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Gas-liquid microextraction coupled with magnetic-assisted dispersive solid-phase extraction clean-up for multi-residue pesticide analysis in fatty foods of animal origin

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ABSTRACT

An effective sequential clean-up method by coupling gas-liquid microextraction (GLME) and magnetic-assisted dispersive solid phase extraction (d-SPE) termed as GLME-MA-*d*-SPE has been developed for multi-residue pesticide analysis in different fatty foods of animal origin. GLME is applied as a primary clean-up step to remove low-volatile interferences, followed by a secondary clean-up technique through adsorptive removal using d-SPE to eliminate other co-extracts like organic acids in fatty biological samples. As much as 99.3% of lipid substances were effectively eliminated by this powerful clean-up method, and the chromatographic analysis by GC-MS showed at least two orders of magnitude reduction for peaks of interference. Analytical results verified the accuracy and precision of this method with recoveries of 50 pesticides ranged from 60.5% to 119.7%, and RSDs of less than 20%. Permethrin was present in salmon, pork and egg samples, but the concentrations were within the maximum residue levels (MRLs) permitted by both national and international regulations. The GLME-MA-*d*-SPE technique minimizes matrix effects, and it exhibits significant potential as an analytical technique of food safety control systems for broad-spectrum screening trace-level environmental pollutants in complex biological matrices.

1. Introduction

The application of pesticides plays an indispensable role on increasing the yield of agricultural crops to produce adequate food supply for satisfying the escalating world population. Without proper management, the escalating production and consumption of a multitude of pesticides may release or leak into the environment, thus causing pollution. Living organisms are greatly exposed to these anthropogenic pesticides in the environment, which may eventually enter the human body and bring about detrimental impact to human health. Numerous literatures have related the toxicities of pesticides due to high exposure, which include lung cancer (Bonner et al., 2016), neurodegeneration (Sánchez-Santed et al., 2016), and male reproductive disorders (Svingen et al., 2018). Regulations like maximum residue levels (MRLs) that set by European Commissions or tolerances by the United States Environmental Protection Agency (U.S. EPA) are enforced to assess pesticide residues in foods and to ensure human exposure is maintained at a minimum risk. Therefore, constant monitoring endeavors to evaluate the concentration of pesticides in food samples are mandatory.

The heterogeneity of pesticides encompasses several chemical classes, which involve a diversity of structural and physicochemical properties. Organochlorine pesticides (OCPs), also known as legacy

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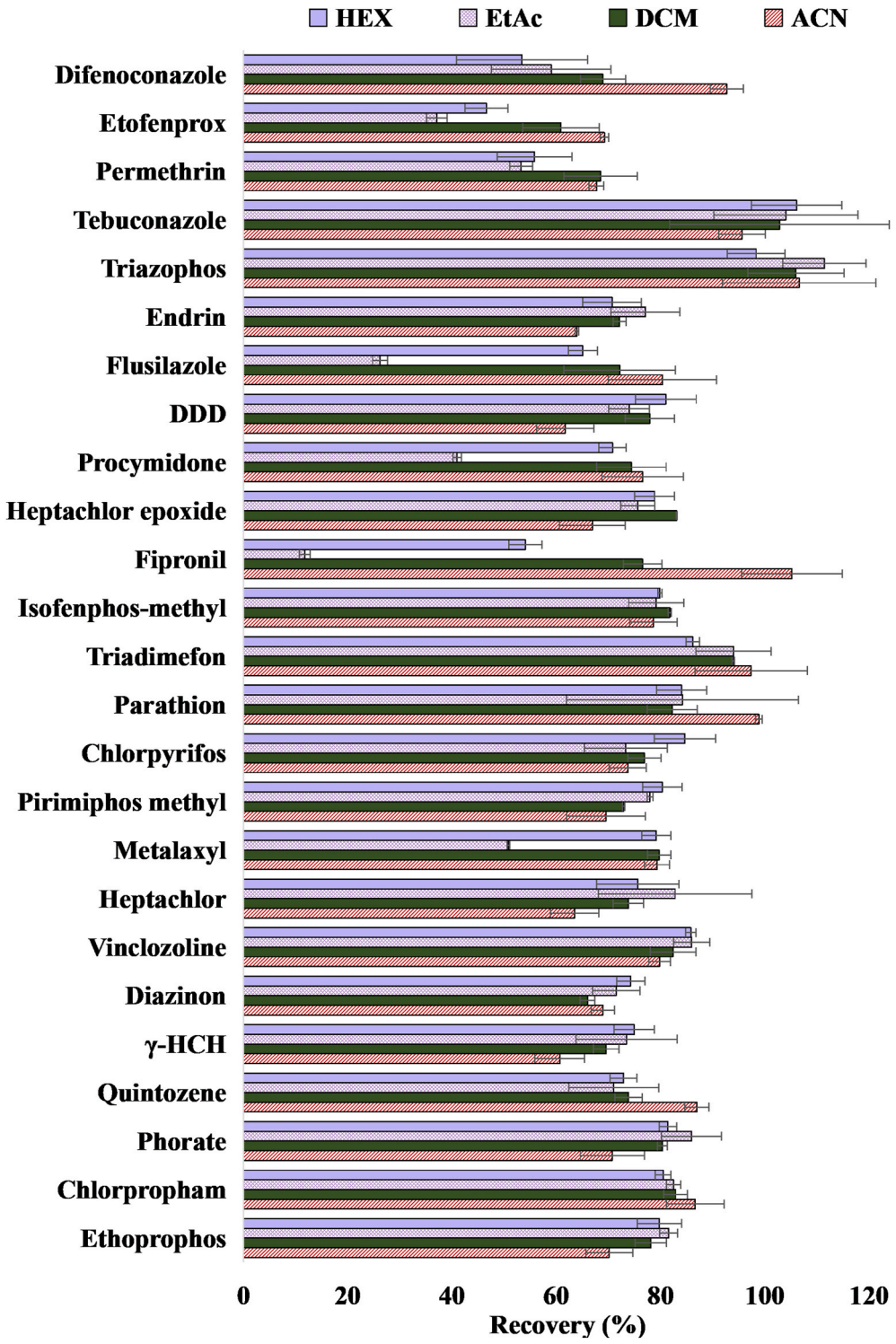

Fig. 1. Comparison of the recoveries of selected target analytes extracted by using different organic solvents (acetonitrile (ACN), dichloromethane (DCM), hexane (HEX) and ethyl acetate (EtAc)) for evaluation of extraction efficiency.

pesticides, still can be detected in various environmental and foodstuffs at trace concentration up till today (Qu et al., 2019), despite being banned for decades. Other categories of pesticides such as organophosphorous, pyrethroids, triazines, conazoles and carbamates were employed to replace the agricultural application of OCPs. Some of these pesticides can be defined as emerging pesticides according to the NORMAN list of emerging substances. Many of these pesticides known as current-use pesticides (CUPs) can be identified in biota in remote regions like the Arctic, accentuating the significance of monitoring multi-class pesticides in biological samples (Balmer et al., 2019).

Multi-residue analysis is the recent trend for determining the heterogeneous pesticide residues in foods. In particular, the determination of hazardous chemicals in fatty biological samples is essential, as lipophilic organic contaminants like chlorinated pesticides and some pyrethroids tend to accumulate in human and animal fat tissues and amplified through the food chain (Sapozhnikova, 2014). Yet, high lipid

