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Improvement of a GC–MS analytical method for the simultaneous detection of 3-MCPD and 1,3-DCP in food

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Abstract Chloropropanols such as 3-monochloropropane-1,2-diol (3-MCPD) and 1,3-dichloropropan-2-ol (1,3-DCP) are produced by heat treatment in the presence of fat and hydrochloric acid during the manufacture of food stuffs such as hydrolyzed vegetable protein and soy sauce. 3-MCPD and 1,3-DCP have been detected in several foods. An efficient, highly selective GC–MS method was developed to determine the concentration of 3-MCPD and 1,3-DCP in food. Calibration curves for 3-MCPD and 1,3-DCP were constructed, and a correlation of determination (r^2) $\geq$ 0.9990 was obtained. The limits of detection and quantitation for 3-MCPD in food were 0.6 and 2.0 µg/kg, respectively, and those for 1,3-DCP were 0.2 and 0.6 µg/kg, respectively. To the best of our knowledge, this GC–MS-based method is a newly improved analytical procedure for the simultaneous separation and determination of 3-MCPD and 1,3-DCP, at once and at low levels (µg/kg).

Keywords Chloropropanol · 1,3-Dichloropropan-2-ol · 3-Monochloropropane-1,2-diol · Gas chromatography–mass spectrometry · Hydrolyzed vegetable protein (HVP)

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Introduction

3-Monochloropropane-1,2-diol (3-MCPD), the most common member of the group of chemical contaminants known as chloropropanols, and 1,3-dichloropropan-2-ol (1,3-DCP) are carcinogenic substances produced by the oxidation of hydrolyzed vegetable protein (HVP) [1]. During the hydrochloric acid hydrolysis of proteins containing low amounts of oil (e.g., beans, wheat, and rice), the proteins break down into amino acids while fat residues degrade into fatty acid esters (glycerol). The resulting glycerol reacts with hydrochloric acid, eliminating the fatty acid and producing chlorinated contaminants [2, 3]. Recent studies have reported that soy sauce contains the highest concentrations of MCPD and 1,3-DCP. These are also found in other foods such as bread, cookies, soups, and sauces [4]. In addition, 3-MCPD is an environmental hormone that is suspected to disrupt the endocrine system and possibly induce genotoxicity, reproductive toxicity, neurotoxicity, renal toxicity, and cancer [5, 6]. 1,3-DCP is highly volatile and flammable and can cause subcutaneous hemolysis on inhalation, sterility and other reproductive problems, acute liver damage, blood coagulation, cancer, and damage to the eyes and skin on contact [7–9]. Several countries have set reference values for the dietary intake of 3-MCPD and 1,3-DCP and strictly evaluate their content in food. In HVP, the maximum concentration of 3-MCPD has been set at 0.02 mg/kg. In soy sauces, 3-MCPD concentrations have been limited to 0.02 mg/kg for products with 40% solids and 0.05 mg/kg for 100% dried products, and these set limits are proportional to the concentration of solid foods for a solid accounting for 40–100% [10–12]. On May 22, 2002, the South Korean government provisionally set the maximum concentrations of 3-MCPD at 0.3 mg/kg for unfermented and mixed soy sauces and

1.0 mg/kg for HVP [13–15]. Therefore, it is important to develop a method for detecting and quantifying 3-MCPD and 1,3-DCP to manage the regulation of food constituents. 3-MCPD and 1,3-DCP are poorly volatile and have a high polarity, resulting in very low sensitivity because of their poor recovery and tendency to adsorb on the GC inlet or column [16]. Consequently, they must be analyzed following derivatization by methods including N,O-bis(trifluoroacetamide acid) (BSTFA), phenylboric acid (PBA), heptafluorobutyrylimidazole (HFBI), and heptafluorobutyric acid (HFBA). In this study, HFBA was used to derivatize 3-MCPD and 1,3-DCP in accordance with the Korea Food Code. In addition, there is no set limit for the concentration of 1,3-DCP in foods. Hence, this study aims to develop an effective method to simultaneously analyze 3-MCPD and 1,3-DCP, which are abundant in soups and soy sauces available across South Korea, Japan, Southeast Asia, and China. The development of such an effective analytical method will provide standard analytical techniques to prevent and regulate the amount of harmful 3-MCPD and 1,3-DCP.

