



Determination of organophosphate flame retardants in meat products from South Korea by QuEChERS-LC-MS/MS

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ABSTRACT

Organophosphate flame retardants (OPFRs) are extensively applied as both flame retardants and plasticizers, however, they have been detected in various matrices, including environmental and food samples. The exposure of humans to OPFRs can cause neurotoxicity, endocrine disruption, and reproductive toxicity. Since food ingestion is the primary exposure pathway, monitoring OPFR levels in food is critical. This study developed and validated a method for analyzing 11 OPFRs in meat products using QuEChERS extraction with liquid chromatography-mass spectrometry. The developed method was validated in terms of linearity, accuracy, precision, and limit of detection (LOD) and quantification (LOQ) according to the CODEX guidelines, exhibiting an accuracy in the range of 53.4 ~ 114.3 % and an LOD between 0.08 ~ 0.62 ng/g. Application to meat samples from South Korean markets revealed six OPFRs above the LOQ, with tris(1-chloropropan-2-yl) phosphate being the most abundant (1.67 ng/g). Duck meat showed the highest total OPFRs concentration (11.27 ng/g). The estimated daily intake (EDI) was calculated based on the monitoring results and was compared with the reference dose (RfD). Collectively, EDI was much lower than RfD; therefore, the risk of OPFRs exposure through meat consumption was low.

1. Introduction

Globally, the demand for meat products has recently increased as a result of population growth and change in dietary preferences to pursue meat products as major sources of nutrition (Henchion et al., 2021). However, the widespread consumption of meat products has raised concerns about their quality and safety, with their safety facing challenges due to several organic contaminants. Such contaminants include residues from veterinary drugs, agrochemicals, mycotoxins, food additives, and environmental contaminants, which have attracted considerable interest worldwide (Hoogenboom et al., 2021; Makowska et al., 2022; Pleadin et al., 2021; Wang et al., 2022). Environmental pollution has rapidly accelerated as technical advances have made mass production possible (Yadav et al., 2021). Consequently, various types of environmental contaminants are detected in everyday environments. Among these contaminants, persistent organic pollutants (POPs) pose a high risk due to their persistence in the environment and their anthropogenic

activity (Jones, 2021). POPs include pesticides, polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), brominated flame retardants (BFRs), and perfluorinated and polyfluorinated alkyl substances (PFAS) (Haedrich et al., 2020), some of which have been banned by the Stockholm Convention. As a result, a movement to stop their use has emerged and novel substances have been introduced to replace these banned compounds, with their production gradually increasing. However, some of these substances have been found to accumulate in the environment, exhibiting a similar behavior to that of the banned compounds, thus being toxic (Qadeer et al., 2024). Organophosphate flame retardants (OPFRs) are representative substances exhibiting this trend, being introduced into the market as a replacement for BFRs that were widely used before being banned.

OPFRs are expected to exhibit lower persistence in the environment than BFRs owing to their physicochemical properties (Zhang et al., 2016). Although OPFRs have a relatively high vapor pressure owing to their short half-life, they are expected to have a shorter long-range

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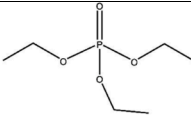
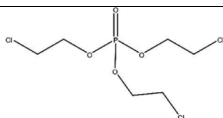
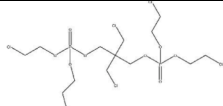
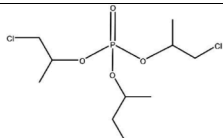
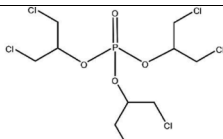
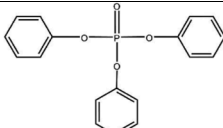
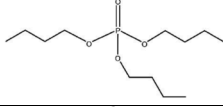
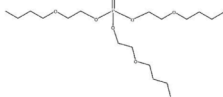
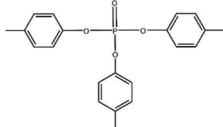
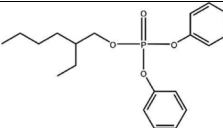
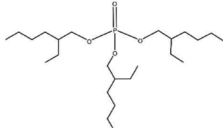
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transport through the atmosphere compared to BFRs (Rodgers et al., 2018). However, compared to BFRs, OPFRs have a higher water solubility and persistence in water, resulting in long-range transport via waterborne routes (Ma et al., 2017; Sühring et al., 2016). Furthermore, previous studies have reported the detection of several OPFRs in air and water samples at higher levels than BFRs (Blum et al., 2019). In the

Great Lakes, OPFRs and polybrominated diphenyl ethers (PBDE) were detected in level ranging between 10–100 ng/L, and 0.05–0.25 ng/L, respectively (Melymuk et al., 2014). OPFRs in the air can enter surface water through rain and transport and persist in aquatic environments due to their high solubility in water (Kim and Kannan, 2018). This leads to their accumulation in the environment thus increasing the

Table 1
Physicochemical properties of OPFRs.

