



# Changes in acetaldehyde and formaldehyde contents in foods depending on the typical home cooking methods

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## ABSTRACT

Analytical methods were validated for the evaluation of acetaldehyde and formaldehyde, which are harmful chemicals, using solid-phase microextraction-gas chromatography/mass spectrometry in four different matrices. Typical home-cooking methods including boiling, pan-frying, and stir-frying, were applied to beef, rapeseed oil, canned pork ham, egg, and rice wine. In addition, monosaccharides, disaccharides, alanine, and glycine were heated for the formation of both aldehydes. All validation parameters, including accuracy, precision, limit of detection, limit of quantification, and uncertainty, for four different matrices were within recommended ranges, confirming the validity of the current method. Acetaldehyde contents ranged from undetectable to 17.92  $\mu\text{g/g}$  and formaldehyde contents ranged from undetectable to 0.27  $\mu\text{g/g}$ . Generally, boiling decreased both aldehydes except acetaldehyde in egg. Pan- and stir-frying increased both aldehyde content substantially in rapeseed oil whereas pan-frying increased acetaldehyde content in canned pork ham and egg. Fructose and sucrose produced higher content of both aldehydes than maltose and glucose when heated. Depending on food type, the cooking process had slightly different effects on the contents of acetaldehyde and formaldehyde.

## 1. Introduction

Acetaldehyde and formaldehyde are highly volatile compounds with low molecular weights. Acetaldehyde is naturally present in fruits, vegetables, dairy products, fruit drinks, meat, and fish (Miyake and Shibamoto, 1993; Yasuhara and Shibamoto, 1995). In the food industry, acetaldehyde is added to various beverages and fruit products as a flavor enhancer or a preservative. Acetaldehyde is an intermediate product of the alcoholic fermentation of fruits or sugar metabolism (Miyake and Shibamoto, 1993). In addition, acetaldehyde is formed by acetic acid bacteria in wine and by the metabolism of ethanol and phenolic compounds (Liu and Pilone, 2000) and a secondary product of the lipid oxidation of polyunsaturated fatty acids (Hsieh and Kinsella, 1989). Acetaldehyde has been shown to form covalent bonds with DNA as well as to induce point mutations and DNA repair deficiencies (Cheng et al., 2003; Wang et al., 2000; Espina et al., 1988). The International Agency for Research on Cancer (IARC) classified acetaldehyde as possibly carcinogenic to humans (Group 2B). In particular, acetaldehyde associated with the consumption of alcoholic beverages is classified as carcinogenic to humans (Group 1) by IARC (International Agency for Research on Cancer, France (IARC), 1999, 2012a).

Formaldehyde is naturally present in foods, such as fruits, vegetables, meats, fish, and crustaceans. Formaldehyde can be used as a preservative, reducing agent, fumigant, or sterilizing agent in various foods (Norliana et al., 2009). Formaldehyde is commonly formed as an intermediate product of normal metabolism in fruits and vegetables and is produced in seafood by the enzymatic decomposition of trimethylamine-oxide (TMAO) (Phillippy and Hultin, 1993; Kimura et al., 2003; Immaculate and Jamila, 2018). Strecker degradation of glycine (Velíšek et al., 1989) and the lipid oxidation of polyunsaturated fatty acids can generate formaldehyde (European Food Safety Authority (EFSA), 2014). The consumption of formaldehyde is related to an increased risk of leukemia and nasopharyngeal cancer (International Agency for Research on Cancer (IARC), 2012b). Formaldehyde is classified as carcinogenic to humans (Group 1) by IARC (International Agency for Research on Cancer (IARC), 2006).

Most previous studies of acetaldehyde and formaldehyde have focused on the contents of target compounds in foods available at markets or restaurants. Usually, food ingredients must be subjected to cooking processes to generate a consumable and/or acceptable product. Therefore, family members are exposed to acetaldehyde and formaldehyde originating from foods after the cooking process. However, the

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effects of home cooking processes on acetaldehyde and formaldehyde contents in foods have rarely been studied. In addition, even though several studies have characterized acetaldehyde or formaldehyde separately by using headspace solid phase microextraction-gas chromatography-mass spectrometry (HS-SPME-GC-MS) (Paiano et al., 2014; Bianchi et al., 2007), only a few reports have reported the determination of both acetaldehyde and formaldehyde in food (Jeong et al., 2015).

The main objectives of this study were to evaluate the effects of typical home cooking procedures on acetaldehyde and formaldehyde contents in foods using HS-SPME-GC-MS and to determine heating process on the acetaldehyde and formaldehyde content in mono- and disaccharides, glycine, and alanine. The analytical method was validated in terms of the linearity of the standard calibration curve, sensitivity (limit of detection and limit of quantification), repeatability (intra-day and inter-day), and accuracy (recovery). The typical home cooking methods used in this study were boiling, pan-frying, and stir-frying.