Materials and methods

Analyzed samples

Food products with a high probability of detecting 3-MCPD and 1,3-DCP were primarily selected. Soybean paste, cookies, and dietary oils were purchased from three supermarkets in Sejong, Deajeon, and Cheongju, and the contents of 3-MCPD and 1,3-DCP were simultaneously analyzed. The purchased samples were stored under conditions identical to those in the supermarket until analysis.

Chemicals and reagents

3-MCPD and 1,3-DCP were purchased from Sigma-Aldrich (St Louis, USA), and 3-MCPD-d5 and 1,3-DCP-d5 internal standards were purchased from CDN Isotopes (Pointe-Claire, Quebec, Canada). The extractant dichloromethane was purchased from Wako (Osaka, Japan), and sodium chloride and sodium sulfate were purchased from Dae Jung (Gyeonggi-do, Korea). The Extrelut-NT3 cartridge, ethyl acetate, and isooctane were purchased from Merck (Darmstadt, Germany), and heptafluorobutyric anhydride (HFBA) used for derivatization was purchased from Sigma-Aldrich (St. Louis, USA). Triple-distilled water used in these experiments was obtained using a Thermo Scientific Barnstead Nanopure system (Marietta, USA).

Preparation of standard solutions and reagents

To prepare 1000 μg/mL solutions, each stock and internal standard solutions was dissolved in ethyl acetate. The working standard solution, which was used to generate the calibration curve, was prepared by diluting the stock standard solution in ethyl acetate to 0.03, 0.1, 0.5, 1, and 2 μg/mL solutions. The internal standard solution was diluted with ethyl acetate to give a 10 μg/mL solution. To perform derivatization, 50 μL of each standard solution at 5 different concentrations, 50 μL of the internal standard solution, 900 μL of isooctane, and 150 μL of HFBA were mixed in vitro in a test tube with a lid, and the results were analyzed. In this study, the samples were classified based on their fat contents (< 10, 10–50, and 70–100%), and only the main samples were selected and analyzed. The stock standard and internal standard solutions were stored in brown bottles at 4 °C until analysis. In addition, a 5 M sodium chloride solution was prepared by dissolving 292.2 g of NaCl in 1 L of triple-distilled water.

Sample preparation

First, 5 g of a homogenized sample was accurately measured, and a 5 M NaCl solution was added up to the 30-mL mark. Second, the mixture was subjected to shaking for 10 min, followed by centrifugation (4500 rpm, 4 °C, 5 min). Third, 1 g of anhydrous sodium sulfate was added on the top of the Extrelut-NT3 column, and 2 mL of the supernatant from the centrifuged sample, as well as 50 μL of the internal standard solution, was added. The resulting mixture was allowed to rest for 10 min to enable adsorption, followed by elution with 60 mL of dichloromethane at a rate of 2 mL/min. The eluted fractions were collected in an evaporating flask, followed by evaporation in a water bath at 35 °C and concentration to 1 mL under high pressure. The concentrated solution was transferred into a graduated test tube using a Pasteur pipette. The residues remaining in the flask were washed with dichloromethane and added to the test tube. The test tube was then subjected to evaporation in a water bath at 35 °C under nitrogen and concentrated to 100 μL. Next, 900 μL of isooctane and 150 μL of HFBA were added to the concentrated sample in the test tube, and the mixture was allowed to react for 30 min in a water bath at 60 °C after sealing the test tube with a lid. Next, the sample was allowed to rest for 10 min at room temperature, 5 mL of triple-distilled water was added, and the mixture was shaken to cease the reaction and separate the HFBA residues. The top organic layer was transferred to a test tube, a small amount of anhydrous sodium sulfate was added, and the layer was subjected to shaking. The mixture was allowed to rest, and the separated layer was extracted into a vial and analyzed by GC–MS.

