

Fluorinated Compounds in North American Cosmetics

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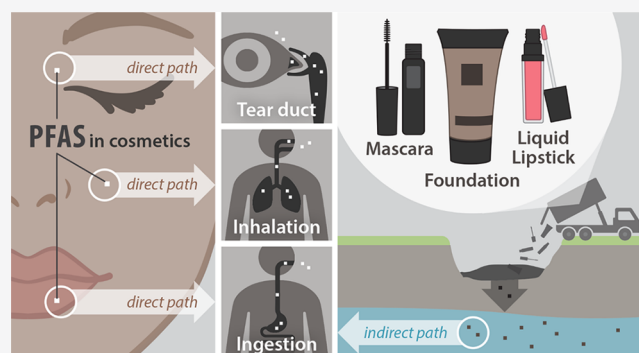
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Supporting Information

ABSTRACT: Per- and polyfluoroalkyl substances (PFAS), a highly persistent and potentially toxic class of chemicals, are added to cosmetics to increase their durability and water resistance. To assess this potential health and environmental risk, 231 cosmetic products purchased in the U.S. and Canada were screened for total fluorine using particle-induced gamma-ray emission spectroscopy. Of the eight categories tested, foundations, mascaras, and lip products had the highest proportion of products with high total fluorine $\geq 0.384 \mu\text{g F}/\text{cm}^2$. Twenty-nine products including 20 with high total fluorine concentrations were analyzed using targeted LC-MS/MS and GC-MS. PFAS concentrations ranged from 22–10,500 ng/g product weight, with an average and a median of 264 and 1050 ng/g product weights, respectively.

Here, 6:2 and 8:2 fluorotelomer compounds, including alcohols, methacrylates, and phosphate esters, were most commonly detected. These compounds are precursors to PFCAs that are known to be harmful. The ingredient lists of most products tested did not disclose the presence of fluorinated compounds exposing a gap in U.S. and Canadian labeling laws. The manufacture, use, and disposal of cosmetics containing PFAS are all potential opportunities for health and ecosystem harm. Given their direct exposure routes into people, better regulation is needed to limit the widespread use of PFAS in cosmetics.



INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) comprise a class of more than 4700 chemical compounds for which the characteristic perfluorinated carbon moiety confers hydrophobic chemical properties and environmental persistence. Since the 1950s, these compounds have been widely used in industrial products such as firefighting foams and consumer products such as coated fabrics, carpets, cookware and other food packaging, and many others.^{1–4} In 2019, the retail value of personal care products was estimated to be more than \$100 billion (USD) in North America, with approximately \$20 billion coming from cosmetics.⁵ While several studies have documented the use of PFAS in cosmetics from Europe and Asia, no such data exist in North America.^{6–8} PFAS in cosmetics may pose a risk to human health through direct and indirect exposure, as well as a risk to ecosystem health throughout the lifecycle of these products. PFAS are used in cosmetics due to their properties such hydrophobicity and film-forming ability, which are thought to increase product wear, durability, and spreadability. Additional claimed benefits are increased skin absorption of the product and improvements in the appearance or texture of skin.⁹

Gluge et al.⁹ recently noted that the magnitude of PFAS use in cosmetics in several European countries was difficult to estimate due to lax regulatory requirements for reporting PFAS

use. Requirements in the United States (U.S.) and Canada are similarly lax. Use and labeling of cosmetic ingredients in the U.S. is regulated by the Federal Food, Drug, and Cosmetics Act of 1938 and the Fair Packaging and Labeling Act of 1967. These acts do not regulate the type or kind of testing that is needed to determine the safety of cosmetic ingredients, and exemptions exist for labeling ingredients which are considered proprietary.¹⁰ In Canada, the Food and Drugs Act, the Cosmetic Regulations, and the Consumer Packaging and Labeling Act and Regulations regulate the labeling and safety of cosmetics.¹¹ These require all ingredients (except “incidental” ingredients such as processing agents) to be disclosed both to federal regulators and to consumers on product packaging.

Cosmetics industry-associated groups usually assess the potential hazards of new and emerging ingredients in North American cosmetics. Ingredient names are defined and

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538

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disclosed per the guidelines given in the International Cosmetic Ingredient Dictionary and Handbook.¹² This document is prepared by the International Cosmetic Ingredient Nomenclature Committee, which is a group sponsored by the Personal Care Products Council.¹³ This council includes more than 600 member companies that produce and distribute the “vast majority” of personal care products in the U.S.¹⁴ The Personal Care Product Council also funds the Cosmetic Ingredient Review (CIR) which seeks to assess the safety of cosmetic ingredients. Assessments made by CIR are subsequently utilized by regulatory bodies when determining the need for new or modified regulations. Without formal regulations that define how cosmetic ingredients should be labeled, the INCI Dictionary and Handbook is used by cosmetic manufacturers when determining how to label chemical ingredients. However, using this handbook for guidance to establish the presence of PFAS from ingredient labels is complicated by the series of exemptions and generalized guidelines for polymers, silanes and siloxanes, color additives, and substituted compounds.