## 2. Materials and methods

### 2.1. Materials

Imported beef, rapeseed oil, canned pork ham, egg, and rice wine were obtained from Chungang University (Ansung, South Korea). Fructose, glucose, sucrose, and maltose were purchased from Junsei Chemical (Tokyo, Japan). Acetaldehyde, formaldehyde, glycine, alanine, and polydimethylsiloxane/divinylbenzene (PDMS/DVB) were purchased from Sigma Aldrich (St. Louis, MO, USA). 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde was purchased from Toronto Research Chemicals (Toronto, Canada). *O*-(2,3,4,5,6-Pentafluoro-benzyl)-hydroxylamine hydrochloride (PFBHA) was purchased from Sigma Aldrich. All chemicals were of analytical grade.

### 2.2. Sample preparation

Three types of standard cooking processes were used to prepare foods, including boiling, pan-frying, and stir-frying. Five types of foods were selected, including beef, rapeseed oil, canned pork ham, eggs, and rice wine, which are typical high-fat raw solid state, high-fat liquid, high-fat processed solid, low-fat solid, and low-fat liquid food matrices, respectively. Boiling was performed by heating foods in boiling water near 100 °C. Pan-frying and stir-frying were conducted on a pan. The heating temperatures for pan-frying and stir-frying differed depending on the types of food. For beef rib, the temperature was 180 °C, while other types of foods were heated at 170 °C. All three cooking procedures were used to prepare beef rib, rapeseed oil, canned pork ham, and eggs, whereas boiling and stir-frying were used for rice wine preparation.

In detail, beef rib was boiled for 17 min in water. Beef rib was pan-fried and stir-fried for 5 min in a pan. In the case of rapeseed oil, the cooking times for pan-frying and stir-frying were 2 min. The canned pork ham was boiled for 20 min. Canned pork ham was pan-fried and stir-fried on a pan for 3 min at 180 °C. In the case of eggs, egg yolk and egg white were mixed gently and used for cooking treatment. The cooking times of eggs for boiling, pan-frying, and stir-frying were 9, 2, and 2 min, respectively. Rice wine (100 mL) was added to 100 mL of boiled water and heated for 6 min, as a type of boiling. Rice wine in a pan was heated for 2 min with stirring. Cooked samples were kept at -70 °C until analyses.

Mono- and di-saccharide including fructose, glucose, sucrose, and maltose and amino acid such as glycine and alanine were prepared in deionized water at the final concentration of 0.1 M. An aliquot of 6 mL solution was transferred into a 10-mL glass vial (Agilent, USA), which was hermetically sealed with a silicone coated rubber septa and an aluminum cap using a crimper. The sample vial was placed in a pre-heated convection oven (Hysclab, Seoul, Korea) for 2 h at 160 °C. Samples were prepared in triplicate.

### 2.3. Sample preparation and derivatization

Acetaldehyde, formaldehyde, and 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde stock standard solutions were prepared at 10 µg/mL in deionized water and stored at 4 °C. Samples other than rice wine (1 g) were mixed with 9.950 mL of 30% NaCl solution and 50 µL of 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde as an internal standard (10 µg/mL, w/v) and then treated by sonication at room temperature for 30 min. After the sample was centrifuged (1660 × g) at 4 °C for 10 min and vortex-mixed for 30 s, the supernatant (5 mL) was transferred to 10-mL headspace vials with screw caps. For rice wine, 4.475 mL of 30% NaCl solution and 25 µL of the internal standard were added to 0.5 mL of rice wine and then sonicated at ambient temperature for 30 min.

For derivatization, 100 mg of potassium hydrogen phthalate and 50 µL of PFBHA (10 mg/mL) were added to 5 mL of the above samples and maintained at 45 °C for 40 min in a water bath (Lab Companion, Seoul, Korea). Analyses of aldehydes in each sample were conducted in triplicate.