Name	Abbreviation	CAS#	Structure	Formula	Molecular weight (g/mol)	Log Kow
Triethyl phosphate	TEP	78-40-0		C ₆ H ₁₅ O ₄ P	182.2	0.80
Tris(2-chloroethyl) phosphate	TCEP	115-96-8		C ₆ H ₁₂ Cl ₃ O ₄ P	285.5	1.78
2,2-Bis(chloromethyl)trimethylene bis(bis(2-chloroethyl)phosphate)	V6	38051-10-4		C ₁₃ H ₂₄ Cl ₆ O ₈ P ₂	583.0	2.83
Tris(2-chloroisopropyl) phosphate	TCIPP	13674-84-5		C ₉ H ₁₈ Cl ₃ O ₄ P	327.6	2.59
Tris(2-chloro-1-(chloromethyl)ethyl) phosphate	TDCIPP	13674-87-8		C ₉ H ₁₅ Cl ₆ O ₄ P	430.9	3.65
Triphenyl phosphate	TPhP	115-86-6		C ₁₈ H ₁₅ O ₄ P	326.3	4.59
Tributyl phosphate	TBP	126-73-8		C ₁₂ H ₂₇ O ₄ P	266.3	4.00
Tris(2-butoxyethyl) phosphate	TBOEP	78-51-3		C ₁₈ H ₃₉ O ₇ P	398.5	3.75
Tri-p-cresyl phosphate	TPCP	78-32-0		C ₂₁ H ₂₁ O ₄ P	368.4	5.11
2-Ethylhexyl diphenylphosphate	EHDPP	1241-94-7		C ₂₀ H ₂₇ O ₄ P	362.4	5.73
Tris(2-ethylhexyl) phosphate	TEHP	78-42-2		C ₂₄ H ₅₁ O ₄ P	434.6	9.49

possibilities for human exposure through various routes, with the major route of exposure being the ingestion of contaminated food. OPFRs cause various toxicities, including neurotoxicity and endocrine disruption (Rosenmai et al., 2021; Wang and Song, 2024). Therefore, it is important to monitor OPFRs and evaluate the need for regulation of these substances.

Because OPFRs have been detected in various environments, they are expected to be detected in various food groups including agricultural, aqueous, and livestock products. OPFRs are lipophilic and are expected to be detected at high frequencies in fat-rich foods (Bika et al., 2022). Accordingly, a comprehensive analysis of foods with high lipid content is necessary. Meat products are a representative food group exhibiting a high lipid content; therefore, studying the contamination tendency of OPFRs in various types of meat product is of high value. There are few studies which monitored OPFRs concentration in meat products, however, there is no report of OPFRs monitoring in meat products from South Korea.

In this study, we developed and validated an LC-MS/MS analytical method for 11 OPFRs in meat products using QuEChERS-based extraction and purification method. A monitoring study was conducted by applying the developed method to various meat products including beef, pork, lamb, and duck. This is the first report on the monitoring of OPFRs in meat products in the South Korea, therefore, this study is of high significance for the local society.

2. Materials and methods

2.1. Chemicals and reagents

Analytical grade standards of triethyl phosphate (TEP), tris (2-chloroethyl) phosphate (TCEP), [2-[bis(2-chloroethoxy) phosphoryloxymethyl]-3-chloro-2-(chloromethyl)propyl] bis(2-chloroethyl) phosphate (V6), tris(1-chloropropan-2-yl) phosphate (TCIPP), tris(1,3-dichloropropan-2-yl) phosphate (TDCIPP), tributyl phosphate (TBP), triphenyl phosphate (TPhP), tris (2-butoxyethyl) phosphate (TBOEP), tris(4-methylphenyl) phosphate (TPCP), 2-ethylhexyl diphenyl phosphate (EHDPP), and tris(2-ethylhexyl) phosphate (TEHP) were purchased from AccuStandard (New Haven, CT, USA). Deuterated OPFR standards were used as internal standards (ISs). Ten commercially available ISs excluding EHDPP were purchased from Toronto Research Chemical (Toronto, ON, Canada). The physicochemical properties of the selected OPFRs are presented in Table 1. HPLC-grade methanol, acetonitrile, and water were purchased from J.T. Baker (Phillipsburg, NJ, USA). Dichloromethane and NaCl were purchased from Samchun Chemicals (Seoul, South Korea). A QuEChERS Clean-up Kit (MgSO₄ 900 mg, primary secondary amine (PSA) 150 mg, and octadecyl silica (C18) 150 mg), formic acid, and acetic acid were purchased from Sigma-Aldrich (St. Louis, MO, USA). The OPFRs standards and ISs solution was prepared in the range of 0.5–40 µg/mL by diluting with methanol. The working standard solution was stored in a freezer at −80 °C until analysis.

2.2. Sample preparation

2.2.1. Sample collection

The food samples used in this study were selected according to the 2021 Korea National Health and Nutrition Examination Survey (KNHANES) for the most consumed meat products based on their average intake. Twelve types of meat products were selected, including pork belly, pork leg, pork loin, pork rib, pork by-products, beef loin, beef brisket, beef rib, beef by-products, lamb, duck meat and chicken; a total of 105 samples were collected. Among the different meat products, pork loin was the food with the highest average intake; it was therefore selected as the representative food for developing the analytical method. Six cities in South Korea were selected as the sample collection sites: Seoul, Gangneung, Daejeon, Gwangju, Daegu, and Busan. Food samples

were collected from the local markets and supermarkets in each city. Considering the representativeness of the food samples, at least 1 kg of each sample was collected. The collected samples were homogenized using a grinder, followed by vacuum packaging, and stored in a freezer at −20 °C until further use. Since vacuum packaging used during sample collection could potentially become a source of OPFR contamination, we evaluated the contamination levels in the packaging materials and confirmed that OPFRs were presented at negligible levels.

