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# Levels of Perfluorinated Compounds in Liquid Milk Products in Korea

Na-youn Park[1], Younglim Kho[1], Junghoan Kim[2]*

[1]*Department of Health Environment Safety, Eulji University, Seongnam, Korea*
[2]*Food Technology & Service, Eulji University, Seongnam, Korea*

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**ABSTRACT -** This study was carried out to monitor the level of 17 perfluorinated compounds (PFCs) present in liquid milk products sold on the Korean market. The liquid milk samples were extracted via liquid-liquid extraction and analyzed by LC-MS/MS. Excellent linearity over the calibration range ($r^2$>0.99), and the limit of quantification of perfluorooctane sulfonate (PFOS) was 0.021 ng/g, and perfluorotetradecanoic acid (PFOA) was 0.057 ng/g. The accuracy was in the range of 72.5-115.3%, and the precision was under 20%. The preprocessing method for this experiment is considered appropriate for analysis of milk samples. The proposed analytical method was applied for the determination of PFCs in 98 liquid milk product samples, and the average content of total PFCs was 0.6576 ng/mL. PFOA and PFOS were detected in most samples, and their levels were less than 0.1 ng/mL, which was lower than those in other studies.

**Key words :** Perfluorinated compounds, Milk, PFOA, PFOS, LC-MS/MS

Perfluorinated compounds (PFCs), one of the persistent organic pollutants (POPs), are widely used as industrial surfactants, emulsifier, lubricants, fire-fighting forms, textiles, carpets, and clothing, grease and dirt repellents for more than 50 years, and are also widely applicable to fast food packaging and non-stick cookware[1-6].

It is emphasized that PFCs are remaining a human health hazard without being decomposed in the environment and are accumulated to the human through soil, water, air and food materials[1,2,7-9], and have been detected in human serum, plasma and breast milk[10-15]. The most commonly used PFCs are perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS), which are not destroyed in most environments. The half-life in humans is 3.8 years for PFOA and 5.4 years for PFOS. It is critical to understand the half-life of a compound, because the shorter the carbon chain length, the shorter the half-life, which may relate to the dangerousness of a material to human health[16-18].

The exposure of PFCs in human has been caused negative effects on fetal development, pregnancy, fertility, thyroid hormone levels, immune system, and carcinogenic potency[6,7,11,12]. International agency for research on cancer has classified perfluorotetradecanoic acid (PFOA) as 2B group (possibly carcinogenic to humans) in 2017[19]. Thus, EU and United States Environmental Protection Agency (EPA) reported about harmfulness of PFCs and put in the efforts to reduce the usage of PFCs. The 3M Company, major manufacturer of PFCs, stopped the production of PFOA and PFOS in 2000 and 2002[6,7,20].

In recent, the levels of PFCs in various foodstuffs have been reported in Canada, Spain, America and other countries[3,5,21-23]. The main route of PFCs exposure to human is dietary, particularly high protein foods due to perfluoroalkylated substances bind to proteins[13,24] and PFOS levels were positively associated ($P$<0.05) with intake of red meat, animal fats, and snacks[25]. PFCs has also been detected in milk and dairy products[11,13,21,26,27], Macheka et al.[28] reported that the levels of PFAS were influenced by the milk collection and other manufacturing process of milk products, preparation, manufacturing and packaging processes.

Although milk and dairy products are widely used as raw materials to be added to various foods as well as directly consumed, however, it is less studied and reviewed than the studies related to aquatics and water about PFCs in Korea. Therefore, this study was conducted to investigate the occurrence and levels of PFCs in market milk products distributed in Korea, and to confirm the food safety.

*Correspondence to: Junghoan Kim, Food Technology & Service, Eulji University, Seongnam, 13135, Korea
Tel: +82-31-740-7218, Fax: +82-31-740-7349
E-mail: peterkim@eulji.ac.kr

## Material and Methods

### Samples

The samples used for the research were purchased at large supermarkets located in Seoul and Seongnam-si in Korea between August and December 2013. The liquid milk products samples were classified and grouped by their manufacturing process and the type of food labelled on the package based on the Korean Food Code.