### 2.4. Analysis of acetaldehyde and formaldehyde using SPME-GC/MS

SPME equipped with PDMS/DVB was used to adsorb acetaldehyde and formaldehyde in the headspace of samples and desorbed volatiles were separated using gas chromatography (GC) and analyzed using a mass selective detector (MS). A Gerstel MPS Autosampler (Gerstel, Mülheim, Germany) was used to isolate acetaldehyde and formaldehyde in the samples at 45 °C for 15 min. The GC (6890 N series; Agilent Technologies, Palo Alto, CA, USA) was connected to a 5975B MS (Agilent Technologies) equipped with a DB-5MS column (30 m length × 0.25 mm i.d. × 0.25 µm film thickness; Agilent Technologies). Helium was used as a carrier gas at a constant column flow rate of 0.8 mL/min. A splitless injection mode at 220 °C for 5 min was used to desorb the adsorbed volatiles from SPME. The GC oven temperature was held at 50 °C for 2 min, raised to 120 °C at a rate of 30 °C/min, and then held at 200 °C for 10 min. Mass spectra were obtained at 70 eV by electron ionization (EI). Data were acquired in the selected-ion monitoring mode (SIM mode). Acetaldehyde, formaldehyde, and 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde derivatives were quantified at *m/z* 209, 195, and 211, respectively (Jeong et al., 2015). Additionally, 181 *m/z* was monitored for qualifier ions of acetaldehyde, formaldehyde, and acetaldehyde 1, 2-<sup>13</sup>C<sub>2</sub> (Jeong et al., 2015). For quantification, peak areas of the *m/z* 209 and the *m/z* 195 were obtained acetaldehyde and formaldehyde, respectively and were used for the calculation of aldehyde concentration using peak areas of the *m/z* 211 from 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde.

### 2.5. Method validation

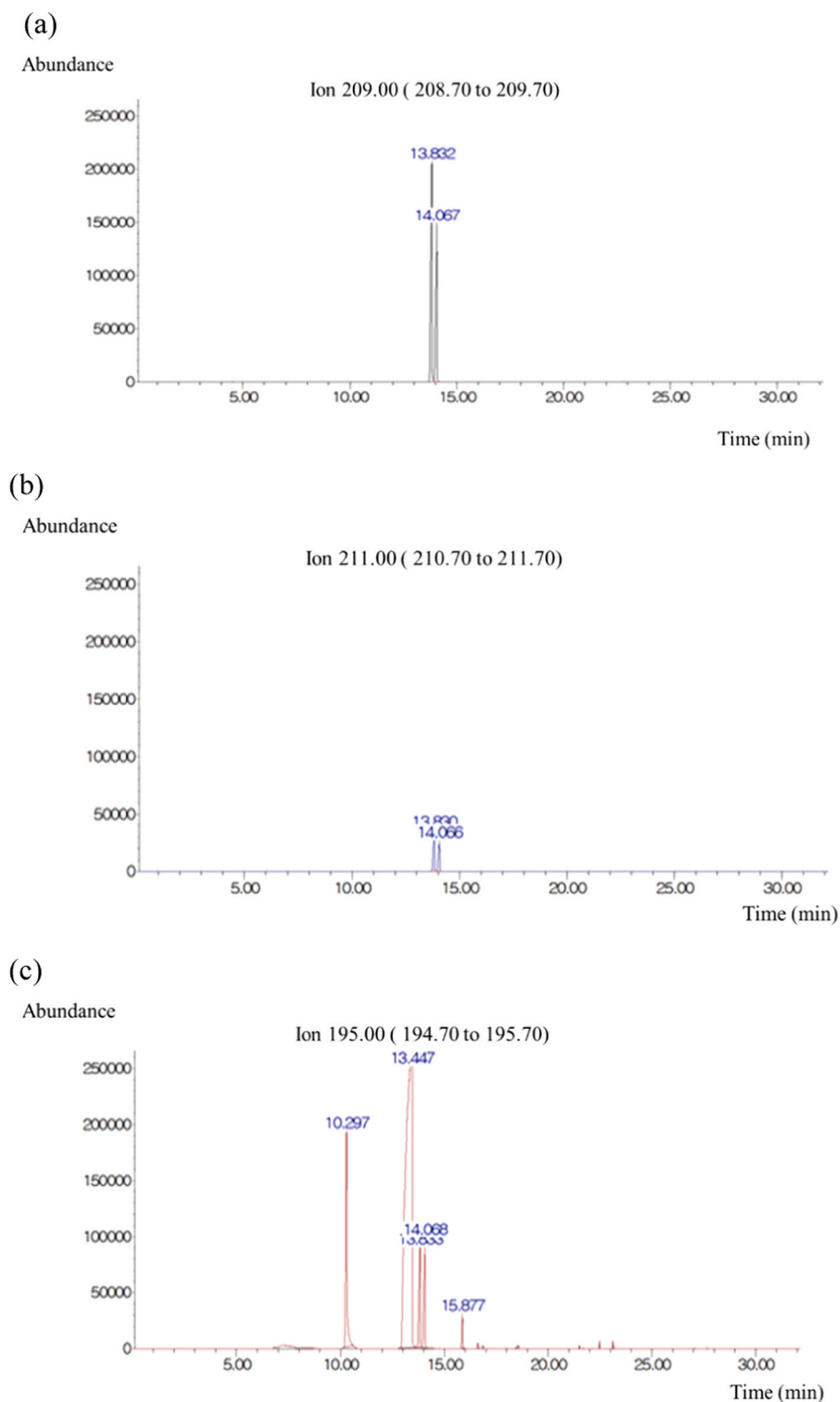
The method was validated in terms of the linearity of standard calibration curves, limit of detection (LOD), and limit of quantification (LOQ) in standard solution and sample matrices, including beef, milk, mayonnaise, and egg, intra-day and inter-day precision, and accuracy. For the determination of linearity, calibration curves were prepared in the range of 0.05–10.00 µg/g using beef, milk, mayonnaise, and egg samples. For LOD and LOQ of the method detection limit (MDL), calibration curves were prepared in the range of 0.001–0.01 µg/g using four types of food matrices and standard solutions. The sample was analyzed at five points, and each point was used for five repeated analyses. LOD and LOQ were calculated using the following formulas:

$$\text{LOD} = 3.3 \times \sigma/S$$

$$\text{LOQ} = 10 \times \sigma/S$$

Where,  $\sigma$  = standard deviation of the y-intercept of the standard calibration curve regression line; S = slope of the regression line.

Recovery tests were performed with the four matrices spiked with authentic acetaldehyde and formaldehyde compounds at three different



**Fig. 1.** Chromatograms of acetaldehyde (a),  $^{13}\text{C}_2$ -acetaldehyde (b), and formaldehyde (c). Quantitation ions of 209  $m/z$  (a), 211  $m/z$  (b), and 195  $m/z$  (c) were for acetaldehyde,  $1,2\text{-}^{13}\text{C}_2$ -acetaldehyde, and formaldehyde, respectively.

concentrations (0.5, 2.5, and 5.0  $\mu\text{g/g}$  for acetaldehyde and 0.25, 0.50, and 1.00  $\mu\text{g/g}$  for formaldehyde). The intra-day repeatability of the analytical method was tested by conducting five repeated analyses of the standard solutions. The contents of acetaldehyde and formaldehyde in four matrices were analyzed on three different days to evaluate the inter-day precision of the analytical method.

## 2.6. Statistical analysis

The acetaldehyde and formaldehyde data were evaluated using one-way analysis of variance (ANOVA) and Duncan's multiple range tests using SPSS version 19 (SPSS Inc., Chicago, IL, USA). Differences with a  $p$ -value of  $< 0.05$  were considered statistically significant.

**Table 1**

Method validation parameters of linearity of standard calibration curves, LOD and LOQ for acetaldehyde and formaldehyde in standard solution.

Analyte	Calibration curve	R <sup>2</sup>	LOD <sup>1</sup> (µg/g)	LOQ (µg/g)
Acetaldehyde	y = 0.085x + 0.048	0.999	0.001	0.003
Formaldehyde	y = 0.008x + 0.379	0.993	0.003	0.007

<sup>1</sup> LOD and LOQ are limit of detection and limit of quantification, respectively.

### 3. Results and discussion

#### 3.1. Validation of methods for acetaldehyde and formaldehyde detection

Gas chromatograms for acetaldehyde (a), 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde (b), and formaldehyde (c) are shown in Fig. 1. The *m/z* 209, 211, and 195 ions were used as quantifying ions for the acetaldehyde, 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde, and formaldehyde, respectively. Acetaldehyde and 1,2-<sup>13</sup>C<sub>2</sub>-acetaldehyde can produce two isomers when derivatized with PFBHA. Two isomer peaks of acetaldehyde were observed at retention times (RT) of 13.832 and 14.067 min. The <sup>13</sup>C<sub>2</sub>-acetaldehyde peaks were observed at RTs of 13.830 and 14.066 min. One chromatographic peak was produced from formaldehyde at a RT of 10.297 min (Fig. 1). Two isomers of the *m/z* 211, two isomers of the *m/z* 209, and one peak of *m/z* 195 were used for the quantification.