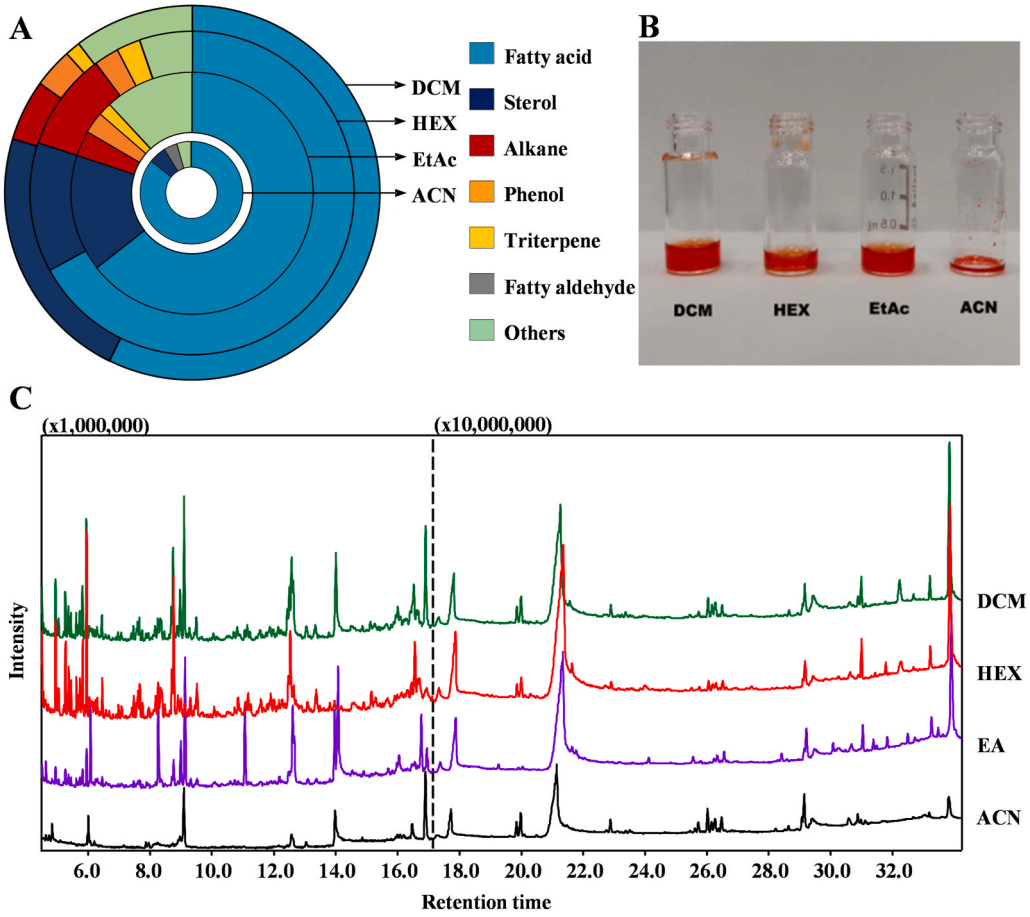

Fig. 2. (A) A diagram formed by different sizes of annuli depicting the difference in quantity of co-extracts being extracted by using different organic solvent. (B) An illustration of the remaining lipid content after evaporation of distinctive extraction solvent. (C) TIC chromatograms by scan mode in GC-MS analysis indicate much lesser and lower peaks for ACN extracts.

interferences and co-extracts in fatty biological samples significantly influence the precision of quantitative analysis, and may cause damage to the detection instruments (Yoon et al., 2015). Therefore, an effective clean up step is indispensable in order to obtain the most accurate result to determine the trace-level pesticide residues in these complex food samples. Conventional clean up techniques like gel permeation chromatography (GPC) is effective in separating the high-molecular mass lipids from low molecular mass analytes, while solid phase extraction (SPE) is a useful purification step to eliminate interfering co-extracts by adsorption using different adsorbents (LeDoux, 2011). However, time-consuming, labor intensive and the consumption of relatively large sample amount, toxic reagent and organic solvent are some of the shortcomings, which make these methods less efficient and environmentally-unfriendly.

Gas liquid microextraction (GLME) technique is a redesigned version of a technology previously named as gas purge microsyringe extraction (GP-MSE) (Yang et al., 2011). It is effective in extracting all kinds of volatile and semi-volatile organic compounds from various matrices (He et al., 2015; Jin et al., 2018, 2020; Zhao et al., 2019). The present study intends to use GLME technique as a primary clean-up method, and subsequently combine with dispersive SPE (d-SPE) as a secondary clean up step to effectively remove lipid interferences in fatty food matrices. The analogous clean-up effectiveness of d-SPE has been justified previously in various published articles, which was normally incorporated in pre-treatment techniques like the quick, easy, cheap, effective, rugged and safe termed as QuEChERS (Al-Nasir et al., 2020) and dispersive liquid-liquid microextraction (DLLME) (Farajzadeh et al., 2017). In short, d-SPE is a comparatively more time- and organic solvent-saving clean-up technique than the traditional cartridge SPE (c-SPE) (Steinbach & Schwack, 2014). Recently, magnetic nanoparticles (MNPs) have been exploited as an effective tool in the field of regenerative medicine, drug delivery and various environmental applications (Gao et al., 2015). Due to their magnetic properties, MNPs were used in this study for the effortless isolation of d-SPE prior to instrumental analysis.

This research aims to effectively eliminate coisolated lipid interferences from the raw extracts of fatty foods of animal origin, and to ensure the accurate determination of multi-residue pesticides in these challenging matrices. Multi-class pesticides comprise legacy pesticides as listed in the U.S. EPA Priority Pollutant List and emerging pesticides were chosen as the analytes in this study. Among which, some of the selected pesticides were also listed under EU Proficiency Test (EUPT) by the European Union Reference Laboratory (EURL) as target list of mandatory analytes to be monitored in food of animal origin and commodities with high fat content (EUPT-AO). After ultrasonic extraction, GLME is employed as a primary clean-up step to remove low-volatile interferences, followed by a secondary clean-up technique to eliminate other co-extracts by adsorptive removal using d-SPE. Multi-class pesticide residues in these fatty food matrices are detected by GC-MS. Several considerations that may affect analytical accuracy such as extraction solvent, temperature for GLME clean-up and the types and quantities of d-SPE sorbents are optimized. The strengths of combining GLME and d-SPE clean-up techniques for efficient lipid removal to achieve multi-residue pesticide analysis are also evaluated.

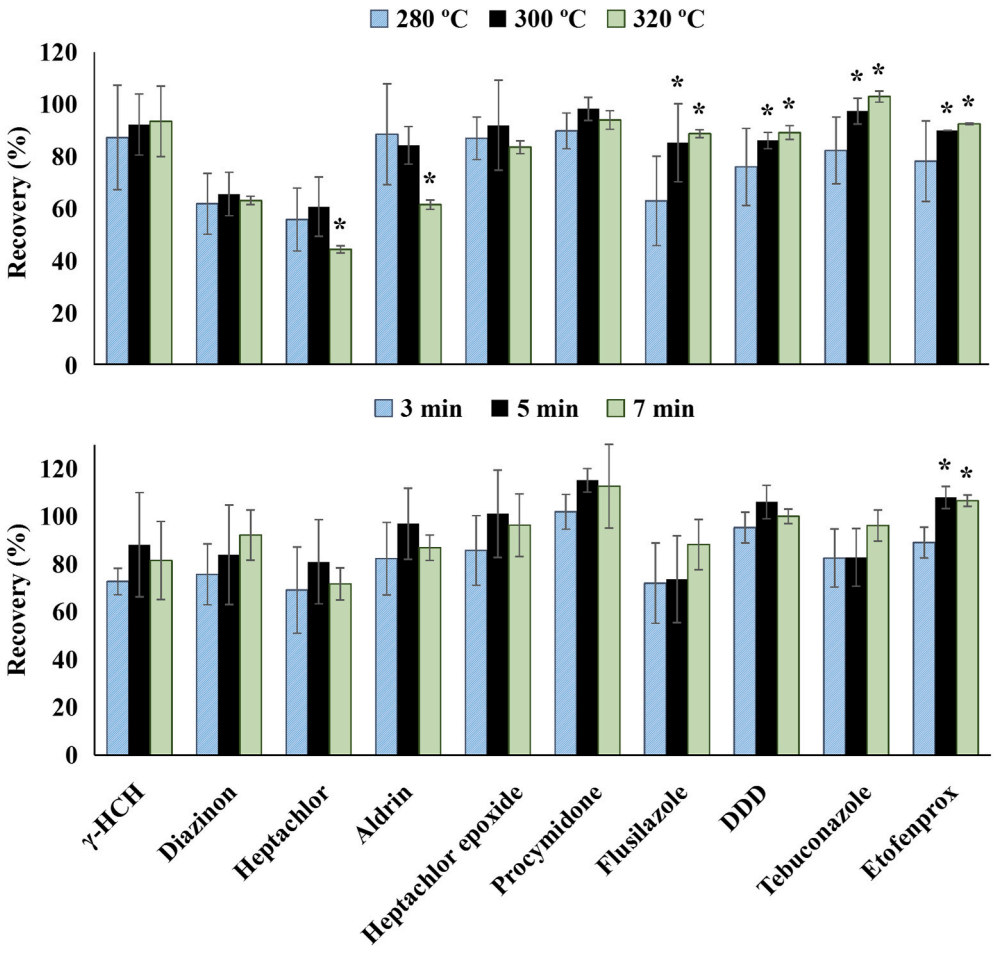

Fig. 3. Optimization of the GLME parameters for primary clean-up. The performance was evaluated based on assessing the recoveries of pesticides according to different (a) temperature and (b) time of GLME treatment. *Asterisk* signifies the statistically significant difference ($p < 0.05$) between GLME extraction conditions.