Three studies have measured PFAS in a total of 72 cosmetics available in Sweden, Denmark, Japan, and the Republic of Korea between 2009–2017.^{6–8} These publications focused on the measurement of PFAS in cosmetics using LC-MS/MS as well as total fluorine through combustion ion chromatography (CIC). Cosmetics analyzed in these studies were those that disclosed a fluorinated ingredient in their labeling, including foundations, facial creams, concealers, and other personal care products such as sunscreens and shaving foams. Lip and eye products, two prominent classes of products in the North American cosmetics market, were surveyed at low frequencies in these previous studies, presumably because labeling did not indicate the presence of a fluorinated ingredient. Of concern is that these classes of cosmetics are applied close to the eyes and the mouth, which could increase exposure and hence risk due to enhanced absorption and ingestion.

Our goal was to determine the occurrence of PFAS in cosmetics in U.S. and Canada, especially mascaras and lipsticks, using particle-induced gamma-ray emission (PIGE) spectroscopy to rapidly screen total fluorine concentrations in 231 cosmetic products, followed by targeted analysis. PIGE has previously been utilized to measure total fluorine levels, the combination of inorganic and organic sources of fluorine in food packaging and firefighting textiles and foams.^{15–17} Targeted analysis of PFAS was conducted on a subset of products by means of liquid chromatography-tandem mass spectrometry (LC-MS/MS) and gas chromatographic mass spectrometry (GC-MS). Finally, we discuss the U.S. and Canadian regulatory regimes with respect to PFAS use in cosmetics by evaluating the relationship between PFAS presence and its disclosure, or lack thereof, on ingredient labels.

MATERIALS AND METHODS

A total of 231 cosmetics were purchased or obtained as free samples for analysis. U.S. cosmetics were purchased from retailers in Indiana and Michigan including Ulta Beauty, Sephora, Target, and Bed Bath & Beyond from 2016 to 2020. Canadian products were purchased online from Sephora and Shoppers Drug Mart in Toronto, ON, Canada in 2020. Canadian products included replicates of cosmetics purchased in the U.S. as well as five products from Canadian brands. Products selected for targeted analysis were repurchased in

2020 in both the U.S. and Canada to compare the amount and kinds of PFAS present. All ingredients were tabulated, including those listed as “may or may not be present”. Samples were grouped into categories based on the intended use of the product as defined by the manufacturer (Table S1). All brands analyzed and their product categories are reported in Table S2. Information on the collection and analysis of ingredient lists are given in the SI.

For PIGE analysis, all samples were applied to the surface of a fluorine-free Whatman 1 qualitative filter paper (Sigma-Aldrich, St. Louis, MO) or standard fluorine-free copier paper. These papers were also used as blanks. Samples were mounted to stainless steel target frames for *ex vacuo* ion beam analysis. These analyses were conducted on two particle accelerators, one at Hope College between 2016 and 2017 and the second at the University of Notre Dame between 2018 and 2020. Instrumental changes were made to the particle accelerator at the University of Notre Dame between 2018 and 2020. Fluorinated paper standards were analyzed at each facility to normalize the fluorine signals given at these different locations and time points. Details of this normalization, as well as the instrumental analysis using PIGE and of the use of inorganic fluoride standards soaked into paper to convert fluorine signals to a total fluorine concentration, are shown in Table S3 and Figure S1. The limit of detection (LOD) was 0.127 $\mu\text{g F}/\text{cm}^2$, and the limit of quantification was 0.384 $\mu\text{g F}/\text{cm}^2$, as determined by the standard response of prepared external inorganic fluoride standards. Replicate PIGE analyses were performed on roughly 5% of the samples, and a blank was measured every ~50 samples as QA/QC. Note that PIGE measures fluorine from inorganic and organic sources giving a measurement of the total fluorine in a sample.

Twenty-nine products including 20 with high total fluorine signals were selected for targeted analysis using LC-MS/MS and GC-MS. Full names, abbreviations, and methods for all 53 PFASs and additional information including the analytes measured for targeted analysis are given in Tables S4–S7 and have been adapted from methods previously published.¹⁸ Details of the targeted analysis are reported in the Supporting Information, including details on QA/QC. Briefly, 50–100 mg of product was spiked with surrogate standards before sonicating twice with 3 mL of 4:1 hexane–isopropanol and twice with 3 mL of 1:1 methanol–acetonitrile. Supernatants were combined and concentrated to a final volume of 5 mL under nitrogen. Concentrates were vortexed and centrifuged with 100 mg of Envi-Carb for cleanup, concentrated again under nitrogen, and filtered. Filtrate was transferred to polypropylene vials and spiked with internal standards for quantitation. LC-MS/MS analysis was done using an ultrahigh performance LC coupled with a triple-quadrupole MS (Agilent 1290 Infinity II UPLC–6470 QQQ-MS) in negative electrospray ionization mode. GC-MS analysis was performed on an Agilent 7890 GC–5977B PCI-MS operated in the positive chemical ionization mode. Additional instrumental parameters can be found in the Supporting Information of Wu et al.¹⁸ A procedural blank and a matrix spike sample were processed along with each batch sample to evaluate possible contamination from laboratory operations and the performance of our method. The recoveries of surrogate standards were all in the range of 60%–130%. Samples were corrected for recovery using the appropriate surrogate standards (Table S7). After this correction, matrix spike recoveries of individual analytes were all within 80%–115%. Additionally, data reported in this