Method validation parameters, including the linearity of standard calibration curves, LOD, and LOQ, for acetaldehyde and formaldehyde in standard solution are shown in Table 1. The linearities (correlation coefficient, *r*) of the standard calibration curves of acetaldehyde and formaldehyde were 0.999 and 0.993, respectively. The LOD and LOQ for acetaldehyde were 0.001 and 0.003 µg/g, respectively, and those for formaldehyde were 0.003 and 0.007 µg/g, respectively. The intra-day and inter-day repeatability, recovery, and LOD and LOQ of acetaldehyde and formaldehyde in four different food matrices are shown in Table 2. The linearity (coefficient of determination, *r*<sup>2</sup>) of the standard calibration curves for acetaldehyde and formaldehyde in the food matrices were in the ranges of 0.998–0.999 and 0.992–0.999, respectively. The LOD and LOQ of acetaldehyde in the food matrices were in the ranges of 0.005–0.050 µg/g and 0.015–0.150 µg/g, respectively. In the case of formaldehyde, LOD and LOQ were in the ranges of 0.010–0.050 µg/g and 0.050–0.151 µg/g, respectively. The relative standard deviation (RSD) (%) of intra-day precision was 1.8–14.4%, whereas that of inter-day precision (%) was in the range of 3.1–9.0%. The recoveries of acetaldehyde and formaldehyde were in the ranges of 87.8–112.5% and 86.0–115.0%, respectively. To further confirm the accuracy of the analytical method, FAPAS proficiency testing for formaldehyde was performed. In this test, a *z*-score of –0.2 was obtained for formaldehyde in 37% ethanol. The submitted value for formaldehyde was 14.6 µg/kg, the mean value was 14.3 µg/kg, and acceptable

**Table 2**

Intra-day and inter-day repeatability, accuracy, and LOD and LOQ, of acetaldehyde and formaldehyde in 4 different food matrices.

Matrix	Analyte	Spiked (µg/g)	Intra-day Precision (RSD)	Inter-day Precision (RSD)	Accuracy (%)	LOD <sup>1</sup> (µg/g)	LOQ (µg/g)	Linearity (R <sup>2</sup> )
Milk	AA <sup>2</sup>	0.50	4.3	3.3	87.8	0.005	0.015	0.998
		2.50	5.7	3.1	102.7			
		5.00	6.5	4.6	105.6			
	FA	0.5	3.8	3.3	104.2			
		0.25	1.8	3.3	89.8			
		1.00	6.6	6.3	91.7			
Egg	AA	0.50	5.0	3.8	109.7	0.050	0.150	0.999
		2.50	4.5	4.0	107.6			
		5.00	10.1	5.4	112.5			
	FA	0.25	14.4	9.0	99.8			
		0.50	2.8	7.9	110.6			
		1.00	8.5	6.6	115.0			
Mayonnaise	AA	0.50	7.6	6.4	93.5	0.028	0.085	0.999
		2.50	3.3	3.8	96.5			
		5.00	4.5	3.3	92.4			
	FA	0.25	4.2	5.7	86.0			
		0.50	8.7	9.0	99.5			
		1.00	6.7	5.7	105.4			
Beef	AA	0.50	7.9	7.6	113.7	0.028	0.085	0.998
		2.50	8.8	4.4	110.6			
		5.00	5.4	3.4	106.2			
	FA	0.25	4.6	5.5	100.1			
		0.50	4.1	7.2	101.7			
		1.00	2.4	4.8	95.0			

<sup>1</sup> LOD and LOQ are limit of detection and limit of quantification, respectively.

<sup>2</sup> AA and FA are acetaldehyde and formaldehyde, respectively.

**Table 3**

Effects of cooking processing on the changes of acetaldehyde and formaldehyde in beef rib, rapeseed oil, canned pork ham, egg, and rice wine.

Cooking methods	Beef rib <sup>1</sup> (µg/g)		Rapeseed oil (µg/g)		Canned pork ham (µg/g)		Egg (µg/g)		Rice wine (µg/g)	
	AA <sup>2</sup>	FA	AA	FA	AA	FA	AA	FA	AA	FA
Non-cooked	2.94 ± 0.27 <sup>3</sup>	0.65 ± 0.03	0.16 ± 0.08	N.D. <sup>4</sup>	0.82 ± 0.09	0.20 ± 0.02	N.D.	0.22 ± 0.07	17.92 ± 0.77	0.10 ± 0.01
Boiling	0.34 ± 0.03	0.08 ± 0.01	N.D.	N.D.	0.47 ± 0.04	0.19 ± 0.03	0.26 ± 0.01	0.09 ± 0.01	0.38 ± 0.04	0.10 ± 0.02
Pan-frying	1.18 ± 0.10	0.06 ± 0.01	1.17 ± 0.05	0.18 ± 0.02	1.49 ± 0.01	0.27 ± 0.01	0.52 ± 0.12	0.17 ± 0.04	N.A. <sup>5</sup>	N.A.
Stir-frying	0.47 ± 0.07	0.08 ± 0.01	0.47 ± 0.07	0.21 ± 0.09	1.55 ± 0.08	0.32 ± 0.01	0.21 ± 0.02	0.22 ± 0.01	0.37 ± 0.01	0.07 ± 0.01