2. Materials and methods

2.1. Chemicals and reagents

All organic solvents (HPLC grade) including acetonitrile (ACN), dichloromethane (DCM), hexane (HEX) and ethyl acetate (EtAc) were purchased from Thermo Fisher Scientific (Waltham, MA, USA). Details of the pesticide reference standards and the preparation procedures can be found in Supplementary Materials and Methods section. Different sorbents including N-propylethylendiamine or primary secondary amine (PSA), graphitized carbon black (GCB), C18 (bonded octadecyl silica), florisil (magnesium silicate), alumina (Al_2O_3) and silica gel were purchased from Shanghai BioSun Sci & Tech Co., Ltd.

2.2. Sample pre-treatment procedures

Different foods of animal origin (pork, beef, salmon and egg) were purchased from a local supermarket in Yanji city, Northeast China. Approximately 0.5 kg of each food types were immediately homogenized and then freeze-dried for 24 h. All samples were stored in amber bottle and kept in the freezer at - 20 °C until further analysis.

A total of 0.75 g (±0.01 g) sample was accurately weighed and put into a 5 mL centrifugation tube. Each sample was spiked with 150 ng surrogate standards (150 ng mixed target analytes was spiked in fortified samples), followed by the addition of 1.5 mL acetonitrile. After ultrasonic extracted for 15 min, 1 mL of the supernatant was injected into GLME under optimized condition for primary clean-up. The optimized amount of MNPs and d-SPE adsorbent were added into the post-GLME extract for secondary clean-up. The solution mixture was vortexed for 30 s to ensure both the MNPs and adsorbent were fully dispersed in the solution. An external magnetic field was applied by using a magnet to isolate the aggregated MNPs and adsorbent, and the purified solution was withdrawn. It was concentrated under a gentle flow of nitrogen gas to 500 μL. Internal standard was added and it was subjected to GC-MS analysis.

2.3. Determination of matrix effect, lipid content and lipid removal efficiency

Method matrix effect (ME%), also can be referred to the ionization signals of enhancement or suppression for each analytes in instrumental analysis, was assessed according to the methodology published elsewhere (Dong et al., 2019). Briefly, matrix effects (ME%) were calculated based on the comparison between the seven-point slope of matrix-matched calibration curves and the corresponding slope of solvent calibration curves, as shown in equation (1).

$$\mathrm{ME}\ \ (\%) = 100 \times \left(\frac{S_m}{S_s} - 1\right) \tag{1}$$

where S_m and S_s represent the slopes of calibration curve in sample matrix and solvent, respectively.

The method for determining lipid content in fatty food of animal origin is performed according to a published method with slight modification (Morrison et al., 2016). In brief, lipids of 9.0 g homogenized samples were ultrasonic extracted by 18 mL ACN, and allowed to incubate for 15 min. Samples were then centrifuged at 6000 rpm (4226

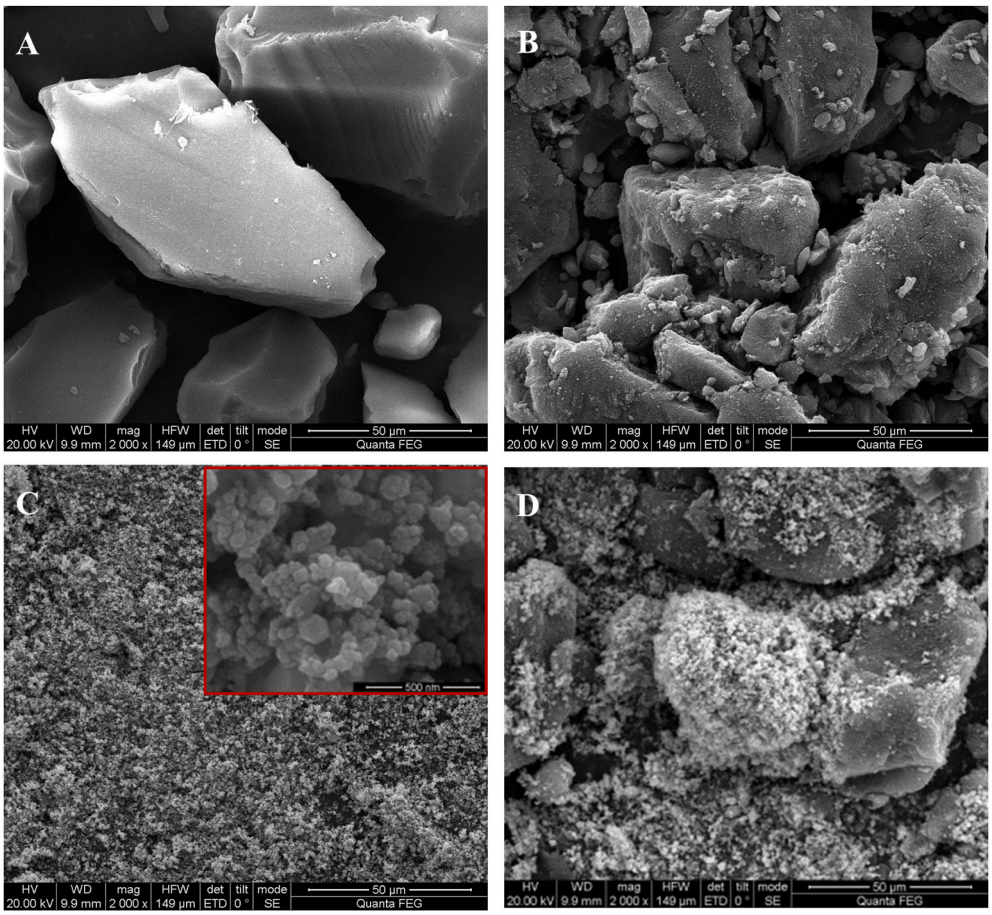

Fig. 4. SEM images of (A) Primary Secondary Amine (PSA); (B) Graphitized Carbon Black (GCB); (C) Magnetic Fe_3O_4 nanoparticles (MNPs) and the subfigure indicates MNPs under higher magnification; (D) Agglomerate of PSA/GCB/MNPs.

rcf) for 3 min, and 2 mL of the supernatant aliquot was transferred to a pre-weigh aluminium foil. Gravimetric analysis was performed to determine the lipid percentage of foods by dividing lipid weight by initial weight for each sample. An equal volume of vegetable oil was used as a lipid surrogate throughout lipid analysis to normalize the potential losses.

The lipid elimination rate (LER) is evaluated based on the difference in lipid weight before and after clean-up, and the calculation is shown in equation (2).

$$\text{LER}\ \ (\%) = 100 \times \left(1 - \frac{W_c}{W_r}\right) \tag{2}$$

where W_c and W_r represent the weight of lipids in post-clean-up extract and raw extract, respectively.

2.4. Instrumental analysis

Multi-class pesticide residues were analyzed using a GC2010 gas chromatograph (Shimadzu, Kyoto, Japan) with a DB-5 fused-silica capillary column (30 m × 0.25 mm × 0.25 µm) coupled to a QP2010 mass spectrometer (Shimadzu, Kyoto, Japan). Helium with a purity of 99.999% was used as the carrier gas. Sample was injected in splitless mode with an injection temperature of 290 °C, and the carrier gas was held at a constant flow rate of 1.0 mL min^{-1}. The GC-MS interface temperature was held at 280 °C and the ion source temperature was set at 200 °C. The electron energy for ionization was set to 70 eV. The initial oven temperature of 40 °C was ramped at a rate of 30 °C min^{-1} to 130 °C, followed by a 5 °C min^{-1} ramp to 250 °C, then a final ramp of 10 °C min^{-1} to 300 °C. Selected ion monitoring (SIM) mode with a sampling rate of 1.0 s was used.

2.5. Quality assurance/quality control

All analytical processes were performed according to quality assurance and quality control measures. Amber glass bottles were used for the storage of food samples. In order to eliminate the risk of contamination, all glassware was rinsed with chromatographic grade organic solvents and subsequently heated at 400 °C for 8 h before use. For our QC/QA program, standards were run to check for instrumental performance before analysis. Quantification was done by internal standard method, and the correlation coefficients (r^2) of calibration curves were assured to be higher than 0.99. Two surrogate standards (TCMX and TEB-D9) were spiked in all matrices of fatty food samples to evaluate the accuracy of analytical procedures. The recoveries of surrogate standards ranged between 60.6 and 90.8%, and 76.1–115.6% (average values of 71.8 and 91.2%) with relative standard deviation (RSDs) of less than 19.1 and 15.2%, respectively, which justify the accuracy of analytical results.