### Reagents

The 17 PFCs included perfluorpentanoic acid (PFPeA), perfluorhexanoic acid (PFHxA), perfluorheptanoic acid (PFHpA), perfluoroctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), per-fluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorobutane sulfonate (PFBS), per-fluoro-hexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS), perfluorodecane sulfonate (PFDS) perfluorooctane sulfonamide (PFOSA), 2-N-methyl-perfluorooctane sulfonamido acetic acid (MePFOSAA), and 2-N-ethyl-perfluorooctane sulfonamido acetic acid (EtPFOSAA). The internal standards included $^{13}C_4$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_4$-PFNA, $^{13}C_4$-PFDA, $^{13}C_4$-PFUnDA, $^{13}C_4$-PFDoDA, $^{13}C_4$-PFHxS, $^{13}C_4$-PFOS, $d_3$-N-MeFOSAA and $d_5$-N-EtFOSAA. All standards and internal standards were purchased from Wellington Laboratories (Guelph, ON, Canada). Protease from *Streptomyces griseus* (≥3.5 units/mg solid, powder) and lipase from *Candida rugosa* (≥700 units /mg solid) were purchased from Sigma-Aldrich (St. Louis, MO, USA). All solvents (acetonitrile, methanol, water and methyl -*t*-butyl ether (MTBE)) used in this study were of chro-matographic grade and were purchased from Burdick and Jackson (Muskegon, MI, USA).

### Pretreatment of sample

In this study, the analytical method followed on Kang et al.[11]. The analysis method is as follows: The 20 μL of internal standard was added into 1 mL of milk samples. Then, 300 μL of two enzyme solutions (Protease and lipase) were added and reacted at 37°C for 16 hours. After the enzyme reaction, reacted with 2 mL of 5 N $H_2SO_4$ in rotator for 30 min. The PFCs in milk sample were extracted by liquid–liquid extraction (LLE) with 5 mL of MTBE. Finally, the extracted solution was dried use of a vacuum-concentrator and reconstituted in 200 μL of acetonitrile.

### Instrumentation and Analytical conditions

The quantitative analysis was performed by liquid chromatography (Agilent 1100 Series, Agilent Technologies Inc., Santa Clara, CA, USA) tandem mass spectrometry (API 4000, Applied Biosystems, Foster City, CA, USA). The $C_{18}$ column (YMC-Pack ODS-AQ, 2.0×150 mm, 3.0 μm) was used for peak separation. Injection volume was 3 μL and the flow rate was 200 μL/min. The mass spectrometer was operated in negative mode. The conditions of the

**Table 1.** Instrumental condition of LC-MS/MS

| Parameter | Condition |
|---|---|
| Column | YMC-Pack ODS-AQ, 2.0×150 mm, 3.0 μm |
| Mobile phase | A : 5 mM ammonium acetate in water<br>B : methanol |
| Gradient mode | Time: 0, 5, 10, 10.1, and 22<br>B (%): 30, 100, 100, 30, and 30 |
| Flow rate | 200 μL/min |
| Injection volume | 3 μL |
| Ionization mode | ESI / negative |
| Curtain Gas | 25 psi |
| Gas temperature | 400°C |
| Ion spray voltage | -4500 V |
| Ion source gas 1 | 40 psi |
| Ion source gas 2 | 60 psi |
| Collision gas (CAD) | 7 |