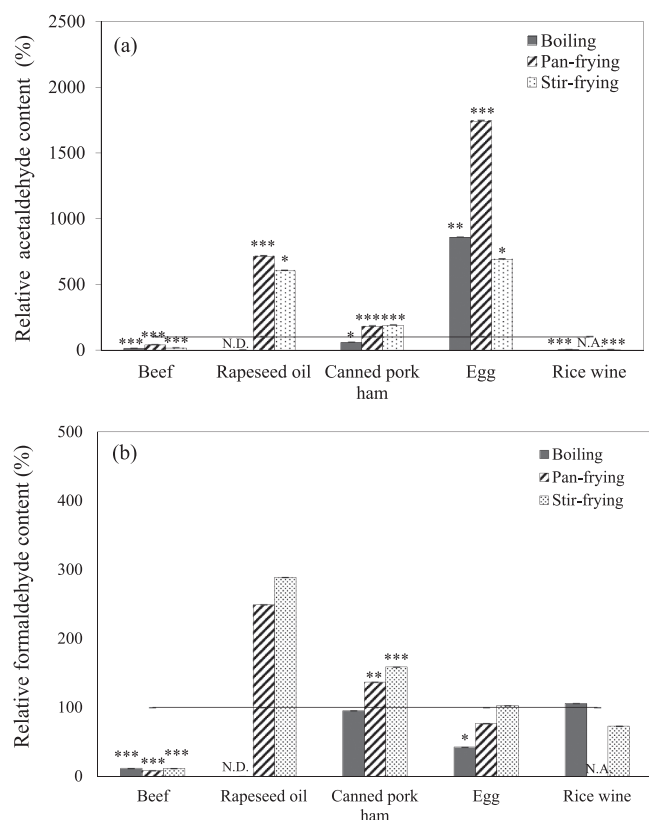
<sup>1</sup> Imported beef was used for non-cooked sample while beef rib was used for boiling, pan-frying, and stir-frying process.

<sup>2</sup> AA and FA are acetaldehyde and formaldehyde, respectively.

<sup>3</sup> Mean ± standard deviation (n = 3)

<sup>4</sup> Aldehyde was not detected.

<sup>5</sup> Sample was not available.



**Fig. 2.** Relative comparison of acetaldehyde and formaldehyde in food matrix depending on cooking processes. Values over and below 100 indicate that cooking treatment increases or decrease the content of aldehydes, respectively. N.D. means aldehydes were not detected. N.A. indicates that samples were not available. The values of 0.03  $\mu\text{g/g}$  AA from egg and 0.05  $\mu\text{g/g}$  FA from rapeseed oil were used for calculation. The values were lower than LOD and marked as N.D. in Table 3. \*, \*\*, and \*\*\*\* on the bar were significantly different at 0.05, 0.01, and 0.001, respectively.

tolerance levels were 11.4–17.7  $\mu\text{g/kg}$ , confirming the high accuracy of the analytical method (data not shown).

The UPLC method has been validated for the determination of formaldehyde in smoked meat products (Li et al., 2018). The reported detection limit of the UPLC method for formaldehyde was 0.25  $\mu\text{g/g}$ , which was higher than that of current results obtained using GC/MS (Table 1). The reported repeatability RSD was 2.10% and recovery of standard addition was 86.71–93.08% for the UPLC method, similar to the results of the current study (Table 2). The difference could be explained by differences in extraction methods, analytic instruments, and matrix types.

### 3.2. Effects of cooking on acetaldehyde and formaldehyde in foods

Effects of cooking processing on the acetaldehyde and formaldehyde contents in beef rib, rapeseed oil, canned pork ham, eggs, and rice wine are shown in Table 3. The acetaldehyde content ranged from undetectable to 17.92  $\mu\text{g/g}$ , whereas the formaldehyde content ranged from undetectable to 0.27  $\mu\text{g/g}$ . The acetaldehyde and formaldehyde contents in the imported beef were 2.94 and 0.65  $\mu\text{g/g}$ , respectively. High acetaldehyde content in the beef was also reported by Frank et al. (2020). Acetaldehyde is one of the most detected volatiles in beef and volatiles including acetaldehyde increased gradually in beef stored  $-1^\circ\text{C}$  for 140 days (Frank et al., 2020). Generally, cooking process especially boiling decreased acetaldehyde and formaldehyde contents in beef. The contents of acetaldehyde and formaldehyde in rapeseed oil were 1.17 and 0.18  $\mu\text{g/g}$  for pan-frying, and boiled samples did not have

detectable aldehyde contents. Pan-frying induced the highest level of acetaldehyde and formaldehyde in canned pork ham. The highest acetaldehyde content in eggs was 0.52  $\mu\text{g/g}$  after pan-frying, while acetaldehyde was not detected in uncooked eggs. For formaldehyde in eggs, no cooking was the highest content, while boiling was the lowest. For rice wine, contents of acetaldehyde and formaldehyde were significantly affected by the cooking method. The acetaldehyde content in rice wine was the highest in uncooked samples and decreased substantially after stir-frying and boiling.