Fig. 1 Chromatograms of
(**A**) 1,3-DCP, 1,3-DCP-d5,
3-MCPD, and 3-MCPD-d5 in
standard mixtures and
(**B**) HFBA derivatized
chloropropanols in soy sauce
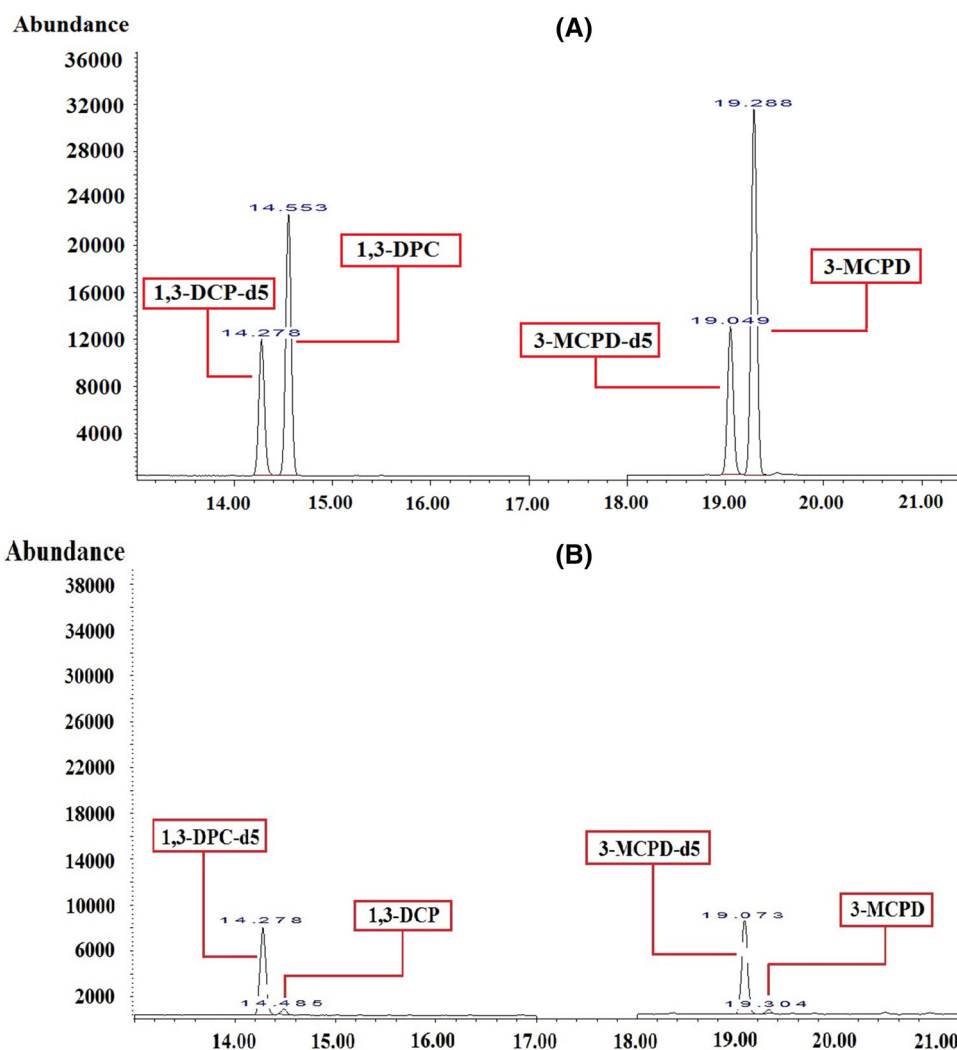
GC–MS analysis

GC and MSD instruments with model numbers 7890B and
5977A (Agilent, USA), respectively, and an Agilent DB-
5MS column (30 m × 0.25 mm, 0.25 μm) were used for
analysis. The injector temperature was 250 °C and 1 μL of
the sample was injected into the GC–MS column with He
at a flow rate of 0.8 mL/min. The analysis was conducted
for 40 min in the split mode (10:1). The oven temperature
was set to 50 °C for the initial 5 min, increased at a rate of
2 °C/min until the temperature reached 90 °C, maintained
at 90 °C for 5 min, and increased at a rate of 30 °C/min
until the temperature reached 280 °C, which was main-
tained for the final 3 min. The ion source temperature was
230 °C and the analysis was conducted in the single-ion
monitoring (SIM) mode with an ionization energy of 70 eV
obtained in the electron ionization (EI) mode. In the
derivatized samples, ions were observed at m/z 253, 275,

289, 291, and 453, corresponding to 3-MCPD, whereas
those observed at m/z 257, 278, 294, and 456 corresponded
to 3-MCPD-d5 (internal standard solution). Peaks at m/z
253 and 257 corresponded to the quantitative ions of
3-MCPD and the internal standard solution, respectively.
Peaks observed at m/z 275, 110, and 277 corresponded to
1,3-DCP, whereas those at m/z 278 and 116 corresponded
to 1,3-DCP-d5 (internal standard solution). Peaks at m/z
275 and 278 corresponded to the quantitative ions for 1,3-
DCP and the internal standard solution, respectively. For
qualitative analysis, the reference range of ion proportion
was set to ± 20% (Figs. 1, 2, 3).