2.2.2. Sample extraction optimization

The laboratory equipment used in this study including tubes and pipette tips was washed twice with dichloromethane to remove any existing OPFRs and prevent possible false positives. The extraction of OPFRs was conducted using 2 g of the homogenized sample in a 15 mL conical tube. A 9:1 mixture of acetonitrile and dichloromethane (5 mL) containing 1 % acetic acid was added together with 1 mL of 0.1 mol/L EDTA and ammonium formate in distilled water, followed by vortexing. NaCl (1.5 g) was added, and the mixture was vortexed for 10 min, followed by centrifugation at 2700 g at 4 °C for 10 min. The supernatant was transferred to a new 15 mL conical tube. For sample clean-up, 75 mg of MgSO₄, 12.5 mg of PSA, and 12.5 mg of C18 were added for dispersive solid phase extraction (dsPE) as sorbents, vortexed for 10 min, centrifuged, and the supernatant was transferred to a new tube. The solvent was completely dried using a gentle nitrogen stream and the sample was redissolved with 0.4 mL of methanol, vortexed for 10 min, and centrifuged at 13500 g at 4 °C for 10 min. The supernatant was transferred to LC vials and was analyzed by LC-MS/MS.

2.3. LC-MS/MS analysis

OPFRs analysis was conducted on a Vanquish HPLC system equipped with an Altis triple quadrupole mass spectrometer (Thermo Scientific, Waltham, MA, USA) using a Gemini C18 column (2 × 150 mm, 3 µm, Phenomenex, Palo Alto, CA, USA). The mass spectrometer was installed with a heated electrospray ionization probe (HESI-II) as the ion source. The sample injection volume was 5 µL, and the flow rate was maintained at 0.25 mL/min. The mobile phases consisted of water with 0.1 % formic acid (A) and acetonitrile with 0.1 % formic acid (B) at the following gradient elution: 0–1.5 min, 38 % B; 1.5–3.5 min, increase to 100 % B; 3.5–9.5 min, 100 % B; 9.51–13 min, 38 % B. The analysis was performed in the positive mode. The ion source conditions were set as: ion spray voltage, 3500 V; ion transfer temperature, 325 °C; sheath gas, 40 (arbitrary unit); auxiliary gas, 5 (arbitrary unit). For each 11 standards and 10 ISs, the precursor ion, product ion, and collision energy were optimized by tuning with the infusion of standards and then analyzing in the MRM mode using the corresponding conditions (Table 2).

Table 2

Retention times and MRM parameters for LC-MS/MS determination of OPFRs in meat products.

OPFRs	Retention time (min)	Precursor ion (m/z)	Product ion (m/z)
TEP	2.84	183.1	155.0; 127.0; 99.0
TCEP	4.79	285.0	223.0 ; 161.0; 99.0
V6	5.55	582.8	360.9 ; 358.9; 234.9
		327.1	99.0
TCIPP	5.57	329.1	254.0; 99.0
		430.9	321.0; 99.0
TDCIPP	6.01	432.9	99.0
TPhP	6.21	327.1	232.9; 215.0; 152.1
TBP	6.28	267.1	211.1; 155.0; 99.0
TBOEP	6.45	399.2	299.1 ; 199.1; 101.1
TPCP	6.78	369.1	243.0; 166.1; 165.1
EHDPP	7.09	363.2	250.9 ; 152.9; 151.9
TEHP	10.68	435.3	323.2; 211.0; 99.0

2.4. Method validation

The developed method was validated for its specificity, range, linearity, accuracy, precision, limit of detection (LOD), and limit of quantitation (LOQ), according to the CODEX guidelines. Selectivity was evaluated by comparing the chromatograms of the procedural blanks and standard-spiked samples, confirming the absence of any overlapping peaks at the retention times of OPFRs. Five concentrations were selected to evaluate linearity, and a calibration curve was plotted with a spiked IS. The linearity was confirmed by regression coefficient values higher than 0.99. Accuracy was evaluated based on the recovery by selecting three concentrations in the linearity range, namely low, medium, and high, and using pork loin as a representative meat product. Each evaluation was conducted in triplicate, and the relative standard deviation (RSD) values were calculated to evaluate intraday and inter-day precision. The LOD and LOQ were calculated using the standard deviation obtained by injecting seven replicates of the lowest linear concentration. This standard deviation was multiplied by 3.3 and 10 for the LOD and LOQ, respectively. Additionally, to evaluate laboratory interference and contamination of OPFR, three procedural blanks were prepared and analyzed for every batch of samples, and the OPFRs detected in the blank were subtracted from the OPFRs detected in the sample.

2.5. Data processing

The estimated daily intake (EDI, ng/kg bw/day) of OPFRs was calculated using Eq. (1).

$$EDI = \frac{\text{Concentration of OPFRs in food} \times \text{Daily food consumption}}{\text{Body weight}} \quad (1)$$

Food consumption data were collected from the 2021 KNHANES and 62.4 kg was used as the average body weight of the Korean population. The risk of OPFRs can be evaluated using hazard quotients (HQ), calculated by dividing the EDI by the reference dose (RfD) using Eq. (2).

$$HQ = \frac{EDI}{RfD} \quad (2)$$

RfD (ng/kg bw/day) is the quantity of chemicals that a person can be exposed to every day for their entire life, with no adverse health effects. If the HQ value is less than 1.0, the risk of OPFRs from the intake of meat products would be negligible, and no hazardous effects are observed in the exposed population (Román-Ochoa et al., 2021).

