous pesticides, and other semi-volatile compounds) from various solid matrices (soils and sediments).

In MAE, the solvent used should generally be capable of absorbing the microwave energies. The amount of microwave energy absorbed owing to the reorientation of permanent dipoles by the electric field is proportional to the dielectric constant of the solvent. Most of the time, absorption is also proportional to the solvent polarity. For example, hexane-acetone 1 : 1 (v/v) was efficient in extracting PAHs from sediments, with recoveries similar to those obtained when dichloromethane was used (6). The best results for MAE of sulfonylurea herbicides from soil were obtained when dichloromethane-methanol (90 : 10, v/v) mixture was used (7). The MAE of phenols from soils with a hexane-acetone mixture was more efficient as the percentage of acetone in the mixture was increased (8).

In some cases, the solvent volume may be important in facilitating efficient extractions. For example, recoveries of methylmercury from sediments depended largely on the volume of hydrochloric acid (9).

Temperature is of prime importance in ensuring efficient extraction, as elevated values usually enhance the extraction process because of the solvent's increased diffusivity into the internal parts of the matrix under high temperatures. This is also brought about by an enhanced desorption of the components from the active sites of the matrix. Hoogerbrugge et al. (10) reported that temperature was found to be a significantly influential parameter on the extraction efficiency of triazines using MAE; values of 80–100°C were found acceptable.

The organic carbon content of the matrix is known to hinder extraction, owing to strong analyte-matrix interactions that are difficult to disrupt (11,12). Since fly ash is a carbonaceous particle adsorbing strongly with PCBs and PCDDs, extraction is difficult. In this paper, microwave-assisted extraction (MAE) is employed to extract PCBs and PCDDs from municipal solid waste incinerator (MSWI) fly ash. The efficiency of toluene-IPA mixture compositions on MAE of PCBs and PCDDs from fly ash has rarely been compared. It should be noted that the purpose of this investigation is to compare the effect of extraction parameters on MAE rather than to obtain quantitative recoveries. The optimum conditions of MAE are established for higher recoveries of PCBs and PCDDs from fly ash. The extraction parameters studied are toluene-IPA mixture compositions, solvent volume, temperature, extraction total time (rise + hold time), and microwave power. The effect of turning the vessels within the cavity on the recoveries is also studied.

EXPERIMENTAL

Reagents

All organic solvents (methanol, dichloromethane, *n*-hexane, toluene and acetonitrile) used were of HPLC grade and were obtained from Burdick & Jackson (Muskegon, MI, USA). Isopropyl alcohol (IPA) was obtained from Kanto Chemical (Tokyo, Japan). Silica gel (230–400 mesh, Merck, Darmstadt, Germany) was initially rinsed with methanol twice, and then with dichloromethane twice. Consecutively, it was activated at 180°C for at least 12 hours. Anhydrous sodium sulfate (Merck, Darmstadt, Germany) was used to protect the packing materials in the multi-layer silica gel column.

PCBs consisting of 2,2′,4,5′-tetrachlorinated biphenyl (4B), 2,3,4,5,6-pentachlorinated biphenyl (5B), and PCDDs consisting of 1,2,3,4-tetrachlorinated dibenzodioxin (4D), 1,2,3,4,7-pentachlorinated dibenzodioxin (5D), 1,2,3,4,7,8-hexachlorinated dibenzo-dioxin (6D), 1,2,3,4,6,7,8-heptachlorinated dibenzodioxin (7D), and OCDD (8D) were obtained from Ultra Scientific (250 Smith Street, North Kingstown, USA). A stock solution containing a mixture of PCBs of 5.0 μg/mL and PCDDs of 0.45 μg/mL was prepared in toluene for each compound.

Sample Preparation

The fly ash samples collected from the Mokdong municipal solid waste incinerator (Seoul, Korea) were used as a fortified sample matrix (FSM) during the recovery studies. The fly ash was air-dried to remove water content. The 0.5 g fly ash samples were spiked at 500 ng/g level with a stock solution of 4B and 5B, and at 45 ng/g with 4D, 5D, 6D, 7D, and 8D.

Microwave-assisted extraction (MAE) was employed for the extraction of PCBs and PCDDs from fly ash samples. The sample was transferred to Teflon-lined extraction vessels. After the extraction using a MARS X microwave extraction system (CEM, Matthews, NC, USA), the vessels were allowed to cool down in room temperature before the caps were opened. After filtering and concentration, the multi-layer silica gel column was employed for the clean-up of the extract.