A comparison of relative acetaldehyde and formaldehyde levels in food matrixes depending on the cooking process is shown in Fig. 2. Because acetaldehyde and formaldehyde contents in uncooked samples were regarded as 100%, values greater than and less than 100% indicate that cooking treatment increased or decreased the aldehyde content, respectively. All cooking processes decreased the both aldehyde contents in beef and rice wine while increased the acetaldehyde content in egg. Generally, boiling decreased both aldehyde contents except acetaldehyde in egg. Edible oil with high content of linolenic acid can generate more aldehydes when high temperature treatment was applied such as pan-frying and stir-frying (Fig. 2). Depending on food types, the cooking process had slightly different effects on the acetaldehyde and formaldehyde contents.

Formation of acetaldehyde and formaldehyde from mono- and disaccharides (a) and alanine or glycine (b) are shown in Fig. 3. Formation of acetaldehyde and formaldehyde was greatly affected by the type of sugars. The acetaldehyde content in heat-treated fructose and sucrose were significantly higher than heat-treated maltose and glucose. In the case of formaldehyde, heat treatment on fructose showed the highest content, followed by sucrose, maltose, and glucose (Fig. 3a). When glycine was heat-treated, formaldehyde was mainly produced whereas acetaldehyde was mainly produced in alanine (Fig. 3b).

Several studies have evaluated toxic aldehyde (acetaldehyde and formaldehyde) contents in various foods, including fruits and wines. Lachenmeier et al. (2013) reported that the acetaldehyde and formaldehyde contents in rice wine are 6–75  $\mu\text{g/mL}$  and 0–7.4  $\mu\text{g/mL}$ , respectively. Smoked meat contains considerable levels of formaldehyde (up to 125  $\mu\text{g/g}$  wet weight) (Zhu et al., 2012). In the smoking process, formaldehyde is generated by the oxidation of methanol, which is produced by dry distillation under oxygen-deficient conditions (Zhu et al., 2012). Huang et al. (2004) reported that pan-fried pork contains 7 ppm acetaldehyde and 6 ppm formaldehyde. The authors reported that superheated steam fried pork contains 11 ppm acetaldehyde and 16 ppm formaldehyde. However, the initial acetaldehyde and formaldehyde contents in raw pork were not reported. Accordingly, the extent to which the toxic compounds were actually induced by the cooking processes could not be estimated. Li et al. (2018) reported that the internal formaldehyde contents in smoked meat products is 25.55–49.20 mg/kg, while the surface formaldehyde content is 34.04–165.25 mg/kg. Reported formaldehyde contents in poultry products and alcoholic beverages are 5.7–20 and 0.27–3.0 ppm (European Food Safety Authority EFSA, 2014). Chung et al. (2015) reported that the acetaldehyde content in Korean rice wine is 4.99–11.57  $\mu\text{g/mL}$ . Peng et al. (2017) studied the total aldehyde concentrations in cooking oil fumes produced in a typical kitchen by different cooking oils (palm oil, rapeseed oil, sunflower oil, and soybean oil), cooking methods (stir frying, pan frying, and deep frying), and food types (potato and pork loin). Deep-frying showed the highest total aldehyde emissions among cooking methods, followed by pan-frying and stir-frying. Sunflower oil produced the highest levels of total aldehydes, regardless of cooking method and food type, as compared to rapeseed oil and palm oil, which produced relatively lower aldehyde emissions. However, the levels of individual aldehydes produced during cooking were not evaluated. In addition, information on changes in toxic aldehyde contents in food during household cooking processes, such as boiling, pan-frying, and stir-frying, is scarce.

The major mechanism underlying acetaldehyde formation in alcoholic beverages is the oxidation of ethanol while glucose metabolism can