3. Results and discussion

3.1. Method optimization

For the simultaneous analysis of OPFRs, the MRM parameters were optimized via direct infusion of the standard solution. OPFRs were efficiently analyzed in the ESI positive mode, and three MRM transitions were set for each OPFR according to the tuning results. The ion with the highest intensity was used as the quantitative ion, and the remaining two ions were used as the qualitative ions. All analytes were detected as protonated ions $[M+H]^+$, and 0.1 % formic acid was added to the mobile phase to improve the peak shape and sensitivity of the detected analytes. The gradient elution of mobile phases A and B was adjusted to establish the HPLC conditions affording maximum separation of the 11 OPFRs and 10 ISs, thus minimizing the matrix effect and maximizing the sensitivity for each analyte. The optimized retention times and MRM transitions of the OPFRs and ISs are summarized in Table 2.

Because food samples have a complex matrix, sample preparation is a critical step for the simultaneous analysis of analytes with satisfactory sensitivity. OPFRs commonly contain phosphate triesters as the backbone, with each OPFR having a different side chain. Therefore, they exhibit similar chemical properties, which is advantageous for their

simultaneous extraction. In this study, it was necessary to establish an extraction and purification procedure that would efficiently remove interfering substances from the matrix, considering that meat products include muscles which mainly contain proteins and lipids depending on the part of meat product. Therefore, a purification step is required to eliminate any interferences caused from such substances. The extraction of OPFRs from homogenized meat products was conducted using a mixture of dichloromethane and acetonitrile as the extraction solvent, followed by the addition of acetic acid and an aqueous solution of EDTA with ammonium formate. NaCl (1.5 g) was then added, and the mixture was vortexed for 10 min. NaCl was selected as the salt for this method to control the polarity of the extraction solvents thereby increasing selectivity (Perestrelo et al., 2019). EDTA was added to prevent the formation of complexes between the cations of the added salt and analytes (Cho et al., 2015; Zhang et al., 2019). EDTA can form chelates with metal ions instead of analytes, thereby increasing the recovery and sensitivity of the method. The mixture was centrifuged to remove any substances that can induce matrix effects, such as proteins, and the supernatant was transferred to a new tube for the subsequent steps. Then, the dSPE sorbent was added to remove any co-extracted interferences from the matrix. C18 and PSA were used as the dSPE sorbents, along with $MgSO_4$ as a salt. PSA is used to remove polar interferences such as fatty acids, sugars, and organic acids, while C18 is used to adsorb and remove non-polar substances such as high-content lipids. Thus, interfering substances contaminating the mass spectrometer and reducing its sensitivity can be efficiently removed via adsorption onto the dSPE sorbent. The sorbent was then precipitated via centrifugation, and the supernatant was transferred to an LC vial and analyzed by LC-MS. The resulting analytical chromatogram of OPFRs from meat products is shown in Fig. 1. After optimization of the gradient conditions, most of OPFRs were baseline separated, except for V6, TCIPP, TBP, and TPhP. However, these compounds have different MRM transitions which could be successfully quantified.

3.2. Method validation

Before applying the developed method to food samples, method validation was conducted according to the CODEX guidelines (CODEX CAC-GL71–2009). The developed method was validated in terms of specificity, linearity, accuracy, precision, LOD, and LOQ. The specificity was evaluated by comparing the chromatograms of the standard solution and procedural blanks, revealing the absence of interference at the same retention time as those of OPFRs. The linearity was evaluated by plotting a calibration curve with five concentration points. The linear concentration range was determined based on the sensitivity for each analyte at concentrations of 1.0, 5.0, 10.0, 15.0, 20.0 ng/g for TCEP; 2.0, 10.0, 20.0, 30.0, and 40.0 ng/g for TDCIPP; and 0.5, 2.5, 5.0, 7.5, and 10.00 ng/g for the remaining analytes. All 11 OPFRs showed regression coefficients greater than 0.99, indicating satisfactory linearities. The LOD and LOQ values were calculated by repeated analysis of OPFRs at the lowest concentration in the linearity range, using the standard deviation from the repeated analysis. As a result, the LOD and LOQ were confirmed to be 0.06–0.62 ng/g and 0.16–1.59 ng/g, respectively (Table 3). Our results were compared with those of previous studies on OPFRs; previous LC-MS-based methods reported LOQ values ranging between 1.4–3.7 ng/g (Xu et al., 2015), and 0.07–0.42 ng/g (Ding et al., 2018), while GC-MS based analysis were ranged 0.15–1.00 ng/g (Poma et al., 2017), and 0.001–1.61 ng/g (Poma et al., 2018). Such values are in agreement with our results. Since OPFRs are ubiquitous environmental contaminants, evaluation of procedural blanks is essential when establishing the LOQ. According to CODEX guidelines for such contaminants, the concentration in procedural blanks should ideally not exceed 30 % of the signals from LOQ. In compliance with this guideline, we assessed the procedural blanks and confirmed that the detected levels were below 30 % of the corresponding signals in LOQ level.