Verification of the effectiveness of the test method

In this study, the selectivity, linearity, limit of detection
(LOD), limit of quantification (LOQ), precision, and
accuracy were investigated. Standard solutions of 3-MCPD

Fig. 2 EI mass spectra of 1,3-DCP and 1,3-DCP-d5
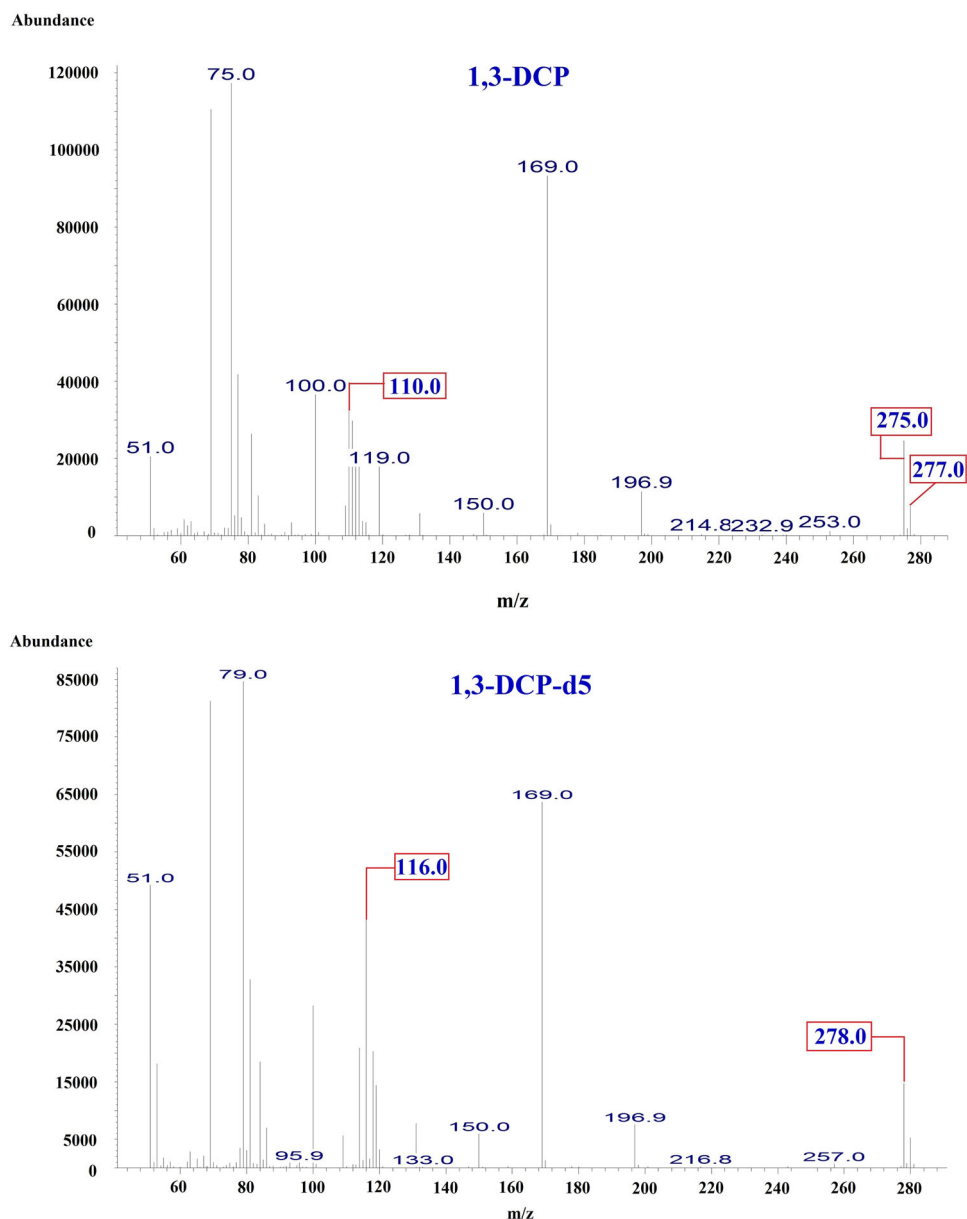