MAE variables, consisting of extracting solvents, solvent volumes, temperatures, total times (rise + hold), and microwave powers, were evaluated in this study to compare recoveries of analytes. Rise time is the time required to reach a temperature during microwave irradiation, while hold time is the time required to maintain a temperature during microwave irradiation.

First, four different extracting solvent compositions of toluene-IPA, 100 : 0, 95 : 5, 90 : 10, and 80 : 20 (v/v) were compared for rise time of 10 min or at 100°C. Secondly, the extractions were carried out at five different temperatures, 72, 85, 94, 100, or 110°C for rise times corresponding to each temperature, or a total time of 25.3 min. Third, four different total times, 13.0, 25.3, 35.3, and 45.3 min were applied, and the relative contributions between rise and hold time to recoveries were compared for a total time of 25.3 min. Fourth, three different powers (300, 600, and 1200 W) and four different solvent volumes (1.3, 2.5, 5.0, and 7.5 mL) were evaluated. Lastly, whether the recoveries were influenced by turning the vessel in the microwave cavity, or not, was determined.

The extracts were cleaned up using a multi-layer silica gel column (13,14), which had been packed in neutral (2 g), acidic (6 g), and neutral (4 g) silica gel, in this particular order. The column was eluted with 50 mL of *n*-hexane. The eluent was concentrated using a rotary evaporator in 2–3 mL and transferred into a 7-mL vial. Nitrogen evaporation was carried out to remove *n*-hexane, and then 100 μL of acetonitrile was refilled for quantification using HPLC-UV.

Analysis of Extracts

The amount of PCBs and PCDDs in the liquid extracts was determined using a Kromasil KR100-5C18 (250 × 4.6 mm, Eka Chemicals AB, Bohus, Sweden) column. The HPLC system used in this work was an M920 delivery pump (Young-Lin, Korea) equipped with an SPD-10A UV-visible detector (Shimadzu, Japan) and C-R6A integrator (Shimadzu). The injected volume was 20 μL, and the flow rate of the mobile phase was 1.0 mL/min at 40°C. An acetonitrile-water (90 : 10, v/v) solution was used to separate the analytes within 40 min. The chromatograms were recorded at 250 nm (A.U.F.S. = 0.005).

RESULTS AND DISCUSSION

The relative ratios of recoveries were obtained by dividing the highest % recovery into the corresponding % recovery in the tables.

Effect of Toluene-IPA Mixture Compositions

The effects of extracting solvent compositions on the temperature, the MAE recoveries and the relative recovery ratios of PCBs and PCDDs are presented in Table 1. The effect of temperature was strongly dependent on the content of isopropyl alcohol (IPA) in toluene because temperature was proportional to the content during the rise time of 10 min. It has been proven that higher temperature results in higher recovery. However, the higher recoveries of PCBs and PCDDs were obtained at 94°C induced by 10% IPA level rather than 110°C by 20%. This MAE system usually increases sample throughput, thereby reducing the extraction time if multiple extraction using several vessels is carried out. However, from the comparison of results between single and multiple extractions in Table 1, it was determined that single extraction yielded higher recoveries than multiple extractions.

Table 2 shows the effect of solvent compositions on the rise time, the recoveries, and the relative ratios of recoveries, at a constant temperature of 100°C. As the content of IPA in toluene was increased, the rise time was reduced from 30.0 to 4.5 min. Although, the corresponding extraction time was shorter, the use of toluene-IPA 90 : 10 (v/v) mixture yielded higher recoveries than the other mixtures (100 : 0 and 95 : 5, v/v).

From the results of Table 1 and Table 2, it was concluded that the best MAE could be carried out at 10% IPA level out of four % IPA levels. This observation is in agreement with the fact that microwaves are slightly absorbed by toluene, so the presence of IPA molecules was necessary to absorb microwave energy, resulting in increased temperature, and microwave heating of IPA in the matrix resulted in more matrix surface available for the extracting solvent (toluene). Similar to the results of enhanced-fluidity liquid extraction (EFLE) and

Table 1. Recovery and Relative Ratio of Recovery of PCBs and PCDDs in Fly Ash Extracts and Variation of Temperature by Different Toluene-IPA Mixtures