The accuracy of the developed method was confirmed via recovery

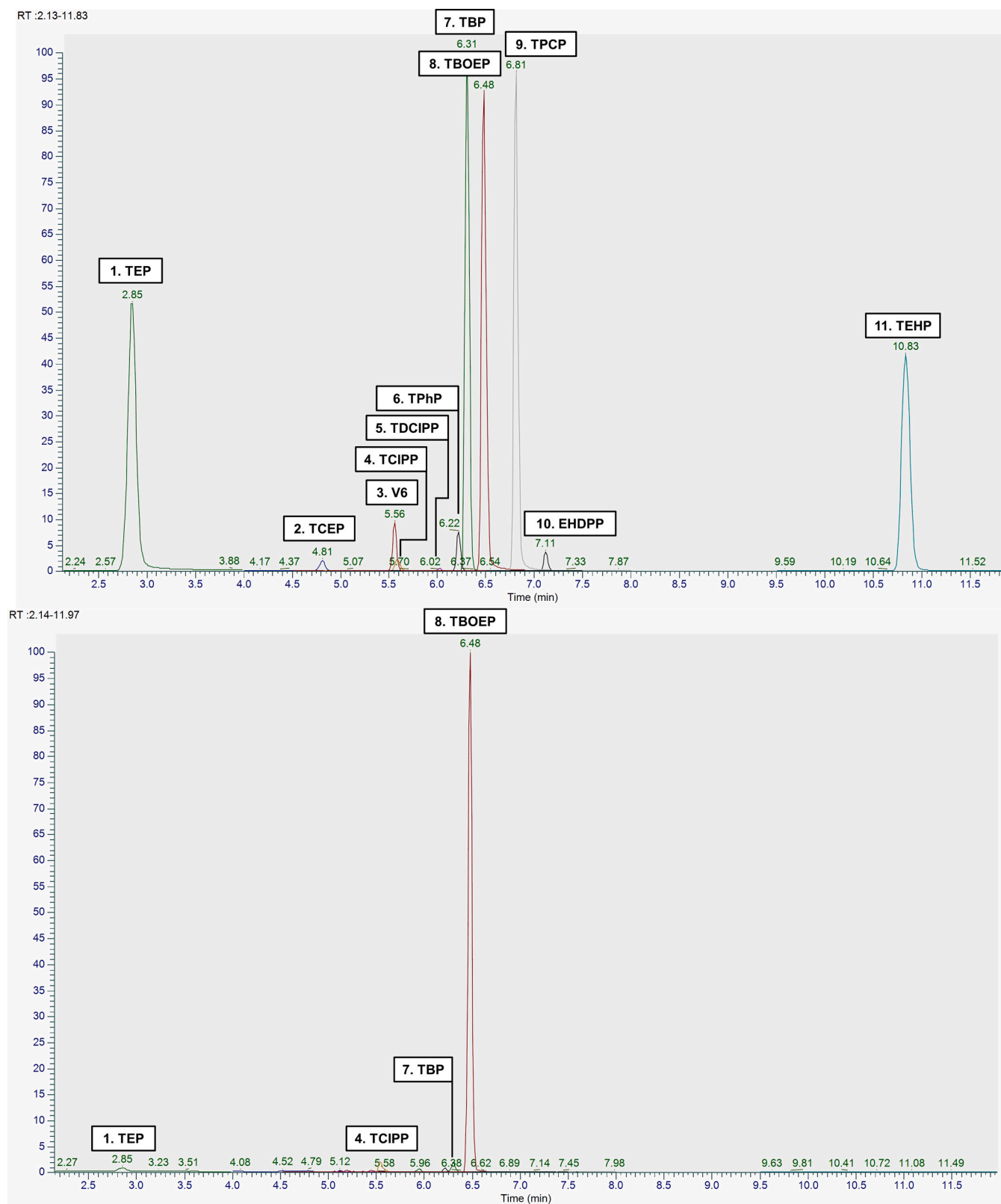


Fig. 1. (A) Chromatograms of 11 OPFRs standard (B) Chromatograms of OPFR from duck meat.

experiments using standard-spiked samples and matrix-matched standards. For the evaluation of accuracy, three concentrations (low, medium, and high) were selected based on the linearity range. As a result, the recovery of 11 OPFRs at low concentration was 48.4–115.4 % which was within the 40–120 % range suggested by the CODEX guidelines at

concentrations lower than 1 ppb. The recovery at medium and high concentrations also met the requirements in the range of 60 ~ 120 %. Precision was confirmed by calculating the RSD of the analytical results, exhibiting RSD values less than 35 % according to the CODEX guidelines. The accuracy and precision of the results for the 11 OPFRs are

Table 3

Range, linearity, LOD, and LOQ values of the developed method.

