Journal of Dairy Research

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Perfluorinated compounds in milk in Shanxi Province, China: A three-year consecutive study

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Research Communication

Cite this article: Dang DX, Xu SQ and Wang H (2025) Perfluorinated compounds in milk in Shanxi Province, China: A three-year consecutive study. *Journal of Dairy Research* **92**, 276–279. https://doi.org/10.1017/S0022029925101167

Received: 8 October 2024
Revised: 15 February 2025
Accepted: 26 February 2025
First published online: 20 October 2025

Keywords:
agricultural products; food safety; milk contamination; perfluorinated compounds exposure

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Abstract

Perfluorinated compounds (PFCs) are synthetic chemicals commonly used in various industries for their water-, grease-, and stain-repellent properties. These compounds are highly persistent in the environment and can be absorbed by farm animals, subsequently contaminating animal-derived products. This contamination poses a significant health risk to humans who consume these products. Previous studies have identified cow's milk as one of the primary animal products contaminated with PFCs. However, it remains unclear which specific PFCs increase in concentrations over time. In this study, we analysed data on the concentrations of 24 PFCs in cow's milk sourced from a milk processing plant in Taiyuan, Shanxi Province, China, over a three-year period, as provided by the National Agriculture Science Data Centre. Our analysis revealed that perfluoropentanoic acid (PFPeA) and perfluorobutanoic acid (PFBA) were the dominant PFCs that tended to accumulate in cow's milk over time. Consequently, consumers and milk producers should monitor the levels of PFPeA and PFBA in cow's milk to mitigate potential health risks associated with these pollutants.

Perfluorinated compounds (PFCs) are a group of synthetic chemicals commonly used in products such as non-stick cookware, food packaging, textiles, and firefighting foams due to their water-, grease-, and stain-repellent properties (Barbo *et al.*, 2023). PFCs contains strong carbon-fluorine bonds, which make them highly resistant to degradation (Wee and Aris, 2023). PFCs have been associated with various toxicological effects, including liver damage, hormonal disruption, immune system suppression, cancer, developmental issues, elevated cholesterol, and kidney damage (Cai *et al.* 2021). Notably, PFCs are bioaccumulative, meaning they can accumulate in organisms and move up the food chain (Savoca and Pace, 2021).

Animal-derived products are a primary source of nutrition for humans, providing essential nutrients. With the development of liquid chromatography-mass spectrometry, the presence of PFCs has been detected in various animal products, including fish, eggs, pork, beef, and milk (Vicente *et al.*, 2012; Wang *et al.*, 2021; Park *et al.*, 2021; Barbo *et al.*, 2023). In the case of milk, Wang *et al.* (2010) detected perfluoroheptanoic acid (PFHpA) in over half of the cow's milk samples collected from markets in China. However, Tao *et al.* (2008) did not detect PFCs contamination in milk obtained from the market in the USA. More recently, Park *et al.* (2021) found that PFHpA, perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), and perfluoropentanoic acid (PFPeA) were the predominant PFCs pollutants in milk products sold on the Korean market.

The accumulation of PFCs in milk poses significant health risks, potentially leading to developmental issues, hormonal imbalances, and immune system dysfunction (Qian and Ying, 2021). For consumers of milk, exposure to PFCs can present serious health hazards. Given the bioaccumulative properties of these contaminants, understanding their time-dependent accumulation is crucial for identifying which specific PFCs warrant greater attention. However, it remains unclear which PFCs increase in concentration over time. Therefore, this study aims to identify which specific PFCs accumulate in milk over time. We hypothesize that the concentrations of all 24 measured PFCs will exhibit a gradual increase over the study period.

Materials and methods

Data collection

Data for this study were obtained from the National Agriculture Science Data Centre (http://www.agridata.cn/#/home; NASDC), uploaded by the Shenzhen Institute of Inspection

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Table 1. Comparative analysis of the concentrations of 24 perfluorinated compounds in milk samples from taiyuan, collected at consistent timepoints over three consecutive years