	Recovery (%)	Relative Ratio of Recovery		
Run (single or multiple):	Single			
Toluene-IPA (v/v):	90:10	100:0	95:5	80:20
Resulting Temperature (°C):	94	72	85	110
Analytes				
4B	53.3	0.77	0.84	0.89
5B	58.5	0.89	0.98	0.96
4D	51.5	0.85	0.88	0.93
5D	58.5	0.86	0.96	0.94
6D	56.1	0.84	0.95	0.89
7D	69.8	0.87	0.94	0.88
8D	72.4	0.89	0.95	0.93
Average (SD)	60.0 (2.6)	0.85 (0.03)	0.93 (0.02)	0.92 (0.03)
	Relative ratio of recovery			
Run (single or multiple):	Multiple			
Analytes				
4B	0.85	0.71	0.77	0.79
5B	0.95	0.84	0.86	0.91
4D	0.87	0.65	0.73	0.74
5D	0.89	0.68	0.78	0.79
6D	0.86	0.69	0.72	0.83
7D	0.92	0.75	0.85	0.85
8D	0.94	0.78	0.89	0.93
Average (SD)	0.89 (0.04)	0.73 (0.05)	0.80 (0.03)	0.83 (0.03)

Condition: rise time 10 min, hold time 0 min, power 600 W, solvent volume 5 mL.

pressurized fluid extraction (PFE) in the previous paper (15), a larger proportion of IPA to toluene, i.e. 20%, reduced the extraction recoveries. This can be explained by the strong analyte-matrix interaction, which is better overcome using toluene than IPA.

Effect of Temperature

The effect of varying extraction temperature on the recoveries and the relative recovery ratios of the analytes is illustrated in Table 3. Increased

Table 2. Recovery and Relative Ratio of Recovery of PCBs and PCDDs in Fly Ash Extracts and Variation of Rise Time by Different Toluene-IPA Mixtures

	Recovery (%)	Relative Ratio of Recovery		
Toluene-IPA (v/v):	90:10	100:0	95:5	80:20
Resulting Rise Time (min): Analytes	13.0	30.0	17.5	4.5
4B	67.0	0.76	0.93	0.73
5B	73.7	0.76	0.99	0.74
4D	70.2	0.60	0.95	0.70
5D	75.6	0.64	0.94	0.72
6D	75.8	0.66	0.93	0.66
7D	80.4	0.74	0.99	0.71
8D	84.1	0.82	0.97	0.79
Average (SD)	75.2 (2.2)	0.71 (0.03)	0.96 (0.04)	0.72 (0.04)

Condition: temperature 100°C, hold time 0 min, power 600 W, solvent volume 5 mL.

recoveries of analytes were obtained from 72 to 100°C because an increase in the extraction temperatures results in a corresponding increase in desorption of the analytes. Increased recoveries were not observed between 100 and 110°C. Our previous paper also reported higher recoveries of PCBs and PCDDs using PFE and EFLE from fly ash at 100°C (15). Therefore, it can be suggested that the optimum temperature is independent of the extraction methods as the same temperature is also optimal for MAE, just as 100°C is suitable for PFE and EFLE.

The differences among the relative ratios of recoveries with temperatures at hold time of 0 min were larger than those with temperatures at total (rise + hold) time of 25.3 min. As a result, no significant difference was observed on the recoveries of the analytes between 94 and 100°C at total time of 25.3 min, while the average relative ratio of the recovery at 94°C to the recovery at 100°C was 0.80 ($= 0.74/0.92$) at hold time of 0 min. From these observations, it was concluded that the extraction time including rise and hold time influenced variations in MAE recoveries.

Effect of Hold Time

The effect of varying hold time on the recoveries and the relative recovery ratios of the analytes is illustrated in Table 4. An increase in hold time 0 to

Table 3. Recovery and Relative Ratio of Recovery of PCBs and PCDDs in Fly Ash Extracts by Different Temperatures

	Recovery (SD, %)	Relative Ratio of Recovery			
Rise + Hold Time (min):	25.3				
Temperature (°C):	100	72	85	94	110
Resulting Hold Time (min):	12.3	19.5	17.1	15.3	6.1
Analytes					
4B	74.6	0.70	0.75	0.94	0.98
5B	83.9	0.85	0.86	0.95	1.01
4D	78.7	0.75	0.81	1.00	0.92
5D	80.0	0.84	0.90	0.97	1.04
6D	82.4	0.77	0.81	0.97	1.06
7D	83.8	0.86	0.89	0.97	1.01
8D	85.7	0.89	0.94	0.97	0.99
Average (SD)	81.3 (1.8)	0.81 (0.03)	0.85 (0.04)	0.97 (0.04)	1.00 (0.03)
	Relative Ratio of Recovery				
Hold Time (min):	0				
Resulting Rise Time (min):	13.0	5.8	8.2	10.0	19.2
Analytes					
4B	0.90	0.46	0.61	0.71	0.74
5B	0.88	0.42	0.67	0.70	0.89
4D	0.89	0.40	0.60	0.66	0.84
5D	0.94	0.40	0.68	0.73	0.91
6D	0.92	0.35	0.59	0.68	0.87
7D	0.96	0.45	0.70	0.83	0.95
8D	0.98	0.48	0.83	0.85	0.97
Average (SD)	0.92 (0.04)	0.42 (0.04)	0.67 (0.03)	0.74 (0.05)	0.88 (0.03)

Condition: toluene-IPA 90:10 (v/v), power 600 W, solvent volume 5 mL.