2.6. Evaluation of analytical performance

An assessment on the analytical performance of the proposed method was implemented according to European Union SANTE/11813/2017 in order to verify the method performance and to guarantee the quality of the analytical procedure (European Commission, 2017). In this study, a standard mixture comprised a total of 50 multi-class pesticides consisted of organochlorine insecticides, organophosphate insecticides, pyrethroid insecticides, pyrazole insecticides, carbamate herbicides, amide fungicides, and conazole fungicides were used to evaluate the

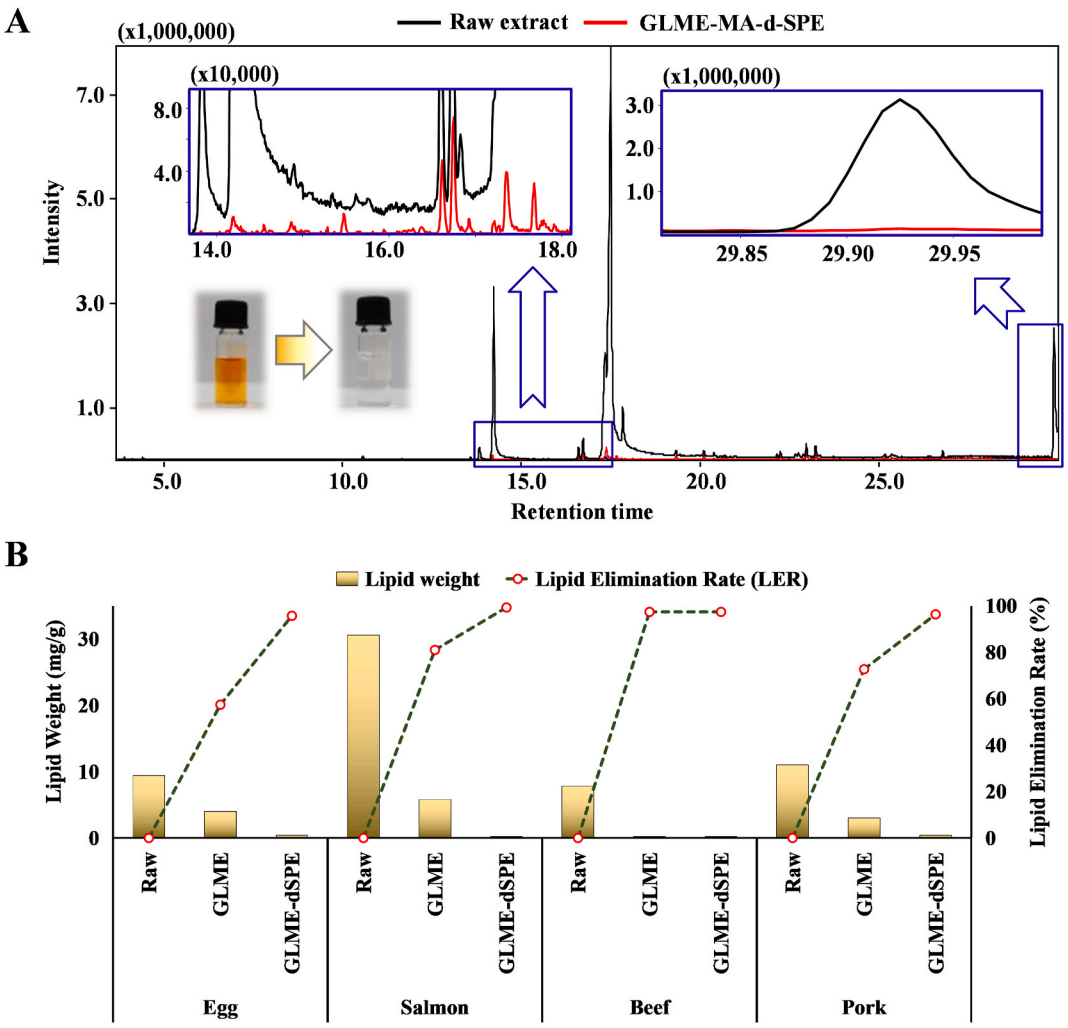

Fig. 5. An evaluation of the Lipid Elimination Rate (LER) by comparing the (A) TIC of GC-MS operating in scan mode and (B) gravimetric analysis before and after GLME-MA-*d*-SPE sequential clean-up method.

reproducibility, linearity, method limit of detection (MLOD) and method limit of quantification (MLOQ) of this method. The recovery and precision (RSD%) of this method were investigated by spiking 200 ng g^{-1} in different food samples (n = 3). The retention times and three characteristics ions including one target and two qualifier ions selected for qualitative and quantitative determination of target compounds are listed in Table S1. Each target compound was confirmed using the retention time match and the intensity ratio of characteristic ions. The concentrations of pesticides ranged from 10 to 1000 ng g^{-1} in spiked samples were used for linearity evaluation. The LOD was calculated as three times the signal-to-noise (S/N) ratio, and the LOQ corresponds to the concentration when the S/N is 10.

2.7. Data analysis

The plot of data and significant analysis were performed using Microsoft Excel 2019 (Redmond, WA, USA). The extraction efficiency of multi-class pesticides influenced by solvent type and GLME parameters (temperature and time) were assessed using one-way analysis of variance (ANOVA) with post-hoc comparisons using Tukey HSD test. A value of $p < 0.05$ was considered to indicate a statistical significance.

3. Results and discussion

3.1. Characterization of the synthesized magnetic nanoparticles

The magnetization of the MNPs can be found in Fig. S1, and the SEM characterization of the MNPs is shown in section 3.2.4. The role of MNPs in this study is merely for forming aggregation with the d-SPE sorbents and then fulfil the purpose of convenient isolation, hence only these two properties were characterized.

3.2. Optimizations of the sample pre-treatment method

3.2.1. Selection of extraction solvent

Different commonly used extraction solvents including ACN, DCM, HEX, and EtAc were experimented to evaluate the extraction efficiency of multi-class pesticides in tested foods of animal origin. In general, the recoveries of all pesticides were identical for all extraction solvents, as can be observed in Fig. 1. Satisfactory recoveries of 60.7–106.6%, 60.9–105.9%, 46.6–106.1%, and 41.0–111.4% (except Fipronil, Flusilazole and Ethofenprox with lower than 40% recoveries) for ACN, DCM, HEX and EtAc, respectively, were achieved, which are in accordance with previous study (Chatterjee et al., 2016). Despite the similar extraction efficiency, the influence of matrix constituents that could adversely affect the chromatographic analysis is an important element to be taken into account. The total ion chromatograms (TIC) of the

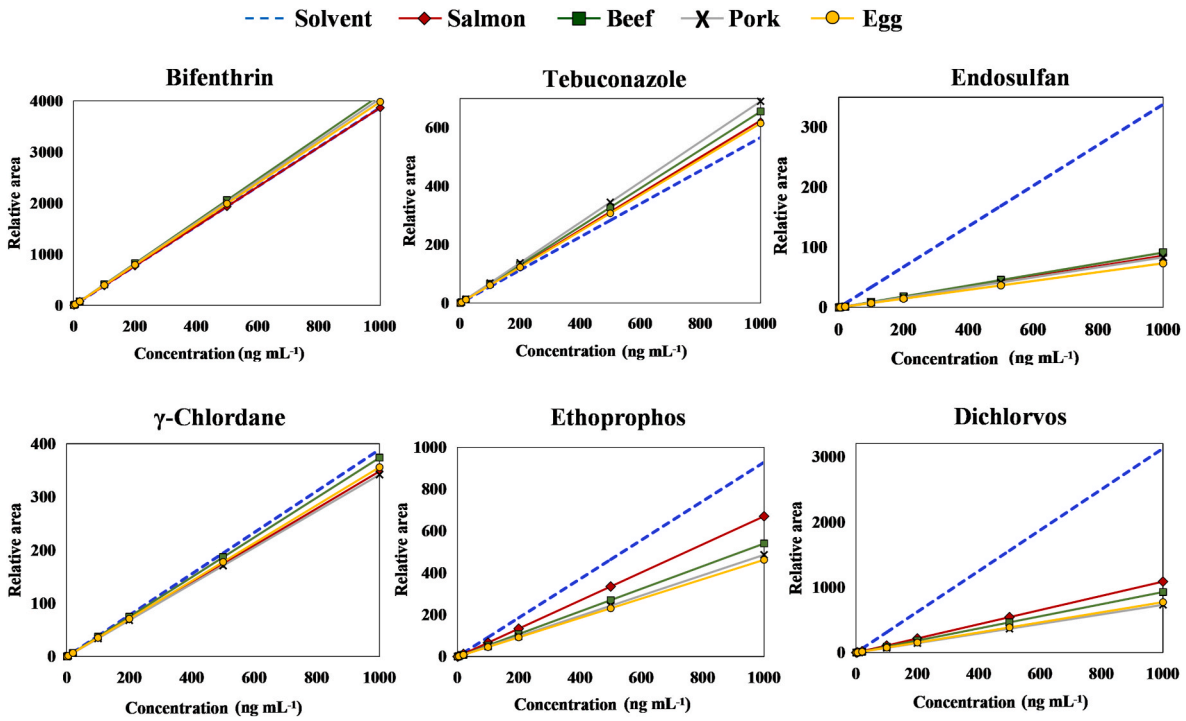

Fig. 6. Matrix effects (ME%) of selected pesticides by comparison between the calibration curves prepared in solvent (dash line) and in different matrices (solid lines with distinctive markers representing each food matrix). Sub-figures (both top and bottom rows) from the left to the right column represent analytes with soft, medium and strong ME%, respectively.

extracts with sample amount of approximately 1 g mL^{-1} were obtained through scan mode in GC-MS analysis to compare the quantity of co-extracts. The annular ring size and color in sub-Fig. 2(A) indicate the ratio amount and profile of co-extracts being detected, respectively. All the sub-figure 2(A), (B) and (C) justified that substantially lesser substances were extracted by ACN compared to DCM, HEX and EtAc as the extraction solvents. Considering the least interfering substances that co-extracted from fatty foods, ACN was eventually selected as the extraction solvent for subsequent experiments.