and 1,3-DCP were added to samples (soy sauce (< 10%), snack (10–50%), and rapeseed oil (> 50%)) with different fat contents to verify the selectivity of the method. Linearity was verified using the coefficient of determination (r^2) and regression, which was obtained from the calibration curve plotted by the analysis of standard solutions with different concentrations. The LOD and LOQ were determined by measuring a matrix blank of fortified samples seven times with standard solutions. The background response and standard deviation were computed and defined as 3 times (3σ) and 10 times (10σ) the standard deviation (σ), respectively. Accuracy was determined by the recovery rate measured by mixing 4.5, 7.5, and 15 μg/mL of a standard solution with the sample, which was measured seven times in 1 day. Precision was verified by the percentage relative standard deviation (%RSD) obtained by conducting seven intraday assays in 1 day. Accuracy and precision were verified to be suitable on the basis of AOAC guidelines [17].

Statistical analysis

The data analysis function of EXCEL (Microsoft Office 2010) was used to calculate the mean and standard deviation. An F-test was conducted with a significance level of 99% ($p = 0.01$).

Fig. 3 EI mass spectra of 3-MCPD and 3-MCPD-d5
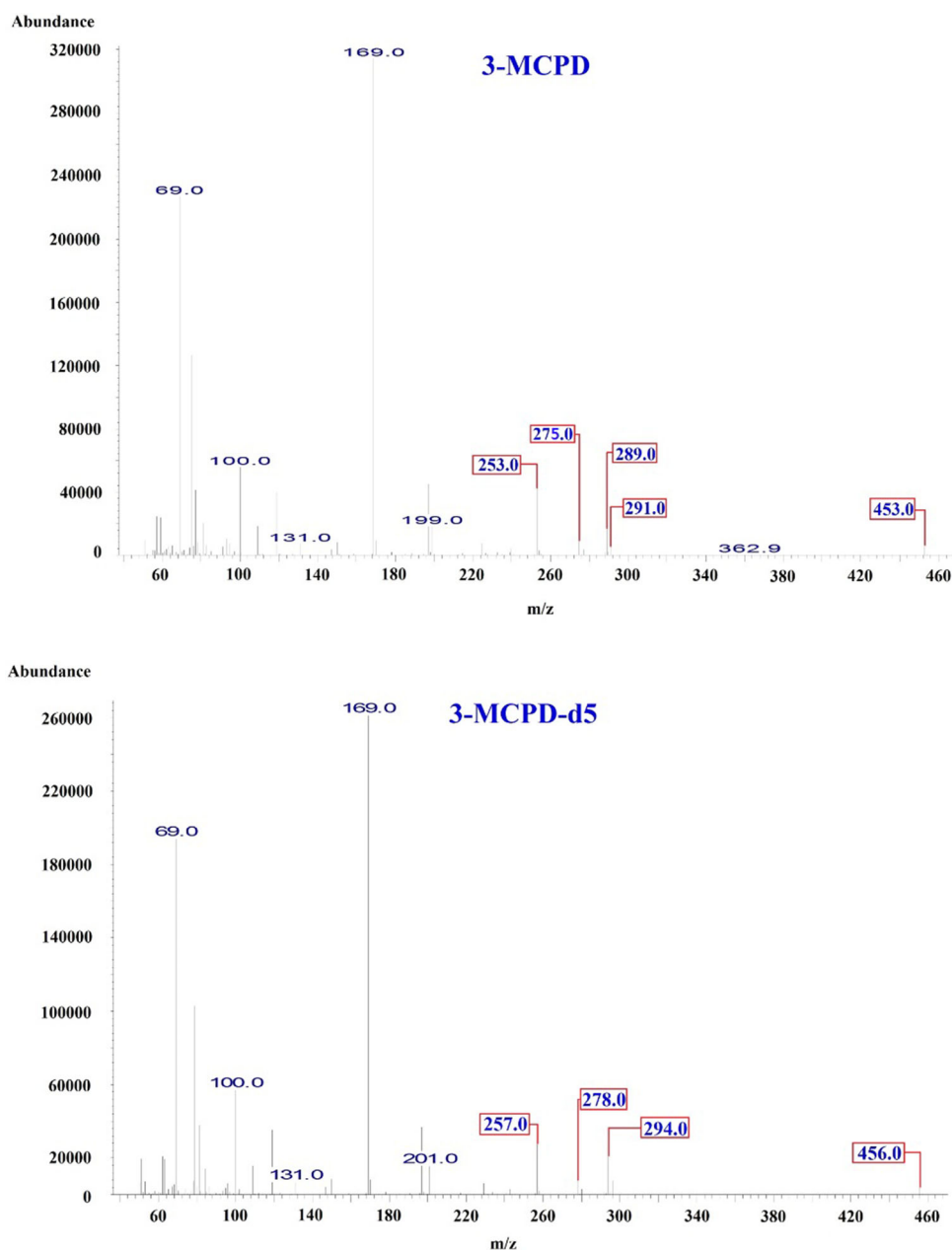
Results and discussion