**Table 2.** Parameters for analysis of the PFCs compounds

| Compound | Q1 (m/z) | Q3 (m/z) | DP | CE | EP | CXP |
|---|---|---|---|---|---|---|
| PFHxA | 313.0 | 268.9 | -35 | -12 | -10 | -7 |
| PFHpA | 363.0 | 318.9 | -35 | -14 | -10 | -7 |
| PFOA | 413.0 | 368.9 | -40 | -16 | -10 | -9 |
| PFNA | 463.0 | 418.8 | -45 | -14 | -10 | -11 |
| PFDA | 513.0 | 468.9 | -45 | -16 | -10 | -13 |
| PFUnDA | 562.9 | 518.9 | -40 | -18 | -10 | -13 |
| PFDoDA | 613.0 | 569.0 | -35 | -18 | -10 | -15 |
| PFHxS | 399.0 | 79.9 | -95 | -78 | -10 | -5 |
| PFOS | 499.0 | 80.1 | -105 | -92 | -10 | -5 |
| PFBS | 298.9 | 79.8 | -70 | -60 | -10 | -3 |
| PFPeA | 263.0 | 218.9 | -25 | -12 | -10 | -13 |
| PFTrDA | 663.1 | 619.1 | -40 | -16 | -10 | -9 |
| PFTeDA | 713.1 | 669.1 | -50 | -20 | -10 | -9 |
| PFDS | 599.0 | 79.9 | -55 | -120 | -10 | -15 |
| N-MeFOSAA | 570.0 | 418.8 | -80 | -28 | -10 | -9 |
| N-EtFOSAA | 584.0 | 419.0 | -60 | -30 | -10 | -9 |
| PFOSA | 498.0 | 78.0 | -85 | -70 | -10 | -3 |

DP: declustering potential.
CE: collision energy.
EP: entrance potential.
CXP: collision cell exit potential.

instrument and optimized mass parameters for each target material are detailed in Table 1 and 2.

### Validation

Linearity, accuracy, precision and limit of detection (LOD) were validated by pooled milk products sample. Calibration curves of PFCs with 7 concentration points, 0.02-10 ng/mL, were calculated, LOD were estimated using a slope and standard error of regression equation[29]. The calculation formula was as follows; LOD=3.3 x standard error of regression equation / Slope, LOQ= 3 x LOD

## Results and discussion

The coefficient of the determination of a calibration curve in this study significantly showed linearity with higher and equal to 0.99 in 17 PFCs measured, as shown as Table 3. The limit of detection (LOD) of PFOS was lowest, with 0.007 ng/g, and the highest was PFTeDA, as 0.057 ng/g. Compared with other studies, Wang et al.[30] reported that LOD of 11 PFCs were 2(PFHxS)–27(PFNS) pg/g in milk, milk powder and yogurt in China, method detection limit of PFCs in cow milk in Italy was 0.5(PFTeDA)-3.0(PFDA and PFPeA) ng/mL[26].

The result of the experiment for validation of the analytical method employed in the determination of PFCs using the pooled samples made by mixing milks under the concentration of 0.1, 0.5 and 2 ng/mL can be seen in Table 4, except PFPeA of 2 ng/mL, the accuracy of the analysis of PFCs in milk was in range of 72.5-115.3%, and the precision was under 20%. Therefore it is considered that the analytical method employed for this study was appropriate for determination of PFCs in milk samples, and we could analyze the 17 PFCs simultaneously more than previous researches[26,30].

The 16 PFCs except PFTeDA were detected in milk products samples. The highest content of total PFCs was found in liquid milk as 0.9037 ng/mL, whereas the liquid milk samples were higher than those of other groups with 0.5237(milk beverage)-0.6368(fortified milk) ng/mL. PFPeA, PFDS and PFHxA remained at a relatively high concentration of 0.1 ng/mL or more in liquid milk. PFDoDA, PFTrDA and PFTeDA were not detected in low fat milk, and in processed milk and fortified milk, there were no detection of PFNA, PFDoDA, PFTrDA and PFTeDA. PFNA were found in liquid milk as low concentration of 0.0250 ng/mL, PFCs seemed to be reduced in the manufacturing process but not statistically[28], PFPeA also showed drastic decrease in low fat and process milk. The average concentration of the total PFC for all of the samples was 0.6576 ng/mL, and the levels of the total PFCs in other milk samples grouped by the Korean Food Sanitation Act are shown in Table 5. In the China, Yu et al.[27] analyzed 20 PFCs in 46 milk samples. As a result, there were 16 kinds of PFCs that were detected in 46 milk samples, ND (not detected) to 0.11 μg/L for PFOA, ND to 0.12 μg/L for PFOS, ND to 0.34 μg/L for PFBA 0.19 to 0.66 μg/L for total PFCs. In another study, in a review of 11 PFCs in milk, milk powder and yogurt in the China, the concentration of PFOA and PFOS were <18 to 178 pg/g and <5 to 695 pg/g in milk, <36 to 482 pg/g and <10 to 175 pg/g in milk powder and <18 to 229 pg/g and <5 to 32 pg/g in yoghurt[28]. In the study in Italy, the contamination monitoring of PFCs was shown to have progressed in cow's milk. In that region, PFOA and PFOS were detected in 27 and 29 out of 67 samples and the maximum concentrations were 32 ng/L for PFOA and 97 ng/L for PFOS[21]. Compared with the research conducted in China and Italy, the residual pattern of PFOA and PFOS in market milks in Korean were shown to have been similar, and the concentrations of them were less than 0.1 ng/mL.