3.2.2. GLME clean-up parameters

The application of GLME proposed herein is intended for the clean-up of fatty foods of animal origin by separating analytes from non-volatile interfering substances through difference in volatility. The time and temperature required for clean-up, which have been justified previously as the most influential parameters, were sequentially evaluated. Based on the optimization results (Fig. 3), the clean-up time of 5 min and temperature of 300 °C were chosen for further experiments.

3.2.3. Removal efficiency of lipid-soluble interferences by GLME

After ultrasonication extraction, the purification performance of the GLME clean-up technique to remove lipid-soluble interferences in fatty foods of animal origin was assessed. The lipid elimination rate (LER) was examined through gravimetric analysis to evaluate the clean-up effects. LER values of 97.4% were successfully achieved in beef, followed by 81.0%, 72.7% and 57.5% in salmon, pork and egg extracts, respectively. Nonetheless, traces of highly volatile organic acids and other co-extracts still pose an unfavorable impact on the accurate quantification of multi-class pesticides in fatty foods. The utilization of clean-up adsorbent is significant for minimizing the damage to the analytical instrumentation, liners and columns, hence assuring the preventive maintenance for instrumentation (Hakme et al., 2018). Therefore, a secondary clean-up step that involves d-SPE approach is mandatory to effectively purify high-lipid samples.

3.2.4. Optimal amount of MNPs and d-SPE adsorbent

Subsequent to primary clean-up by using the GLME technique, a secondary purification step through d-SPE was implemented. In this study, different kinds of adsorbent (alumina, silica gel, florisil, C18, GCB and PSA) were sequentially examined and selected based on the chemical structure or properties of co-extract after GLME clean-up. According to the results displayed in Fig. S2, a significant clean-up performance was achieved by using PSA alone, and most of the recoveries for detected pesticides were in between 60 and 120%. Previous studies have proven that PSA, a weak anion exchanger, is effective in removing organic acids, fatty acids, sugars and ionic lipids, and most importantly, it generally does not adsorb many pesticides (Kim et al., 2019). Thus, PSA was selected as the primary d-SPE sorbent in this study, and the most ideal quantity was sequentially examined. Co-extracts were gradually being eliminated through increasing the amount of PSA, but the clean-up effect was less obvious after adding 40 mg. Hence, 40 mg PSA was chosen as the optimized quantity. Nonetheless, some peaks in the TIC of salmon sample remained unchanged even if PSA was added (data not shown), indicating that these compounds cannot be adsorbed by PSA. These compounds were identified as sterols with 94% similarity based on the NIST database, which can be found abundant in biological samples (Copeman & Parrish, 2004).

Due to the excellent performance of GCB to remove sterols and pigments (Wilkowska & Biziuk, 2011), it was selected as the secondary d-SPE clean-up sorbent in high-lipid food samples, and the adequate amount of GCB was sequentially evaluated. According to the removal efficiency, the magnitude of sterols being eliminated was proportionate to the amount of GCB used. However, lower recoveries of hexachlorobenzene, terbufos, metalaxyl and pendimethalin were observed when 50 mg of GCB were added (Fig. S8). This phenomenon is in accordance with previous studies which stated that GCB not only can adsorb matrix components, but also pesticides with planar ring structure that has high affinity towards GCB due to its great specific surface (Bruzzoniti et al., 2014). Consequently, 40 mg of GCB plus 40 mg of PSA were selected as the optimal amount of d-SPE adsorbents for secondary

Table 1
Matrix effect (ME%), method limit of detection (MLOD, ng g^{-1}) and method limit of quantification (MLOQ, ng g^{-1}) for 50 legacy and emerging pesticides in the four tested food matrices (salmon, beef, pork and egg).

	ME%				MLOD (ng g^{-1})				MLOQ (ng g^{-1})			
	Salmon	Beef	Pork	Egg	Salmon	Beef	Pork	Egg	Salmon	Beef	Pork	Egg
Dichlorvos	−65	−70	−77	−75	0.67	3.02	1.91	2.18	2.22	10.05	6.36	7.26
2-Phenylphenol	−36	−52	−53	−57	0.32	0.29	0.23	0.10	1.06	0.98	0.77	0.34
Ethoprophos	−28	−42	−48	−50	0.92	2.08	1.24	0.56	3.08	6.94	4.14	1.86
Chlorpropham	−22	−37	−40	−44	0.65	1.84	1.47	0.79	2.18	6.12	4.92	2.62
Phorate	−27	−40	−44	−46	0.74	2.25	1.80	0.82	2.47	7.49	6.02	2.72
α-HCH	−35	−45	−51	−50	1.00	2.94	2.67	4.18	3.35	9.81	8.89	13.95
Simazine	−18	−26	−30	−33	4.35	2.53	2.34	1.31	14.51	8.44	7.81	4.35
β-HCH	−16	−22	−29	−37	1.05	3.08	1.90	1.92	3.49	10.28	6.35	6.38
Quintozene	−29	−31	−39	−43	1.24	3.23	3.16	2.52	4.12	10.77	10.55	8.39
γ-HCH	−19	6	−20	−10	3.49	5.58	5.22	5.63	11.64	18.60	17.41	18.76
Terbufos	−28	−32	−40	−42	1.14	3.22	1.89	1.31	3.80	10.73	6.29	4.38
Diazinon	−33	−37	−47	−46	0.93	2.47	1.70	1.21	3.09	8.24	5.67	4.03
Pyrimethanil	−25	−27	−40	−35	0.36	0.52	0.63	0.21	1.21	1.74	2.09	0.70
Isazofos	−34	−31	−47	−40	2.03	3.46	2.16	2.11	6.76	11.55	7.18	7.02
Vinclozoline	−42	−37	−57	−48	3.05	5.98	5.34	2.33	10.18	19.92	17.79	7.77
Heptachlor	−46	−35	−54	−47	2.11	3.98	2.92	1.36	7.04	13.27	9.73	4.53
Metalaxyl	−40	−33	−51	−43	1.47	1.05	1.13	0.67	4.91	3.49	3.75	2.24
Chlorpyrifos	−30	−33	−44	−40	3.66	7.49	7.80	6.99	12.21	24.97	26.01	23.31
Aldrin	−40	−45	−52	−54	1.90	3.86	3.33	1.61	6.34	12.87	11.10	5.36
Parathion	−14	−19	−29	−30	3.58	7.19	3.70	5.67	11.95	23.98	12.32	18.90
Triadimefon	−23	−22	−31	−27	4.45	3.55	4.54	2.04	14.83	11.83	15.13	6.81
Dicofol	43	25	4	28	1.52	1.82	2.64	0.68	5.07	6.06	8.78	2.26
Isofenphos-methyl	−14	−12	−20	−14	1.65	1.60	1.47	1.61	5.51	5.33	4.89	5.36
Pendimethalin	−4	−9	−19	−16	3.38	4.12	2.98	1.33	11.28	13.73	9.95	4.43
Fipronil	9	15	12	2	1.32	1.63	1.36	2.86	4.39	5.43	4.52	9.53
Heptachlor epoxide-A	−14	−14	−24	−19	2.87	3.16	2.95	1.06	9.56	10.52	9.82	3.53
Heptachlor epoxide-B	−18	−19	−21	−21	10.97	8.00	11.01	6.77	36.56	26.67	36.70	22.57
Procymidone	−13	−13	−21	−12	1.31	1.89	1.57	0.94	4.35	6.30	5.23	3.12
γ-Chlordane	−11	−4	−12	−9	0.98	1.17	1.22	0.55	3.27	3.92	4.07	1.82
α-Chlordane	−9	−3	−14	−6	0.96	1.45	1.71	1.22	3.19	4.84	5.70	4.07
Endosulfan	−74	−73	−75	−78	9.54	6.83	7.81	9.27	31.80	22.75	26.04	30.91
Hexaconazole	1	2	−3	5	2.49	1.96	1.81	1.15	8.32	6.52	6.02	3.84
DDE	−10	−7	−15	−1	0.85	0.84	1.16	0.55	2.85	2.80	3.86	1.83
Dieldrin	−7	3	−6	3	3.77	5.69	4.88	2.80	12.58	18.98	16.27	9.35
DDD	−7	−1	−7	−6	1.00	0.82	1.25	1.10	3.33	2.72	4.18	3.66
Flusilazole	−2	1	−2	0	0.58	0.82	0.63	0.37	1.93	2.72	2.10	1.23
Endrin	5	16	5	−3	2.39	2.65	2.79	1.70	7.98	8.83	9.31	5.67
DDT	7	13	7	1	2.18	0.81	0.71	0.61	7.26	2.71	2.36	2.04
Triazophos	10	17	13	5	2.37	1.98	1.13	0.66	7.89	6.59	3.76	2.21
Benalaxyl	−2	4	−1	0	1.58	0.70	1.01	0.59	5.28	2.33	3.36	1.98
Tebuconazole	10	16	22	9	3.70	3.46	2.40	2.94	12.32	11.52	8.00	9.79
Piperonyl butoxide	7	14	9	6	0.62	0.81	0.57	0.47	2.07	2.69	1.90	1.56
Bifenthrin	0	6	5	3	0.86	2.44	1.44	1.09	2.86	8.13	4.80	3.64
Methoxychlor	6	22	19	14	0.94	2.70	1.39	2.65	3.12	9.01	4.64	8.82
Fenpropathrin	−1	12	11	8	1.58	1.37	1.97	1.26	5.25	4.57	6.58	4.20
Permethrin	−9	0	−3	−4	1.21	0.84	0.32	0.80	4.04	2.81	1.07	2.68
Pyridaben	−22	−3	−10	−3	1.95	0.55	0.52	0.47	6.50	1.83	1.75	1.58
Etofenprox	−24	−8	−23	−11	0.23	0.25	0.23	0.09	0.76	0.83	0.76	0.30
cis-Difenoconazole	−24	−5	−16	−12	1.62	1.32	1.40	1.40	5.41	4.39	4.68	4.68
trans-Difenoconazole	−12	−8	−13	−1	1.16	1.01	1.07	0.70	3.86	3.37	3.56	2.32