OPFRs	Linear ranges (ng/g)	R ²	LOD (ng/g)	LOQ (ng/g)
TEP	0.5–10.0	0.999	0.09	0.22
TCEP	1.0–20.0	0.999	0.22	0.56
V6	0.5–10.0	0.997	0.10	0.27
TCIPP	0.5–10.0	0.998	0.18	0.45
TDCIPP	2.0–40.0	0.999	0.62	1.59
TPhP	0.5–10.0	0.999	0.06	0.15
TBP	0.5–10.0	0.997	0.20	0.50
TBOEP	0.5–10.0	0.993	0.19	0.48
TPCP	0.5–10.0	0.999	0.06	0.16
EHDPP	0.5–10.0	0.999	0.08	0.20
TEHP	0.5–10.0	0.996	0.13	0.33

presented in Table 4.

3.3. Application to real samples

The developed method was applied to determine the OPFR content in 12 types of meat products obtained from local Korean markets. Twelve types of products were selected based on the consumption of meat products in Korea: pork belly, pork leg, pork loin, pork rib, duck, pork by-products, beef loin, beef brisket, beef rib, beef by-product, lamb, duck meat, and chicken. Seven to ten replicates were collected from six Korean cities. Considering the ubiquitous contamination of OPFRs during the monitoring process, procedural blanks were analyzed in every batch. When OPFRs were detected in a procedural blank, the corresponding concentrations were subtracted from the values obtained in the sample batch, and the adjusted concentrations were used as measured values. The measured OPFRs concentrations in the meat

Table 4

Accuracy and precision of the developed method.

OPFRs	Spiked Conc. (ng/g)	Intraday		Interday	
		Accuracy (Recovery, %)	Precision (RSD)	Accuracy (Recovery, %)	Precision (RSD)
TEP	0.5	103.0 ± 4.8	4.7	105.6 ± 3.9	3.7
	5	106.7 ± 2.3	2.1	107.8 ± 0.8	0.7
	10	106.6 ± 1.5	1.4	106.7 ± 0.5	0.5
TCEP	1	96.2 ± 17.2	17.9	97.6 ± 6.0	6.1
	10	102.8 ± 2.6	2.5	104.0 ± 0.8	0.8
	20	104.2 ± 1.8	1.7	104.4 ± 1.2	1.1
V6	0.5	97.6 ± 1.7	1.8	95.8 ± 1.3	1.4
	5	107.1 ± 4.5	4.2	107.7 ± 1.3	1.2
	10	105.8 ± 4.7	4.4	106.1 ± 1.0	0.9
TCIPP	0.5	48.4 ± 11.3	23.4	57.4 ± 6.4	11.2
	5	109.9 ± 6.3	5.7	111.7 ± 1.5	1.3
	10	110.4 ± 4.2	3.8	110.5 ± 0.9	0.8
TDCIPP	2	114.3 ± 19.5	17.1	107.5 ± 5.0	4.7
	20	80.4 ± 16.5	20.6	94.9 ± 10.7	11.2
	40	104.6 ± 26.5	25.3	109.0 ± 3.7	3.4
TPhP	0.5	93.2 ± 12.6	13.5	91.7 ± 1.9	2.1
	5	114.6 ± 7.0	6.1	116.6 ± 1.5	1.2
	10	107.9 ± 6.0	5.5	108.3 ± 1.0	0.9
TBP	0.5	53.4 ± 5.5	10.3	53.8 ± 8.3	15.4
	5	90.0 ± 4.9	5.5	91.3 ± 1.6	1.7
	10	87.5 ± 6.4	7.3	86.3 ± 1.3	1.5
TBOEP	0.5	98.9 ± 3.5	3.5	93.3 ± 4.4	4.7
	5	98.7 ± 3.5	3.5	97.4 ± 3.0	3.0
	10	98.0 ± 5.9	6.1	98.2 ± 1.8	1.9
TPCP	0.5	99.2 ± 7.0	7.0	99.8 ± 4.4	4.4
	5	94.5 ± 2.4	2.5	95.0 ± 0.5	0.5
	10	98.6 ± 8.5	8.6	100.0 ± 1.1	1.1
EHDPP	0.5	98.5 ± 1.5	1.6	95.9 ± 7.2	7.5
	5	86.9 ± 4.4	5.1	83.9 ± 3.7	4.4
	10	99.7 ± 16	16.0	99.5 ± 0.2	0.2
TEHP	0.5	104.4 ± 8.0	7.7	107.2 ± 3.3	3.0
	5	109.4 ± 5.6	5.1	114.2 ± 3.6	3.2
	10	114.2 ± 4.4	3.9	122.6 ± 6.0	4.9

products are presented in Fig. 2 and Table 5. In this study, OPFR with the highest average concentration was TCIPP, accounting for 39 % of the total OPFRs, with an average concentration of 1.56 ng/g. The sample with the highest TCIPP content was the pork by-products, with an average concentration of 8.45 ng/g. The second highest TCIPP concentration was detected in chicken samples, with an average concentration of 5.38 ng/g. The OPFR with the second highest average concentration was TBOEP, accounting for 28 % of the total detected OPFR amount with an average concentration of 0.78 ng/g, followed by TEP (0.27 ng/g) and TPhP (0.21 ng/g), exhibiting the highest detection amounts in duck meat and pork belly, respectively.

In this study, the tested samples mainly contained the edible parts of chicken, duck, pork, beef, and lamb. All samples exhibited a total OPFRs concentrations of less than 10 ng/g, with the highest total OPFRs concentration below 10 ng/g were duck meat (9.96 ng/g) followed by pork by-products (8.72 ng/g), and chicken (5.87 ng/g). The food type with the least amount of OPFRs was pork rib (0.10 ng/g). The OPFR levels did not exhibit any tendency with respect to the type or species of meat product. Contamination of OPFRs in meat products has also been reported in Sweden, Belgium, and China, and these findings can be compared with the present results to assess the OPFRs contamination status in Korea.