Table 4. Recovery and Relative Ratio of Recovery of PCBs and PCDDs in Fly Ash Extracts by Different Hold Times (Temperature °C = 100)

	Recovery (SD, %)	Relative Ratio of Recovery			
Rise Time (min):	13.0	13.0	16.9	13.0	13.0
Hold Time (min): Analytes	12.3	0	8.4	22.3	32.3
4B	74.6	0.90	0.88	1.11	1.08
5B	83.9	0.88	0.94	1.05	1.08
4D	78.7	0.89	0.89	1.16	1.17
5D	80.0	0.94	0.94	1.33	1.21
6D	82.4	0.92	0.95	1.09	1.12
7D	83.8	0.96	0.97	1.10	1.10
8D	85.7	0.98	0.97	1.11	1.14
Average (SD)	81.3 (1.8)	0.92 (0.04)	0.94 (0.03)	1.14 (0.04)	1.13 (0.05)

Condition: toluene-IPA 90:10 (v/v), power 600 W, solvent volume 5 mL.

22.3 min led to an increase in the recoveries of PCBs and PCDDs, while the recoveries between 22.3 and 33.3 min did not yield any difference.

From the comparison of rise/hold time between 16.9/8.4 and 13.0/12.3 min, the recoveries at 13.0/12.3 min were higher than 16.9/8.4 min. Therefore, although the MAE was carried out for the same total duration of 25.3 min, hold time other than rise time was a more effective parameter for higher recoveries.

Effect of Microwave Power, Solvent Volume, and Vessel Turning

The effect of varying microwave power on the mean recoveries and the relative ratios of the analytes is illustrated in Table 5. An increase in power from 300 to 600 W produced a corresponding increase in the recoveries. Although hold time was proportional to strength, further increase in power to 1200 W did not improve recoveries. A rapid rise in temperature by 1200 W seemed to prohibit IPA from making toluene desorb the analytes between the sample matrices, owing to the faster evaporation of IPA resulting in condensation on the vessel's internal wall.

Table 5. Recovery and Relative Ratio of Recovery of PCBs and PCDDs in Fly Ash Extracts by Different Powers and Solvent Volumes, and by Vessel Turning or Not

	Recovery (SD, %)	Relative Ratio of Recovery	
Solvent Volume (mL):	5		
Power (W): Analytes	600	300	1200
4B	74.6	0.81	1.00
5B	83.9	0.89	0.97
4D	78.7	0.82	1.03
5D	80.0	0.86	1.01
6D	82.4	0.83	0.93
7D	83.8	0.84	0.95
8D	85.7	0.83	0.93
Average (SD)	81.3 (1.8)	0.84 (0.05)	0.98 (0.03)
	Relative Ratio of Recovery		
Power (W):	600		
Solvent Volume (mL): Analytes	1.3	2.5	7.5
4B	0.93	0.78	0.91
5B	0.82	0.90	0.98
4D	0.86	1.05	0.96
5D	0.72	0.94	1.03
6D	0.73	1.00	1.04
7D	0.74	0.99	1.03
8D	0.64	0.98	0.99
Average (SD)	0.78 (0.03)	0.95 (0.02)	0.99 (0.02)
	Relative Ratio of Recovery		
Power (W): Solvent Volume (mL):	600 5		
Turning, Temp (°C): Analytes	Turning, 72	Not turning, 72	Not turning, 100
4B	0.68	0.72	0.88
5B	0.79	0.85	0.97
4D	0.74	0.83	1.02
5D	0.78	0.85	1.07
6D	0.75	0.80	1.07
7D	0.76	0.79	1.05
8D	0.76	0.70	1.02
Average (SD)	0.75 (0.05)	0.79 (0.04)	1.01 (0.03)

Condition: toluene-IPA 90:10 (v/v), rise + hold time 25.3 min.

The effect of varying solvent volume on the recoveries and the relative recovery ratios of the analytes is also illustrated in Table 5. Solvent volume had no significant effect on the recoveries except for 1.3 mL, where the volume was not sufficient to ensure that the sample was immersed after MAE.

As the electric field is generally non-homogeneous in the cavity, the vessels are placed on a turntable. However, in this study, regardless of the vessel's being turned or not, the recoveries did not differ at 72 and 100°C, respectively. Therefore, 600 W of power was sufficient to heat the samples homogeneously.