clean-up (Fig. S7).

MNPs were utilized in this study for the easy isolation of d-SPE adsorbents. The d-SPE adsorbents were endowed with magnetic property when they were integrated with MNPs during the mixing process, in which MNPs completely wrapped around the surfaces of adsorbents resulting in a physical adsorption interaction termed the “aggregation wrap” mechanism (Fig. S3) (Sun et al., 2018). The amount of MNPs needed for the effective isolation of d-SPE adsorbents was determined. Observational results showed that 25 mg was adequate to fully isolate the d-SPE adsorbents (Fig. S9), and MNPs were recycled for subsequent batches of experiments. The SEM images of d-SPE adsorbents and MNPs were shown in Fig. 4 (A-C), and Fig. 4 (D) demonstrates the aggregation of adsorbents and MNPs.

3.3. Lipid elimination rate (LER) of the GLME-MA-d-SPE clean-up method

The LER of the optimized sequential clean-up technique by combining GLME with magnetic-assisted d-SPE (GLME-MA-*d*-SPE) was systematically determined through GC-MS operating in scan mode and gravimetric analyses. As shown by the GC-MS TIC in Fig. 5(A), approximately 100-fold reduction indicated by the two orders of magnitude lower peaks was achieved by employing the sequential GLME-MA-*d*-SPE clean-up method. The raw extract of salmon sample with orange colour displayed in sub-Fig. 5(A) was successfully purified to a colourless solvent, verifying the strength of this combined clean-up technique. Similarly, gravimetric analysis (Fig. 5(B)) also confirmed the effectiveness of GLME-MA-*d*-SPE clean-up method to eliminate lipid interferences in fatty foods by showing the LER values of 99.3%, 97.4%, 96.4% and 95.7% for salmon, beef, pork and egg samples, respectively. A comparison of lipid elimination rates between different purification

Table 2
Recovery and relative standard deviation (RSD) for 50 legacy and emerging pesticides in spiked fatty foods of animal origin.