A monitoring study from Sweden showed different profiles of OPFRs compared with studies from South Korea. In Swedish meat products, unlike the trend observed in meat products in South Korea, EHDPP exhibited the highest concentration (0.64 ng/g), whereas TCIPP, which was the OPFR with the highest concentration in South Korea, exhibited the lowest concentration (0.08 ng/g) (Poma et al., 2017). TBOEP and TBP, which are highly ranked compounds in South Korea, were not detected in the Swedish samples. In another study, Belgian meat product samples were analyzed for seven OPFRs, revealing a total average concentration of 4.11 ng/g detected in the following order: TPhP (1.52 ng/g) > TDCIPP (0.8 ng/g) > EHDPP (0.72 ng/g) > TEHP (0.35 ng/g) > TCIPP (0.29 ng/g) > TBP (0.23 ng/g) > TCEP (0.2 ng/g) (Poma et al., 2018). EHDPP is a compound used in food packaging that is approved by the U.S. Food and Drug Administration (US-FDA). Therefore, it can be transferred to food when used as a packaging material. TPhP can be derived from the electrical equipment used for flame retardants and plasticizers. With the different OPFR profiles from previous studies, OPFRs appear to have various detection tendencies depending on industrial characteristics, region, and country.

China is one of the countries with high OPFR production, and several monitoring studies have been conducted, revealing various trends depending on the food sampling region. Chen et al. (2021) analyzed food collected from markets in 24 provinces in China and found that meat products had the highest average OPFR concentration (16.3 ng/g) (Chen et al., 2021). Among the detected OPFRs, EHDPP (1.63 ng/g) exhibited the highest median concentration, followed by TCIPP (0.977 ng/g), TCEP (0.812 ng/g), TDCIPP (0.414 ng/g), and TPhP (0.274 ng/g). Several studies have confirmed that pollution levels vary across countries. Therefore, it is necessary to assess and manage OPFR pollution in each country.

Using OPFR monitoring data from meat products, EDI was calculated to evaluate the risk of OPFR exposure through the intake of meat products. The reference doses (RfD, ng/kg bw/day) reported by the U.S. Environmental Protection Agency (US EPA), the EU Commission, and previous studies were used for the risk assessment which were 20000, 7000, 20000 ng/g bw/day for TBP, TCEP, and TCIPP, respectively. The calculated EDI of OPFRs for TBP, TCEP, and TCIPP by intake of meat products were 1.87, 1.55, and 81.0 ng/g bw/day, respectively. The HQ values for these OPFRs ranged from 9.34×10^{-5} to 4.05×10^{-3} , exhibiting very low values. For other OPFRs, sufficient toxicological data are currently unavailable, making it challenging to establish appropriate reference dose (RfD) values. Regarding TBOEP, which exhibited the second highest mean concentration (0.78 ng/g), similar levels have been reported in food samples studied from China (Bi and Su,