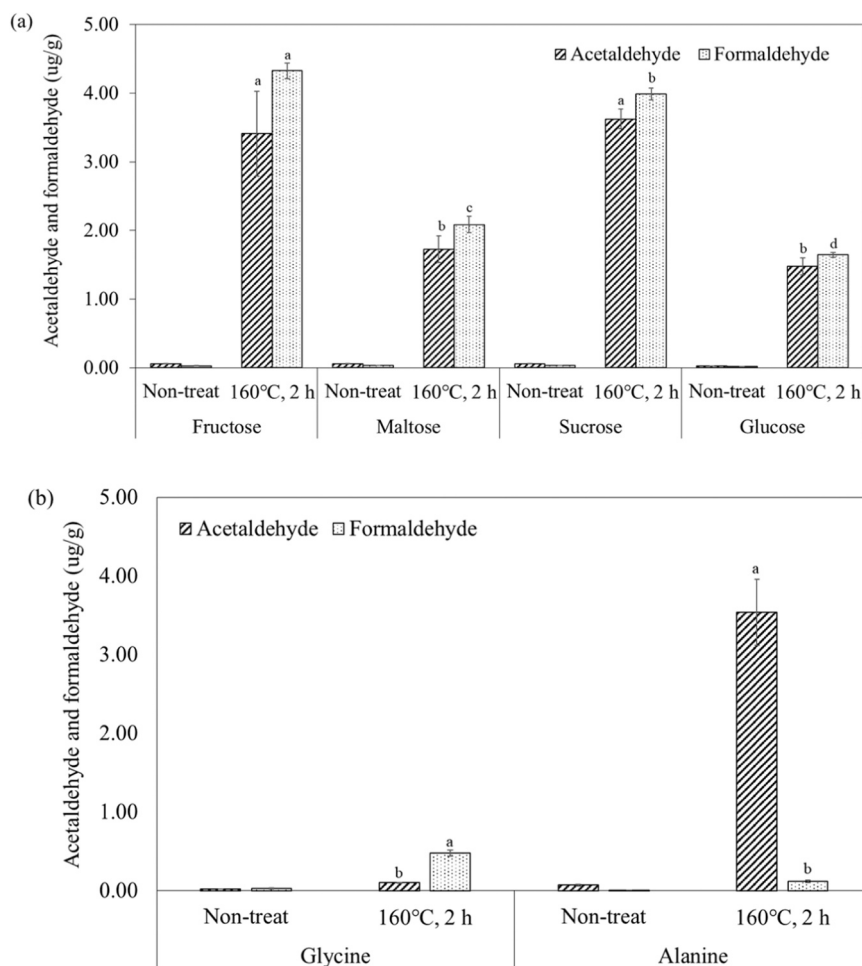


Fig. 3. Formation of acetaldehyde and formaldehyde from mono- and di-saccharides (a) and alanine or glycine (b). Different letters on the bar are significant at 0.05.

generate acetaldehyde in the presence of phenolics by lactic acid bacteria (Liu and Pilone, 2000). The quick volatilization of ethanol in foods by cooking process may prevent substantial increases of acetaldehyde, which could apply to rice wine. In addition, thermal energy could induce formation of acetaldehyde through Maillard reaction of alanine by Strecker degradation (Cobb et al., 2015). As shown in Fig. 3, heat treatment can produce both aldehydes from amino acids in the absence of reducing sugar. Significant increases in acetaldehyde in canned pork and eggs could be due to the high protein content in the matrix.

Formaldehyde in alcoholic beverages can be formed due to the bacterial oxidation on methanol (Norliana et al., 2009). Because formaldehyde can be formed by the Strecker degradation of amino acids like glycine (Velfšek et al., 1989) and the lipid oxidation of polyunsaturated fatty acids (European Food Safety Authority (EFSA), 2014), foods containing proteins or unsaturated fatty acids could generate more formaldehyde upon exposure to high thermal energy. Major formaldehyde formation in rapeseed oil could be due to lipid oxidation of linoleic acid, while formaldehyde formation in canned pork ham, could be explained by the Strecker degradation of glycine. Mono- and di-saccharides can generate formaldehyde and acetaldehyde when thermal energy was applied (Fig. 3). Pyrolysis of sugars can produce diverse aldehyde compounds including formaldehyde. Baker et al., (2006) detected formaldehyde in brown sugar, white sugar, fructose, and glucose between 220 and 550 °C thermal treatment. Therefore, presence of mono- and di-saccharides in foods may contribute the generation of aldehydes after cooking.

#### 4. Conclusion

The acetaldehyde and the formaldehyde content determined in foods after cooking was lower than established criteria. Depending on the type of food, slightly different effects of the cooking process on the acetaldehyde and formaldehyde contents were observed. Boiling decreased both aldehydes in beef rib and edible oil, while boiling increased acetaldehyde in egg. Rapeseed oil subjected to pan- and stir-frying contained more formaldehyde than that in uncooked samples. Foods containing oils with a high degree of unsaturation, high content of monosaccharides and disaccharides, and proteins containing glycine and alanine may generate aldehydes upon exposure to cooking with high thermal energy. Although the level of acetaldehyde and formaldehyde in foods after cooking was relatively low, adjusting the cooking methods or conditions can help to reduce the level of those aldehydes in foods.

#### CRediT authorship contribution statement

**SeHyeok Kim:** Validated the analysis methods for aldehydes. **HyunJeong Jung:** Designed experiment and analyzed aldehydes in foods. **KeunCheol Yoo:** Analyzed aldehydes in foods and participate revision process. **JaeHwan Lee:** Mainly wrote original and revised manuscript.

#### 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.

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