	Beef						Salmon						Pork						Egg					
	Spiked level (ng g^{-1})																							
	20		50		200		20		50		200		20		50		200		20		50		200	
	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)
Dichlorvos	105.3	9.4	104.9	1.1	91.3	8.6	80.9	13.6	95.8	11.2	83.0	20.2	80.8	8.3	76.0	18.1	104.3	8.4	95.6	1.1	104.9	3.0	94.3	10.0
2-Phenylphenol	103.8	12.8	119.7	0.5	80.6	8.3	111.4	9.0	110.3	2.8	97.9	6.9	104.5	5.0	102.0	5.3	77.1	4.6	96.8	10.0	115.1	3.6	116.7	2.3
Ethoprophos	82.1	5.7	89.1	13.6	91.8	11.3	101.4	4.6	95.5	5.6	108.5	7.0	98.8	2.7	93.4	1.7	86.0	9.9	72.4	4.7	105.3	11.8	110.1	3.5
Chlorpropham	85.3	10.1	107.7	8.4	86.2	7.9	92.1	6.2	98.0	4.4	84.5	11.1	101.3	0.7	105.4	7.3	78.6	5.7	82.1	8.4	95.1	12.2	109.3	7.3
Phorate	95.3	10.7	80.7	9.8	77.8	12.6	101.2	8.5	89.2	5.2	78.6	15.0	91.9	4.9	97.0	5.6	66.6	6.7	85.4	4.8	90.9	6.6	102.7	3.1
α-HCH	78.1	8.9	63.9	6.4	77.1	4.0	106.8	6.3	81.3	10.8	79.1	10.6	79.6	7.6	72.7	11.0	80.4	10.7	69.1	3.5	64.6	6.9	91.0	2.8
Simazine	100.7	13.3	94.2	11.7	83.9	11.3	83.7	12.2	72.7	12.0	92.1	6.7	78.8	11.0	93.3	13.1	100.6	7.8	92.1	1.4	110.4	7.7	98.9	11.5
β-HCH	81.9	8.9	84.7	5.7	84.4	5.0	94.2	9.1	86.9	4.8	66.8	8.6	77.7	2.9	102.0	7.9	76.8	2.5	64.1	4.2	81.0	12.5	80.1	0.7
Quintozene	71.5	9.1	99.5	10.3	78.4	3.0	95.7	2.3	82.6	11.7	70.1	12.2	72.2	8.2	74.2	6.4	91.7	9.4	74.1	17.2	88.2	9.4	93.8	1.5
γ-HCH	97.2	10.2	85.5	9.0	73.1	2.3	77.4	9.1	72.9	16.8	69.9	15.5	83.6	4.6	82.5	4.6	73.5	4.9	88.0	1.1	87.1	0.2	74.2	3.6
Terbufos	93.4	9.6	98.4	4.3	102.3	7.6	87.1	13.6	104.4	5.6	99.2	10.8	100.7	4.4	82.0	12.7	96.0	2.7	91.4	4.8	95.2	9.7	108.0	6.4
Diazinon	100.3	4.7	87.0	1.4	76.4	12.7	89.9	15.7	71.3	7.0	70.6	15.8	97.5	2.5	109.2	4.9	87.0	4.6	70.5	11.5	86.1	10.7	89.8	5.2
Pyrimethanil	90.8	18.3	88.8	8.0	77.5	13.6	113.2	7.6	102.2	15.6	69.4	9.2	79.3	7.7	76.2	8.6	75.9	16.3	108.6	3.1	101.5	15.9	113.9	7.3
Isazofos	95.5	5.8	88.2	5.6	72.3	9.4	89.5	6.5	81.8	14.4	92.9	13.9	89.0	5.0	110.4	5.2	86.2	3.6	71.6	12.7	102.4	1.7	94.9	3.7
Vinclozoline	89.0	11.2	109.1	10.5	84.1	8.6	104.2	3.9	99.5	14.6	86.8	16.4	77.0	11.7	99.6	3.6	91.9	6.4	87.3	6.1	80.8	5.1	102.4	10.3
Heptachlor	67.5	9.5	73.9	4.5	69.4	5.7	69.0	4.9	64.1	8.9	78.3	14.7	76.0	4.8	86.4	6.1	82.6	2.8	76.6	6.3	90.3	9.4	87.9	13.8
Metalaxyl	86.9	9.1	107.8	4.9	77.5	7.9	92.8	6.4	92.1	12.6	106.5	10.8	93.4	2.1	110.0	1.2	94.1	3.8	91.2	11.4	91.1	9.7	83.6	5.6
Chlorpyrifos	78.1	8.4	76.0	14.5	67.5	10.2	85.0	8.4	86.0	8.4	65.0	7.5	85.1	6.1	74.5	9.5	69.0	7.5	61.8	4.0	77.7	10.8	61.6	2.7
Aldrin	82.9	1.3	109.5	2.2	78.1	6.3	99.4	11.9	80.2	7.0	85.5	12.3	84.1	6.4	92.2	8.3	77.3	3.2	75.0	7.6	86.0	6.6	82.5	7.6
Parathion	79.4	4.0	78.6	6.8	61.8	2.2	96.0	2.6	101.8	6.8	68.9	6.1	93.4	3.3	96.1	4.7	65.6	1.9	71.6	5.2	71.5	4.2	64.3	3.9
Triadimefon	94.4	10.4	115.6	3.0	82.2	9.5	104.2	6.6	89.7	7.7	104.8	6.2	95.4	5.4	116.7	9.5	100.1	3.7	74.1	15.8	109.0	10.7	88.7	5.5
Dicofol	90.0	5.8	99.8	12.9	85.0	8.8	73.6	7.5	94.7	7.1	109.6	14.7	84.6	4.9	81.2	10.9	92.1	5.2	69.9	12.5	109.9	13.7	88.1	8.0
Isofenphos-methyl	70.2	5.9	106.3	5.2	71.2	8.6	89.0	10.4	71.2	13.4	91.3	4.9	88.8	3.6	109.0	7.1	98.1	2.2	83.3	3.6	105.3	6.8	93.3	11.8
Pendimethalin	92.2	1.5	92.2	3.4	63.6	4.2	99.6	5.1	103.2	5.2	71.4	9.2	93.4	3.0	93.4	5.3	73.0	9.6	97.9	6.6	86.7	9.1	108.1	13.2
Fipronil	78.8	7.1	79.9	9.8	63.8	8.0	83.8	6.7	67.7	8.8	66.8	11.0	92.3	6.2	99.5	10.7	72.5	4.2	79.1	3.3	73.3	9.8	68.1	11.6
Heptachlor epoxide-A	86.5	2.7	119.7	2.3	78.4	7.0	82.3	13.3	79.0	4.1	87.0	4.1	88.0	9.5	106.1	10.1	86.9	2.3	77.4	14.0	91.5	14.5	84.7	7.5
Heptachlor epoxide-B	83.8	8.7	81.1	9.4	70.5	9.5	82.0	5.2	88.3	5.8	85.2	7.3	86.6	2.0	92.2	8.6	91.6	8.3	71.6	17.9	71.2	8.0	76.7	8.2
Procymidone	89.1	3.7	95.8	11.7	70.2	8.2	101.4	14.2	72.8	7.2	107.4	11.9	98.7	8.5	108.6	6.4	106.0	6.6	101.8	7.8	96.7	9.5	95.5	7.0
γ-Chlordane	74.6	13.1	101.9	8.1	73.2	7.7	89.9	4.7	68.8	8.6	98.1	3.3	79.8	5.1	102.2	5.8	84.3	3.9	70.7	9.2	84.3	13.1	78.3	5.8
α-Chlordane	69.6	3.9	73.3	11.4	66.0	5.1	64.5	6.5	62.2	5.0	95.0	12.5	80.1	4.8	87.4	7.1	77.8	2.6	84.1	5.4	107.2	11.4	89.2	4.9
Endosulfan	70.1	14.2	85.6	8.8	70.7	6.9	95.2	4.2	78.4	14.9	109.7	4.4	82.8	6.8	89.2	6.1	95.1	4.9	64.9	3.9	76.7	10.9	94.7	2.9
Hexaconazole	103.4	7.1	109.2	9.0	89.9	6.0	100.7	8.7	81.9	4.6	110.5	2.8	105.5	4.0	102.9	1.3	112.7	3.1	107.8	3.9	101.7	6.5	95.5	6.5
DDE	100.6	4.4	93.3	6.8	99.6	8.0	65.6	7.5	64.8	5.5	73.6	10.4	99.5	8.1	104.2	8.5	98.9	4.1	82.8	11.6	111.6	11.2	102.5	4.7
Dieldrin	99.6	6.4	105.9	11.4	87.1	5.4	75.7	8.0	92.9	8.5	74.1	7.0	68.0	11.2	87.2	13.2	88.7	5.0	68.3	8.0	76.9	10.1	80.5	5.0
DDD	61.1	9.3	76.7	13.2	65.8	4.8	63.9	8.1	65.5	7.1	66.3	9.3	82.0	3.5	103.2	13.2	81.2	2.2	86.5	5.1	64.1	4.0	90.3	3.6
Flusilazole	91.0	4.9	107.5	6.4	78.2	6.2	102.6	12.0	76.4	7.9	73.0	13.8	101.8	7.7	75.2	10.2	109.4	3.6	67.6	17.8	99.5	7.7	93.0	5.0
Endrin	79.8	7.7	77.6	10.0	70.0	5.4	67.1	7.5	63.1	4.9	90.9	12.5	76.4	6.2	82.1	16.9	75.2	3.7	73.0	14.8	86.6	9.8	82.1	9.3
DDT	101.2	13.6	101.7	12.7	72.3	4.8	87.2	8.4	78.5	13.8	94.0	8.0	76.9	5.0	97.7	14.5	69.0	5.0	87.6	5.4	88.8	12.7	70.3	8.2
Triazophos	91.1	6.6	68.0	5.1	68.0	11.5	99.3	13.8	76.6	7.7	96.4	15.5	105.4	7.2	97.0	13.7	67.3	6.3	78.2	3.8	65.5	0.9	72.8	5.4
Benalaxyl	76.0	7.0	84.9	5.9	72.7	6.6	101.9	8.5	78.0	7.5	102.3	7.4	100.6	11.1	110.4	7.4	98.7	2.6	107.5	12.6	80.2	8.5	80.3	4.8
Tebuconazole	88.1	9.7	105.2	9.8	83.6	4.7	100.3	9.2	71.7	5.4	87.9	2.2	99.9	3.9	102.3	12.9	102.2	11.2	96.2	1.4	88.9	2.9	100.2	5.2
Piperonyl butoxide	111.6	7.5	106.5	10.8	67.5	9.2	88.1	8.6	72.6	9.6	93.7	3.3	101.6	5.1	85.1	3.8	93.3	10.2	84.5	5.7	84.2	9.4	89.1	4.4
Bifenthrin	109.7	5.1	106.8	16.0	73.1	8.2	89.9	4.9	91.4	19.3	107.0	3.2	99.8	4.5	75.6	18.9	93.2	5.9	100.6	7.8	88.3	4.4	85.6	6.3
Methoxychlor	65.0	2.6	105.5	12.5	70.8	12.1	69.5	4.6	77.0	2.1	78.1	12.5	71.5	8.3	82.1	7.0	65.3	7.7	66.1	5.0	68.2	4.5	105.8	15.8
Fenpropathrin	68.7	8.9	89.8	4.4	68.7	7.6	77.4	15.4	68.3	13.7	95.9	16.8	70.5	16.4	112.9	7.8	85.9	7.0	67.0	15.4	83.1	2.0	73.8	10.8
Permethrin	64.2	0.3	68.7	5.9	64.4	7.4	79.6	3.9	84.9	10.5	70.7	9.0	89.2	11.1	76.6	5.8	62.9	3.7	72.9	1.9	90.2	4.4	81.8	7.8
Pyridaben	83.8	2.0	91.5	9.2	63.2	6.1	97.3	6.9	92.4	12.5	74.1	15.5	83.4	8.5	82.6	15.1	68.9	7.9	95.5	8.7	79.0	13.5	85.5	9.7
Etofenprox	67.1	4.0	62.6	5.3	66.2	9.9	86.9	5.9	79.0	19.7	70.3	15.2	101.9	8.3	74.9	12.0	71.4	6.8	88.2	19.6	101.1	18.9	111.4	9.5

(*continued on next page*)

Table 2 (*continued*)

	Beef						Salmon						Pork						Egg					
	Spiked level (ng g^{-1})																							
	20		50		200		20		50		200		20		50		200		20		50		200	
	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)	R%	RSD (%)
cis-Difenoconazole	70.6	2.0	65.7	2.2	60.5	6.9	81.6	11.1	85.4	9.6	69.2	8.8	65.1	4.9	72.3	11.2	70.3	8.2	68.2	1.2	79.9	13.4	70.1	7.7
trans-Difenoconazole	70.4	1.9	66.1	4.8	72.3	5.6	80.5	11.8	91.4	9.6	71.6	8.9	81.5	10.9	72.5	16.5	67.4	7.1	77.1	5.7	88.4	10.9	83.2	6.8

*R% represents recovery.

techniques was demonstrated in Fig. S6, highlighting further the synergistic clean-up effect of GLME-MA-*d*-SPE technique than using GLME or d-SPE alone.