The relationship of the fat content and PFCs in food, PFOS was strongly displaced from the aqueous phase to the creamed phase, while PFOA stayed preferentially located in water[31], PFOA concentrations in low-fat processed milk

**Table 3.** Linearity (standard error, regression coefficient, slope), limit of detection (LOD) and limit of quantification (LOQ) for the analysis of PFCs in milk

| Compounds | Standard error | Slope | $r^2$ | LOD (ng/mL) | LOQ (ng/mL) |
|---|---|---|---|---|---|
| PFHxA | 0.0018 | 0.2066 | 0.9982 | 0.027 | 0.081 |
| PFHpA | 0.0043 | 0.6345 | 0.9990 | 0.02 | 0.060 |
| PFOA | 0.0038 | 0.6103 | 0.9991 | 0.019 | 0.057 |
| PFNA | 0.0046 | 0.612 | 0.9987 | 0.022 | 0.066 |
| PFDA | 0.0025 | 0.4615 | 0.9993 | 0.016 | 0.048 |
| PFUnDA | 0.0038 | 0.5512 | 0.9989 | 0.021 | 0.063 |
| PFDoDA | 0.0072 | 0.4801 | 0.9949 | 0.045 | 0.135 |
| PFHxS | 0.0103 | 1.6165 | 0.9991 | 0.019 | 0.057 |
| PFOS | 0.0009 | 0.4089 | 0.9999 | 0.007 | 0.021 |
| PFBS | 0.0175 | 1.4359 | 0.9992 | 0.037 | 0.111 |
| PFPeA | 0.0153 | 1.9436 | 0.9986 | 0.024 | 0.072 |
| PFTrDA | 0.0045 | 0.2657 | 0.9936 | 0.050 | 0.150 |
| PFTeDA | 0.0057 | 0.2979 | 0.9917 | 0.057 | 0.171 |
| PFDS | 0.0022 | 0.4873 | 0.9995 | 0.014 | 0.042 |
| N-MePFOSAA | 0.0012 | 0.2583 | 0.9995 | 0.014 | 0.042 |
| N-EtPFOSAA | 0.0013 | 0.2861 | 0.9995 | 0.011 | 0.033 |
| PFOSA | 0.0009 | 0.2629 | 0.9997 | 0.014 | 0.042 |

Standard error: The standard error of the y value predicted for each x in the regression analysis.