	Years				*P*-value	
Levels, μg/kg × 10^{-5}	1st	2nd	3rd	SEM	Linear	Quadratic
PFPeA	6.55[b]	28.25[ab]	200.85[a]	63.88	0.037	0.340
PFHxA	7.49[b]	49.19[ab]	147.74[a]	52.18	0.064	0.659
PFHpA	9.33[b]	28.33[ab]	140.15[a]	51.37	0.079	0.465
PFOA	19.40	258.30	301.70	116.77	0.095	0.498
PF0	7.35	2.45	13.17	6.12	0.505	0.304
ipPF0	0.00	0.00	0.00	–	–	–
PFDA	10.86	10.03	18.55	8.86	0.542	0.669
PFUdA	4.95	13.43	10.42	6.43	0.551	0.470
PFDoA	2.10	22.15	25.70	9.74	0.094	0.493
PFTrDA	3.42	11.79	11.32	5.85	0.345	0.540
PFTeDA	5.60	3.34	0.00	2.99	0.193	0.884
PFHxDA	3.74	0.00	2.37	2.56	0.707	0.335
PFBS	81.73	39.07	108.53	46.20	0.684	0.327
PFPeS	39.13	45.80	7.67	27.26	0.419	0.506
PFHxS	39.87	32.27	25.87	23.22	0.672	0.983
PFHpS	23.73	32.27	35.07	25.18	0.752	0.926
P1MHpS	0.00	0.00	0.00	–	–	–
PFOS	100.00[b]	289.10[ab]	459.40[a]	131.52	0.060	0.954
PFNS	0.00	26.47	34.00	21.51	0.270	0.721
ipPFNS	0.00	0.00	0.00	–	–	–
PFDS	45.80	14.80	16.73	23.13	0.379	0.564
PFDoS	0.00	0.00	0.00	–	–	–
PFBA	15.50[b]	26.67[b]	134.87[a]	36.03	0.024	0.278
PFODA	0.00	0.00	0.00	–	–	–

Abbreviation: PFPeA, perfluoropentanoic acid; PFHxA, perfluorohexanoic acid, PFHpA, perfluoroheptanoic acid; PFOA, perfluorooctanoic acid; PF0, perfluoroacetic acid; ipPF0, isopropyl perfluoroacetic acid; PFDA, perfluorodecanoic acid; PFUdA, perfluoroundecanoic acid; PFDoA, perfluorododecanoic acid; PFTrDA, perfluorotridecanoic acid; PFTeDA, perfluorotetradecanoic acid; PFHxDA, perfluorohexadecanoic acid; PFBS, perfluorobutane sulfonic acid; PFPeS, perfluoropentane sulfonic acid; PFHxS, perfluorohexane sulfonic acid; PFHpS, perfluoroheptane sulfonic acid; P1MHpS, perfluorinated 1-methylheptane sulfonic acid; PFOS, perfluorooctane sulfonic acid; PFNS, perfluorononane sulfonic acid; ipPFNS, isopropyl perfluorononane sulfonic acid; PFDS, perfluorodecane sulfonic acid; PFDoS, perfluorododecane sulfonic acid; PFBA, perfluorobutanoic acid; PFODA, perfluorooctadecanoic acid; SEM, standard error of the mean.
[a,b]Means in the same row with different superscript differ significantly ($P < 0.05$).

and Quarantine (http://www.agridata.cn/data.html#/datadetail?id=4419; Yue, 2020). The NASDC is a publicly accessible data repository, where the collected data is open to the public and available for use by anyone who adheres to the data usage guidelines.

Milk samples were collected from a milk processing plant in Taiyuan, Shanxi Province, China, in July 2015 (n = 15), 2016 (n = 15), and 2017 (n = 15). All samples were analysed using HPLC-MS/MS methods (with Zorbax EP-C18 and Poroshell 120 EC-C18 columns) at the Xiamen Entry-Exit Inspection and Quarantine Bureau Technical Centre to understand the concentration of 24 PFCs, including PFPeA, PFHxA, PFHpA, PFOA, perfluoroacetic acid (PF0), isopropyl perfluoroacetic acid (ipPF0), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUdA), perfluorododecanoic acid (PFDoA), perfluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorohexadecanoic acid (PFHxDA), perfluorobutane sulfonic acid (PFBS), perfluoropentane sulfonic acid (PFPeS), perfluorohexane sulfonic acid (PFHxS), perfluoroheptane sulfonic acid (PFHpS), perfluorinated 1-methylheptane sulfonic acid (P1MHpS), perfluorooctane sulfonic acid (PFOS), perfluorononane sulfonic acid (PFNS), isopropyl perfluorononane sulfonic acid (ipPFNS), perfluorodecane sulfonic acid (PFDS), perfluorododecane sulfonic acid (PFDoS), perfluorobutanoic acid (PFBA), and perfluorooctadecanoic acid (PFODA) in cow's milk samples.

Statistical analysis

All data were analysed statistically using a randomised complete block design, employing the General Linear Model procedure (SAS Inst. Inc., Cary, NC, USA). Each sample was treated as the experimental unit. Orthogonal contrasts were applied to evaluate both linear and quadratic effects in relation to the years. Tukey's test was used to identify differences between groups. Data variability is presented as the pooled standard error of means. Treatment means

https://doi.org/10.1017/S0022029925101167 Published online by Cambridge University Press

were considered significantly different at $P < 0.05$, and trends were noted at $P < 0.10$.