3.4. Matrix effects

Matrix effect (ME%) is an important consideration in multi-residue pesticide analysis as the accuracy of instrumental detection can be influenced by the co-eluted matrix interferences. A positive value of ME represents matrix-induced signal enhancement, while negative value indicates signal suppression. The degree of ME can be generally categorized into soft matrix effects (ME% of approximately ±20%), medium matrix effects ($-50\% < ME\% < -20\%$ and $50\% > ME\% > 20\%$) and strong matrix effects ($ME\% < -50\%$ and $ME\% > 50\%$). Fig. 6 showed the representative analytes with distinctive ME%, and the overall ME% of analytes in fatty foods were categorized accordingly in Fig. S4.

The assessment on matrix effects of 50 pesticides in tested foods of animal origin was indicated in Table 1 and Fig. S5. Among the 50 pesticides being analyzed, more than 50% of the analytes showed soft matrix effect, followed by approximately 40% pesticides with medium matrix effect and less than 10% pesticides showed strong matrix effect (except for pork samples with 16%) (Fig. S5). In comparison of ME% between foods of animal origin after treated by the sequential GLME-MA-*d*-SPE clean-up technique, about 84% of the analytes in salmon sample exhibited soft to slight medium (<35 ME%), followed by beef, egg and pork samples with 80%, 66% and 64%, respectively. As shown in Fig. S4, 72% of the pesticides being analyzed were in the form of ion suppression. This result is in agreement with previous study that observed ion suppression occurred for majority of the pesticides and veterinary drug in meat matrices (Gómez-Pérez et al., 2014). In order to compensate for ME% while determining pesticide residues in fatty foods, matrix-matched calibration standards were used for quantification.

3.5. Method validation

Under optimized condition, the analytical proposed in this study was validated through evaluating the linearity, accuracy and precision. Good linearity with the square of the correlation coefficient ($r^2 > 0.99$) was obtained for all the analyzed compounds in both the solvent and matrix-matched standards. The final method was validated through three different spiking levels (20, 50 and 200 ng g^{-1}) in triplicate in each of all four matrices. The recoveries of both legacy and emerging pesticides in different spiked levels ranged from 60.5% to 119.7%, and the RSDs for all target compounds in different fatty foods were less than 20%, indicating good precision (Table 2). The results showed that the overall MLODs (MLOQs in parentheses) of multi-class pesticides ranged from 0.1 to 11.0 (0.3–36.7) ng g^{-1} in these fatty foods. A comparison of the analytical performances between this method and methods from other literatures was shown in Table 3, signifying the advantageous of the sequential GLME-MA-*d*-SPE clean-up approach.

3.6. Analysis of multi-class pesticides in fatty food samples

To evaluate the applicability of the GLME-MA-*d*-SPE method, it was employed for investigating the occurrence of multi-residue pesticides in foods of animal origin (salmon, pork, beef and egg) from a local market. Analytical results indicated that methoxychlor was detected in egg but it showed concentration below MLOQ, while most pesticides showed concentrations of < MLOD in fatty food samples in this study. Permethrin was the only pesticide detected in salmon, pork and egg at concentration of 27.02, 11.18 and 20.19 ng g^{-1}, respectively. However, the detection concentrations of permethrin were still within the MRLs regulated by European Commission (50 mg kg^{-1}) (European Commission, 2008), U.S. Code of Federal Regulation (0.05 parts per million) (U.S. Code, 2020) and national standard GB 2763-2016 in China (50 mg

Table 3
Comparison of the performances between different methods for multiresidue pesticide analysis in foods of animal origin.

Fatty food matrix	Number of analyzed pesticides	Pesticide family	Extraction	Clean-up	MLOQ (ng g^{-1})	ME% range	References
Salmon, pork, egg, beef	50	OC, OP, PY, CH, TH, AF, PF, DD, MF, CF, DA, PZ, PS, UN	UAE	GLME- MA-dSPE	0.3–36.7	−78 and 43	This study
Pork, beef, chicken, fish	109	CB, OD, OP, PP, PY, AF, OC, DC, UN, PS, BD, TZ, CA, CF, AN, CS, PR, NP, PC	ASE	GPC	0.3–107.7	N/A	Wu et al. (2011)
Salmon, pork, avodaco, kale	65	OC, OP, CT, PY, CB, AF, PF, BD, DA, PP, CF, SF, PS, SE, HI, UN	SLE	EMR-Lipid	5–10[a]	−50 and 120[b]	Han et al. (2016)
Fish liver, crab hepatopancreas	7	OP, CT, UN, PP, CF	Vortex-assisted MSPD		50–1250	N/A	Souza Caldas et al. (2013)
Fish	23	OP, PY, BD, CS, AN, PX, PP, SF, HI, DA, DC, UN	QuEChERS		15–50	−25 and 140	Colazzo et al. (2019)
Egg	9	OC, PY, CS, GI, DC	QuEChERS		3–8	−80.6 and 98.9	Song et al. (2019)
Fish	28	DA, OP, DC, CA, PP, CS, SF, AN, BD, HI, PX, PY	QuEChERS		10.5–85.1	−25 and 324	Ernst et al. (2018)

OC: Organochlorine insecticide; OP: Organophosphate insecticide; PY: Pyrethoid insecticide; GI: Growth inhibitor; CT: Chlorotriazine herbicide; AF: Aromatic fungicide; PF: Pyrimidine fungicide; DC: Dicarboximide fungicide; AN: Anilide fungicide; CF: Conazole fungicide; BD: Bridged diphenyl acaricide; DA: Dinitroaniline herbicide; PP: Phenylpyrazole acaricide; PS: Pesticide synergist; PX: Phenoxypropionic herbicide; SF: Strobilurin fungicide; HI: Hormone insecticide; CB: Carbamate insecticide; SE: Sulfite ester acaricide; CA: Chloroacetanilide herbicide; OD: Oxadiazine insecticide; TZ: Thiazolidine acaricide; PR: Pyrrole insecticide; NP: Nitrophenyl ether herbicide; PC: Pyrazolecarboxamide acaricides; UN: Unclassified.
[a] LOD value.
[b] ME% range for salmon matrix.

kg^{-1}) (GB 2763-2016, 2016). Since the application of pyrethroid pesticides greatly increased to replace the banned legacy pesticides and organophosphate insecticides with restricted usage, they have been commonly detected in biota samples. A study found that the detection frequency of pyrethroids in farmed salmon was 100% due to the function of pyrethroid pesticides as anti-sea lice pesticide formulations (Aznar-Alemany et al., 2017). Two-third of the six commercial farms successfully detected permethrin with the concentration as high as 98.3 ng g^{-1} in chicken eggs, but found none in home-produced eggs (Parente et al., 2017). The results from this study plus recent researches highlight an observation that increased application of pyrethroids in the animal farming industry cause the high anthropogenic occurrence of pyrethroids in biota, human and the environment. Therefore, pesticide residues in foods should be frequently monitored henceforth, especially in lipid-rich tissues where toxic pollutants bioaccumulate.

4. Conclusion

The GLME-MA-*d*-SPE technique sufficiently eliminated co-extracted lipid interferences and successfully achieved multi-residue pesticide analysis in fatty foods of animal origin. One of the advantages of such effective clean-up technique is to protect the detection instruments from being damaged, hence prolonging the lifespan of the expensive instruments and saving cost by minimizing the frequency of maintenance work. In order to tackle the challenges of analytical chemistry and food safety concerning routine simultaneous analysis of various pollutants (pesticides, plasticizers, flame retardants, pharmaceuticals, etc.), this method can be further enhanced to realize the high-throughput, broad-spectrum, on-line and automation potentials for screening different kinds of contaminants in the environment, especially in identifying trace-level of chemicals in complex food and biological matrices.

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CRediT authorship contribution statement

Han Yeong Kaw: Project administration, Methodology, Writing - original draft, Visualization, Formal analysis. **Xiangzi Jin:** Investigation, Validation. **Yunan Liu:** Investigation. **Long Cai:** Investigation. **Xiangai Zhao:** Resources. **Juan Wang:** Resources. **Miao He:** Resources, Supervision. **Donghao Li:** Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.lwt.2020.110448.

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