**Table 4.** Accuracy and precision of the target PFCs from milk

| Compounds | Accuracy (%) | | | Precision | | |
|---|---|---|---|---|---|---|
| | Low (0.1 ng/mL) | Medium (0.5 ng/mL) | High (2.0 ng/mL) | Low (0.1 ng/mL) | Medium (0.5 ng/mL) | High (2.0 ng/mL) |
| PFHxA | 94.5 | 96.8 | 99.1 | 13.9 | 6.9 | 11.3 |
| PFHpA | 103.6 | 90.0 | 93.3 | 9.0 | 10.2 | 12.2 |
| PFOA | 101.1 | 93.4 | 93.0 | 9.9 | 1.7 | 5.9 |
| PFNA | 99.1 | 91.3 | 91.5 | 3.4 | 6.0 | 6.6 |
| PFDA | 103.3 | 103.2 | 103.2 | 7.9 | 5.4 | 5.6 |
| PFUnDA | 103.2 | 98.4 | 98.4 | 4.1 | 4.6 | 6.3 |
| PFDoDA | 102.3 | 99.8 | 99.3 | 3.3 | 6.5 | 7.9 |
| PFHxS | 97.5 | 91.7 | 89.9 | 4.3 | 5.4 | 7.0 |
| PFOS | 101.2 | 90.9 | 94.9 | 11.0 | 7.6 | 5.1 |
| PFBS | 85.3 | 106.0 | 104.7 | 17.7 | 14.4 | 12.9 |
| PFPeA | 86.1 | 83.0 | 72.5 | 11.7 | 14.7 | 18.0 |
| PFTrDA | 109.3 | 106.2 | 110.9 | 5.3 | 9.6 | 9.8 |
| PFTeDA | 94.3 | 92.5 | 94.5 | 4.7 | 7.3 | 11.7 |
| PFDS | 100.5 | 95.7 | 91.4 | 5.9 | 8.0 | 4.3 |
| N-EtPFOSAA | 112.6 | 101.5 | 101.1 | 4.0 | 9.4 | 8.3 |
| N-MePFOSAA | 105.0 | 102.1 | 101.9 | 5.1 | 6.8 | 4.9 |
| PFOSA | 109.9 | 110.6 | 115.3 | 12.5 | 14.6 | 10.2 |

**Table 5.** The concentration ± standard deviation of PFCs by milk product type (Unit: ng/mL)

| Types of products | Milk beverage | Fortified milk | Processed milk | Low-fat processed milk | Low fat milk | Liquid milk | Total sample average |
|---|---|---|---|---|---|---|---|
| Number of samples | 9 | 6 | 11 | 30 | 8 | 34 | 98 |
| PFHxA | 0.0702±0.0597 | 0.0441±0.0319 | 0.0618±0.0372 | 0.0652±0.0446 | 0.0771±0.0416 | 0.1074±0.1322 | 0.071±0.087 |
| PFHpA | 0.0395±0.0335 | 0.0407±0.0247 | 0.0334±0.0126 | 0.0396±0.0234 | 0.0403±0.0184 | 0.0501±0.0477 | 0.0406±0.0337 |
| PFOA | 0.0722±0.0615 | 0.0455±0.0352 | 0.0763±0.0302 | 0.0673±0.0378 | 0.0807±0.0228 | 0.0875±0.0822 | 0.0716±0.0580 |
| PFNA | 0.0148±0.0112 | <LOD | <LOD | 0.0133±0.0070 | 0.0131±0.0059 | 0.0251±0.0341 | 0.0147±0.0214 |
| PFDA | 0.0139±0.0122 | 0.0098±0.0044 | 0.0111±0.0054 | 0.0135±0.0086 | 0.0144±0.0106 | 0.0193±0.0225 | 0.0137±0.0151 |
| PFUnDA | 0.0243±0.0134 | 0.0253±0.0140 | 0.0249±0.0121 | 0.0268±0.0149 | 0.0208±0.0089 | 0.0289±0.0279 | 0.0252±0.0195 |
| PFDoDA | 0.0252±0.0081 | <LOD | <LOD | <LOD | <LOD | 0.0255±0.0102 | 0.0235±0.0066 |
| L-PFHxS | 0.0151±0.0090 | 0.0220±0.0112 | 0.0142±0.0067 | 0.0172±0.0097 | 0.0239±0.0106 | 0.0219±0.0140 | 0.0191±0.0115 |
| L-PFOS | 0.0154±0.0116 | 0.0318±0.0357 | 0.0351±0.0268 | 0.0311±0.0442 | 0.0185±0.0159 | 0.0296±0.0293 | 0.0269±0.0329 |
| L-PFBS | 0.0292±0.0214 | 0.0249±0.0155 | 0.0319±0.0233 | 0.0499±0.0347 | 0.0436±0.0321 | 0.0515±0.0705 | 0.0385±0.0482 |
| PFPeA | 0.0442±0.0435 | 0.0744±0.0655 | 0.0860±0.0925 | 0.0806±0.0607 | 0.0663±0.0553 | 0.2215±0.2562 | 0.0955±0.1732 |
| PFTrDA | <LOD | <LOD | <LOD | <LOD | <LOD | 0.0274±0.0100 | 0.0254±0.0059 |
| PFTeDA | <LOD | <LOD | <LOD | <LOD | <LOD | <LOD | <LOD |
| PFDS | 0.0619±0.0391 | 0.0902±0.0644 | 0.0876±0.0592 | 0.0684±0.0519 | 0.0948±0.0369 | 0.1279±0.1098 | 0.0885±0.0801 |
| N-EtFOSAA | <LOD | 0.0423±0.0527 | 0.0084±0.0051 | 0.0245±0.0449 | 0.0174±0.0091 | 0.0154±0.0171 | 0.0189±0.0305 |
| N-MeFOSAA | 0.0278±0.0148 | 0.078±0.0691 | 0.0404±0.0177 | 0.0467±0.0467 | 0.0256±0.0070 | 0.0251±0.0186 | 0.0406±0.0357 |
| PFOSA | 0.0115±0.0068 | 0.0213±0.0089 | 0.0184±0.0077 | 0.0167±0.0061 | 0.0171±0.0087 | 0.0119±0.0070 | 0.0162±0.0075 |
| Total PFCs | 0.5238±0.2016 | 0.6369±0.2715 | 0.6161±0.2037 | 0.6361±0.2497 | 0.6291±0.1739 | 0.9038±0.5923 | 0.6576±0.4142 |