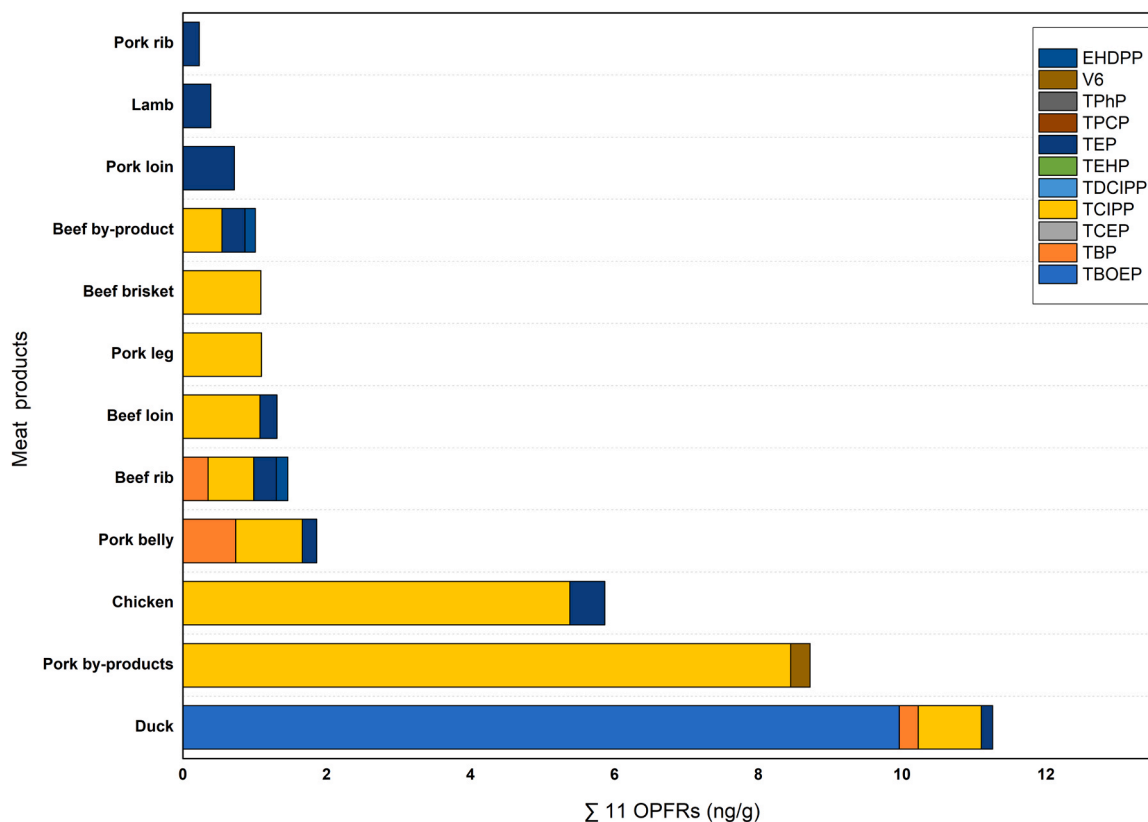


Fig. 2. Average amount of 11 OPFRs detected in 12 meat products.

Table 5

Contents of OPFRs in 12 meat products (ng/g).

Foods	TBOEP	TBP	TCEP	TCIPP	TDCIPP	TEHP	TEP	TPCP	TPhP	V6	EHDPP	Average
Pork belly	0.35	0.74	0.23	0.93	0.07	0.00	0.20	0.00	0.26	0.03	0.00	0.25
Pork leg	0.02	0.00	0.24	1.09	0.00	0.00	0.07	0.02	0.12	0.06	0.00	0.15
Pork loin	0.00	0.00	0.13	0.21	0.17	0.00	0.72	0.02	0.14	0.06	0.00	0.13
Pork rib	0.01	0.00	0.12	0.38	0.00	0.00	0.23	0.00	0.30	0.01	0.04	0.10
Pork by-products	0.00	0.00	0.07	8.45	0.29	0.00	0.07	0.00	0.06	0.27	0.00	0.84
Beef loin	0.28	0.00	0.35	1.07	0.14	0.00	0.24	0.00	0.12	0.09	0.00	0.21
Beef brisket	0.19	0.00	0.50	1.09	0.00	0.00	0.07	0.00	0.18	0.09	0.05	0.20
Beef rib	0.35	0.35	0.16	0.64	0.28	0.00	0.31	0.02	0.34	0.04	0.16	0.24
Beef by-products	0.03	0.00	0.04	0.54	0.00	0.00	0.32	0.00	0.01	0.05	0.14	0.10
Lamb	0.03	0.00	0.02	0.43	0.36	0.00	0.39	0.00	0.19	0.09	0.00	0.14
Duck meat	9.96	0.27	0.19	0.88	0.09	0.00	0.16	0.00	0.38	0.08	0.00	1.09
Chicken	0.00	0.00	0.10	5.38	0.00	0.00	0.49	0.00	0.45	0.04	0.00	0.59
Minimum Value	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	-
Maximum Value	69.70	7.36	2.94	36.19	2.06	ND	4.18	0.24	2.06	0.87	1.58	-
Average	0.78	0.12	0.19	1.56	0.11	0.00	0.27	0.01	0.21	0.07	0.03	-

2023). These findings also indicate a relatively low potential health risk. Additionally, the other OPFRs without EDI values showed low average concentration which are similar to TBP and TCEP. These low values indicate that the dietary intake of OPFRs in Korean meat products does not result in significant health risks in this study. Although individual OPFRs pose a low risk, the combined effects of several OPFRs are unclear, and require further study. As OPFRs are emerging contaminants, it is necessary to continue monitoring their levels in environments that are in close contact with daily life.

4. Conclusions

In this study, we successfully developed an analytical method to simultaneously detect and quantify 11 OPFRs in meat products using QuEChERS and LC-MS/MS. The developed method was successfully

validated based on the CODEX guidelines and was then applied for the analysis of meat products from a local Korean market. Among the 11 OPFRs, 6 compounds exhibited concentrations higher than the LOQ in 12 types of meat products, with TCIPP exhibiting the highest overall concentration. Based on the monitoring results, the human *per capita* exposure to OPFRs from meat product intakes was estimated and found to be much lower than the reported RfD values. Although the risk of meat product intake is low, exposure to OPFRs can happen not only through food intake but also via indoor dust. As OPFRs are not regulated worldwide, there is a possibility that their concentration in food and the environment may increase in the near future. Accordingly, the continuous management of these compounds is suggested in future research.

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CRediT authorship contribution statement

Taek Gu Han: Writing – original draft, Visualization, Validation, Methodology, Investigation. **Eun Seon Jeong:** Validation, Methodology, Investigation, Formal analysis. **Jin Hwan Kim:** Validation, Investigation. **Kyung Tae Kim:** Writing – review & editing, Resources, Conceptualization. **Yong Seok Choi:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Hyung Min Kim:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Dong-Kyu Lee:** Writing – review & editing, Resources, Conceptualization. **Sang Beom Han:** Writing – review & editing, Resources, Funding acquisition, Conceptualization. **Jong Seong Kang:** Writing – review & editing, Supervision, Resources, Conceptualization.

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. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jfca.2025.108775](https://doi.org/10.1016/j.jfca.2025.108775).

Data availability

Data will be made available on request.

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