Verification of the effectiveness of the test method

Generally, hydrolysis is carried out at 100–130 °C using 4–6 M hydrochloric acid for 4–24 h and finally neutralized with NaOH. In this process, 3-MCPD and 1,3-DCP are produced by replacing one or two of the three fatty acids of the triglyceride of glycerol with the chlorine group of hydrochloric acid [18]. 3-MCPD and 1,3-DCP reacted selectively with the isotopically labeled internal standard and chloropropanol to provide stable derivatives of HFBA because of their low volatility and high polarity. These

species decompose into characteristic ions and allow quantification of chloropropanol [19]. Although studies have reported the GC–MS [20] or LC–MS [16] analysis of 3-MCPD with 1,3-DCP using chromatography glass columns [19], to the best of our knowledge, studies have not reported the simultaneous analysis of 3-MCPD and 1,3-DCP by GC–MS using Extrelut-NT3 columns in the past decade. Hence, this study verified the effectiveness of a simple, rapid GC–MS method for the simultaneous analysis of 3-MCPD and 1,3-DCP (Table 1).

Standard solutions were added to the samples prior to GC–MS analysis to verify the selectivity of 1,3-DCP and 3-MCPD. Sample analysis indicated a peak retention time

Table 1 Characteristic ions in the EI spectra of HFBA derivatives

Compound	MW	[M-CH$_2$Cl]$^+$	[M-C$_3$F$_7$CO$_2$]$^+$	[M-C$_3$F$_7$CO$_2$CH$_2$]$^+$	[M-C$_3$F$_7$CO$_2$-HCl]$^+$
3-MCPD	502	453	289/291	275	253
1,3-DCP	325	275/277	110	–	–
3-MCPD-d5	507	456	294	278	257
1,3-DCP-d5	330	278	116	–	–

Table 2 Limits of detection (LOD), limits of quantification (LOQ), accuracy, and precision for 3-MCPD and 1,3-DCP in foods

Sample	Compound	Spiked concentration (µg/mL)	LOD (ng/g)	LOQ (ng/g)	Accuracy (% recovery) (n = 5, a = 0.01)	Precision (%RSD) Intra-day
Soy sauce	3-MCPD	4.5	0.6	2.0	90 ± 0.002[a]	0.5
	1.3-DCP		0.2	0.6	80 ± 0.003	1.6
Snack	3-MCPD	4.5	0.05	0.2	85 ± 0.002	0.8
	1.3-DCP		0.03	0.1	79 ± 0.002	2.0
Rapeseed oil	3-MCPD	4.5	0.2	0.6	88 ± 0.009	3.7
	1.3-DCP		0.2	0.5	75 ± 0.001	0.8