Results and discussion

In this study, we investigated 24 PFCs in cow's milk samples, collected at consistent timepoints over three years. We found that PFPeA ($P = 0.037$) and PFBA ($P = 0.024$) significantly increased over time. Additionally, PFHxA ($P = 0.064$), PFHpA ($P = 0.079$), PFOA ($P = 0.095$), PFOS ($P = 0.060$), and PFDoA ($P = 0.094$) exhibited increasing trends (Table 1). Tukey's test revealed that the concentrations of PFPeA, PFHxA, PFHpA, and PFOS in the third year were significantly higher than those in the first year ($P < 0.05$). Moreover, PFBA levels in the third year were significantly higher than in the other two years ($P < 0.05$). However, ipPF0, P1MHpS, ipPFNS, PFDoS, and PFODA were not detected in any of the years, suggesting these compounds may not contribute to the contamination in the milk samples. Therefore, our fundings indicate that PFPeA, PFBA, PFHxA, PFHpA, PFOA, PFOS, and PFDoA were the main contaminants in the milk, with PFPeA and PFBA being particularly prominent. Similarly, Wang *et al.* (2010) reported PFHpA as a common pollutant in milk products sold on the Chinese market, with detection in over half of the milk samples. Park *et al.* (2021) also identified PFHxA, PFHpA, PFOA, and PFPeA in liquid milk products sold on the Korean market.

PFHpA and PFBA have been shown to bioaccumulate in the human body. Adverse health effects linked to PFHpA exposure include reproductive toxicity, endocrine disruption, liver and kidney damage, obesity, diabetes, high cholesterol, cancer, and immune system impairment (HEAL, 2022). Additionally, human epidemiological studies have found potential associations between PFBA exposure and health issues such as liver and thyroid toxicity, as well as developmental delays in infants (Davis *et al.*, 2022).

The high mobility of PFPeA and PFBA in water allows them to persist and travel long distances, contaminating aquatic environments far from their original emission sources. These compounds have even been detected in remote regions such as the Arctic and Antarctic, potentially causing irreversible harm to both the environment and human health (Gao *et al.*, 2020). Our findings suggest that PFPeA and PFBA are the dominant PFCs that tend to accumulate in cow's milk over time.

Conclusion

This study highlights the accumulation of PFCs in cow's milk over time, with PFPeA and PFBA identified as the most prominent contaminants. Our results suggest that these two compounds tend to increase in concentration over the study period, posing potential health risks to consumers due to their bioaccumulative properties and association with various toxicological effects. The presence of other PFCs such as PFHxA, PFHpA, PFOA, PFOS, and PFDoA also showed increasing trends, underscoring the widespread contamination of milk products.

Limitation

While this study provides important insights into the accumulation of PFCs in cow's milk over time, several limitations should be considered in future research.

Firstly, potential confounding factors such as variations in milk processing methods, storage conditions, and milk composition across the three years may have influenced the detected concentrations of PFCs. These factors were not controlled for in the present study.

Additionally, environmental changes during the study period, including fluctuations in PFCs contamination levels in the surrounding environment or shifts in local industrial activities, could have affected the accumulation of these compounds in the milk samples.

Furthermore, uncertainties related to the sensitivity and precision of the HPLC-MS/MS technique, as well as potential sampling errors, may have impacted the values of the reported concentration.

Future research would benefit from optimizing these limitations.

Highlights

1. The study revealed a consistent presence of PFCs in cow's milk samples from a milk processing plant in Taiyuan, China, over a three-year period.
2. PFPeA and PFBA were the dominant PFCs that tended to accumulate in cow's milk over time.
3. The findings highlight the importance of continuous monitoring of PFCs concentrations in cow's milk to assess and mitigate exposure risks.

Acknowledgements. Acknowledgement for the data support from National Data Centre for Agricultural Scientific, National Science & Technology Infrastructure of China (http://www.agridata.cn)

Funding. This work was partly supported by the Doctoral start-up fund of Xiangyang Central Hospital (No. 2023BS07); Xiangyang Central Hospital Young Talent Incubation and Cultivation Project (2025RCQM-087).

Competing interests. The authors declare that they have no competing interests

Ethics approval and consent to participate. Not required

Availability of data and materials. Data supporting the findings of this study are available in the National Agriculture Science Data Centre (http://www.agridata.cn/#/home; NASDC) under the identifier CSTR:17,058.11.E0002.20201216.00.ds.0056

Authors' contributions. De Xin Dang: writing - original draft, conceptualization, methodology, validation, formal analysis, data curation; Shi Qi Xu: writing - review & editing, methodology, conceptualization; Huan Wang: conceptualization, writing - review & editing, supervision. All authors read and approved the final manuscript.

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