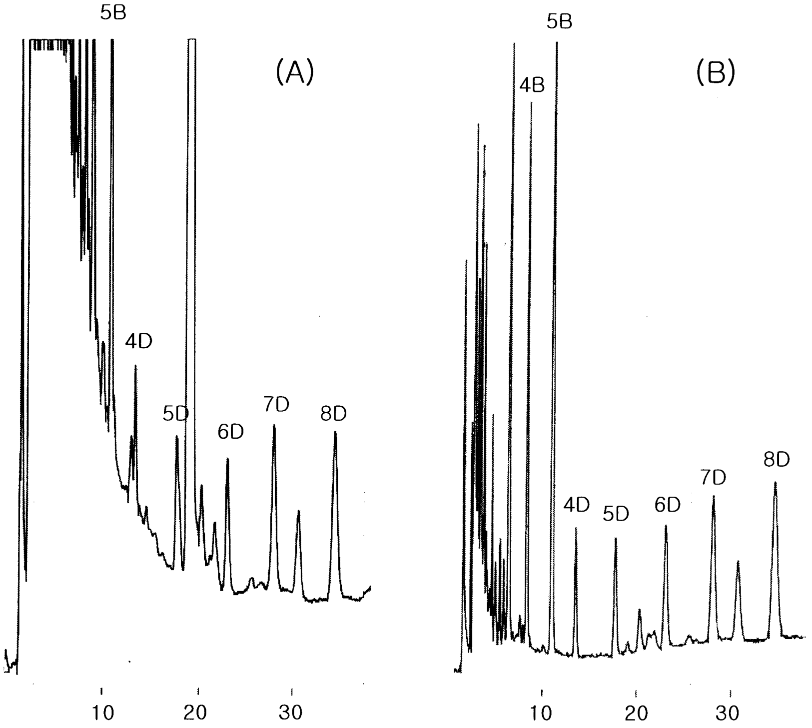

Figure 1. HPLC-UV chromatograms of fly ash sample extracted by MAE, followed by (A) no clean-up and (B) multi-layer silica gel column clean-up. Conditions: temperature, 100°C; toluene-IPA, 90:10 (v/v); total time, 25.3 min. The assignment of peaks are as follow: 4B = 2,2′,4,5′-tetrachlorinated biphenyl; 5B = 2,3,4,5,6-pentachlorinated biphenyl; 4D = 1,2,3,4-tetrachlorinated dibenzodioxin; 5D = 1,2,3,4,7-pentachlorinated dibenzodioxin; 6D = 1,2,3,4,7,8-hexachlorinated dibenzo-dioxin; 7D = 1,2,3,4,6,7,8-heptachlorinated dibenzodioxin; 8D = OCDD.

Removal of Chromatographic Interferences

During the microwave-assisted extraction (MAE) process, all the extracts were analyzed using HPLC-UV. After the extractions, the quantification of the extracts could not be carried out without multi-layer silica gel column clean-up (Figure 1 (A)); chromatographic interferences made quantification inaccurate (Figure 1 (B)). It has been previously reported that interferences could be removed using multi-layer silica gel column clean-up (11,12,15,16).

CONCLUSION

The goal of this study was to evaluate the effect of extraction parameters on efficient MAE of PCBs and PCDDs from fly ash. More efficient MAE was achieved when toluene-IPA mixture 90 : 10 (v/v) as extraction solvent was used rather than toluene alone. IPA was used to make toluene solvate and desorb analytes between sample matrices as well as to absorb the microwave energy and transfer it to the sample. The temperature was the important variable, while the effects of solvent volume and vessel's turning were minimal. Hold time, rather than rise, contributed to the extraction of PCBs and PCDDs under constant total time. An increase in power from 600 to 1200 W did not enhance the recoveries, although hold time was increased. The overall optimum MAE conditions for the extraction of PCBs and PCDDs from fly ash were 5 mL of toluene/IPA (90 : 10, v/v) as the extracting solvent, and an extraction temperature of 100°C for 35.3 min.

Although the optimal temperature and extraction solvents (100°C and toluene-IPA 90 : 10, v/v) for MAE could not always be deduced directly from those used in conventional procedures, it was possible to perform MAE with the similar temperature and solvent composition that had been prescribed for the other extractions, such as EFLE and PFE, in previous papers. Multi-layer silica gel column clean-up was required to remove interferences, which had a significant impact on the determination of PCBs and PCDDs from solid matrices.

This work represents an important evaluation of MAE in isolating adsorptive analytes from a strong interaction with a matrix.

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