[a]Values are mean ± confidence interval

(R$_t$) and a spectrum identical to that observed for the standard solution and a specific peak with no noise. Standard solutions of 3-MCPD and 1,3-DCP were diluted to 0.3–2 µg/mL, followed by derivatization and GC–MS analysis. The analysis revealed that the regression equations for 3-MCPD and 1,3-DCP were $y = 1.767827x + 0.001,670$ and $y = 1.264366x + 0.016974$, respectively, while r^2 was greater than 0.9990 for 3-MCPD and greater than 0.9999 for 1,3-DCP, indicating good linearity. The LOD and LOQ for 3-MCPD were 0.6 and 2.0 µg/kg, respectively. The corresponding LOD and LOQ values for 1,3-DCP were 0.2 and 0.6 µg/kg (Table 2). In other countries, 3-MCPD had LOD and LOQ values of 0.1–10 and 1.2–50 µg/kg, respectively, and the LOD and LOQ of 1,3-DCP ranged between 0.41–3.15 and 2–150 µg/kg, respectively. Results similar to those in this experiment have been obtained previously [16, 19–27]. The recovery of this study was 88–90% for 3-MCPD and 75–80% for 1,3-DCP. Previous studies have shown that the recovery of soy is lower than that of 3-MCPD (61–131%) and 1,3-DCP (71–120%) [16, 19–27]. However, this study is suitable for 75–120% of the AOAC guidelines [17]. The lower recovery of 1,3-DCP than that of 3-MCPD is probably related to how the samples were concentrated [28]; 1,3-DCP should not be completely dried in the concentration process. Experiments were carried out at three concentrations: 4.5 ppm (final concentration 30 ppb), 7.5 ppm (final concentration 50 ppb), and 15 ppm (final concentration 100 ppb). Only the lowest possible concentration of 4.5 ppm is shown (Table 2). Seven intraday assays were conducted in 1 day to verify their precision, and a standard deviation of 5% was observed, satisfying the less than 8% requirement specified by the AOAC guidelines [17]. When the significance of the intraday precision was verified with ANOVA, no significant differences were observed ($p < 0.01$).

Application of the developed GC–MS method to food containing different fat contents

Food products on sale in South Korea were classified based on their fat contents (< 10, 10–50, and 70–100%), and the main samples were selected. Soy sauce was not selected as a monitoring sample because many different sample types of soy sauce, which is widely consumed in Korea, were already used as monitoring samples. Forty-five samples of food products were analyzed (i.e., soybean paste, cookies, and dietary fats). The analysis revealed the absence of 3-MCPD and 1,3-DCP in all samples. The recovery rates for 3-MCPD and 1,3-DCP for soybean paste were 91 and 76%, respectively. The corresponding recovery rates were 91 and 80% for cookies and 76 and 88% for dietary fats. High recovery rates indicated that the GC–MS method for the simultaneous determination of 3-MCPD and 1,3-DCP is reliable (Table 3).

In the present study, 3-MCPD and 1,3-DCP were not detected in food, but the results of previous studies showed that 3-MCPD was detected in 8 of 44 soy sauces (0.02–0.28 mg/kg), 20 of 28 snacks (0.09–1.43 mg/kg), and 8 of 10 oils (0.04–1.22 mg/kg) [3]. In addition, studies thus far have reported that 3-MCPD and 1,3-DCP are produced by mixing and cooking with oil, NaCl, and water. 3-MCPD and 1,3-DCP have been reportedly found in sauces, processed meat products [29], preheated grain products, e.g., cereal, bread, donuts) [30], and compounds used as a seasoning in food products (soups) [31]. Hence, it is necessary to reduce the production of harmful

Table 3 Detection of 3-MCPD and 1,3-DCP in foods

Food categories	Compound	Sample (N)	Detected (N)	Mean (µg/kg)	Recovery (%)
Doenjang (fat content < 10%)	3-MCPD	15	0	ND*	91
	1,3-DCP			ND	76
Snack (fat content 10–50%)	3-MCPD	15	0	ND	91
	1,3-DCP			ND	80
Rapeseed oil (fat content > 50%)	3-MCPD	15	0	ND	76
	1,3-DCP			ND	88

*ND: Below LOD (not detected)

substances. In addition, continuous monitoring and evaluation of different food products are crucial to ensure public health and food safety.

In conclusion, a rapid GC–MS method to simultaneously determine 3-MCPD and 1,3-DCP, typically found in food products such as soup, soy sauce, and HVP, is developed. 3-MCPD and 1,3-DCP are consumed in large quantities not only in South Korea but also in several other Asian countries, which is a social problem with global ramifications. Methods for effectively analyzing both 3-MCPD and 1,3-DCP are being developed in South Korea. The concentrations of 3-MCPD and 1,3-DCP were simultaneously analyzed to determine the content of these harmful compounds in different purchased food products in South Korea. The effectiveness of the developed GC–MS test method is verified by the calculation of selectivity, linearity, the limit of detection (LOD), the limit of quantification (LOQ), precision, and accuracy. Application of the developed method to representative food samples indicated that 3-MCPD and 1,3-DCP were absent in all 45 samples. Hence, the developed GC–MS method is accurate and effective. The results obtained herein can serve as a reference for food safety regulations.

Compliance with ethical standards

Conflict of interest All authors declare no conflict of interest.

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