[1)] Mean levels calculated that levels <LOD are equal to half the LOD.

(0.0672 ng/mL) and low-fat milk (0.0806 ng/mL) were expected to be high, but there was no difference compared to other groups (0.0455~0.0874 ng/mL) as shown in Table 5.

In this study, the 98 market milks samples were classified into six types according to their manufacturing process and type of food labelled on package, and as a result, PFCs are seen to be reduced through various processing steps. The results were similar to those which were reported by Shin et al.[32] that the concentrations of the residual PFCs in food were lower as when the food is processed. Additionally, PFCs with short carbon chain were significantly reduced or not detected. This means that the importance of PFOS and PFOA in food safety.

## 국문요약

본 연구는 국내 유통되는 액상 유제품에 함유된 17종의 과불화화합물 (PFCs) 함량에 대한 모니터링을 위해 수행되었다. 샘플을 액체-액체 추출을 통해 추출하고 LC-MS/MS로 정량 분석하였다. 시중에 유통중인 98개의 액상 유제품 샘플에서 PFCs의 모니터링을 진행하였으며 그 결과, PFTeDA를 제외한 16종의 PFCs가 검출되었다. 총 PFCs의 함량은 0.9037 ng/mL로 액상 우유에서 가장 높았으며, PFPeA, PFDS, PFHxA도 액상우유에서 0.1 ng/mL 이상의 농도 수준을 보였다. 미국과 이탈리아에서 수행한 연구와 비교했을 때 PFOA와 PFOS의 잔류 패턴은 유사한 것으로 나타났으며, 농도 수준은 0.1 ng/mL 미만이었다. 액상 유제품은 다양한 가공단계(살균, 혼합, 지방제거 등)를 거쳐 총 PFCs의 함량이 감소하는 것으로 보여 지며, 특히 짧은 탄소 사슬을 가진 PFCs가 크게 감소하거나 검출되지 않았다. 이는 식품 안전에서 원료의 PFOS와 PFOA의 잔류정도가 중요하다는 것을 의미한다.

## Conflict of interests

The authors declare no potential conflict of interest.

## ORCID

Na-youn Park https://orcid.org/0000-0002-4164-9522
Younglim Kho https://orcid.org/0000-0002-2590-4722
Junghoan Kim https://orcid.org/0000-0002-0162-6326

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