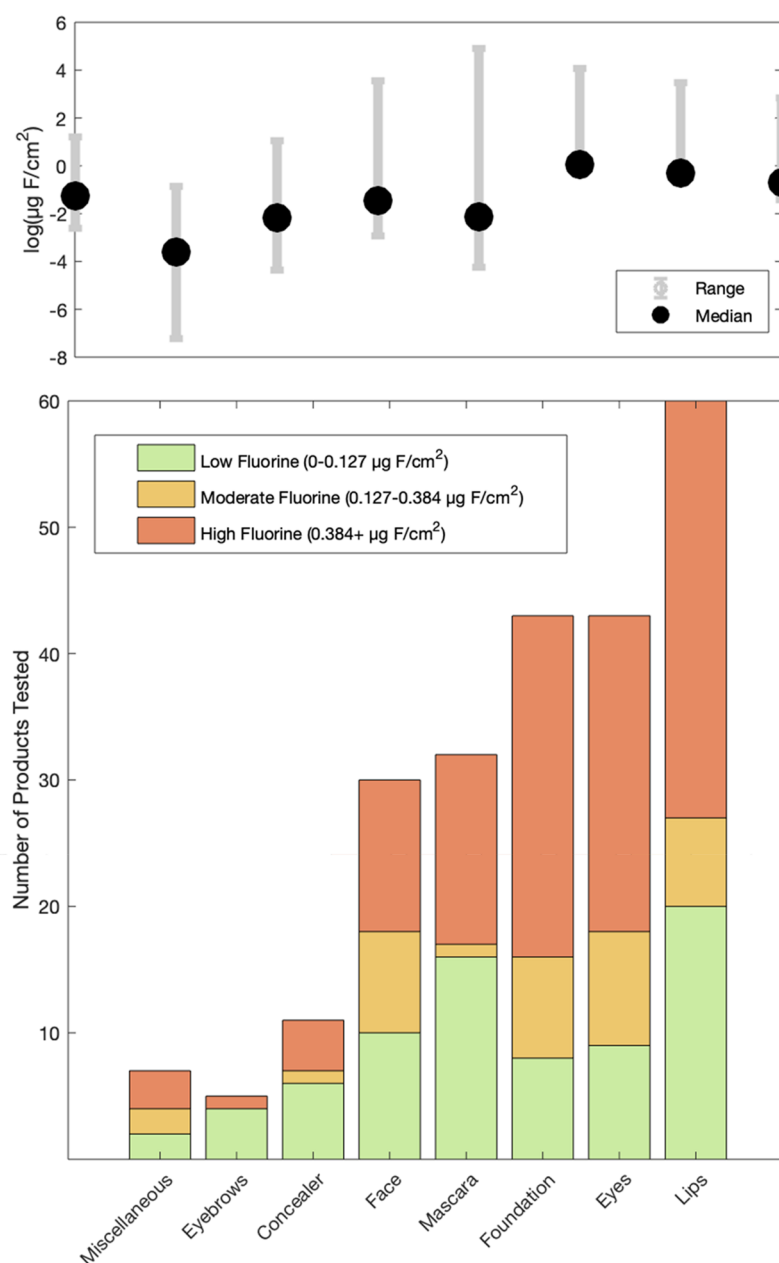


Figure 1. Foundations produced the highest median total fluorine ($\mu\text{g F/cm}^2$) in 231 products groups across eight categories. Breakdown of the number of products tested in each of the eight products categories and proportion of items in each category that was categorized as high, moderate, and low total fluorine concentrations. Several mascaras gave the highest fluorine concentrations measured. All remaining categories produced similar medians but with varying ranges. The three cosmetic categories that had the highest proportion of high fluorine concentrations were foundations, mascaras, and lip products.

study were blank corrected by subtracting the corresponding average blank on a mass basis. The method detection limits (MDLs) were defined as the average procedural blank $+3 \times$ standard deviation ($n = 5$) or the amount of chemical generating a signal-to-noise of 5 if the compound was not detected in the procedural blanks.

RESULTS AND DISCUSSION

Total Fluorine Screening Using PIGE. A total of 231 cosmetics across eight categories including lip products, eye products, foundations, face products, mascaras, concealers, eyebrow products, and miscellaneous products were analyzed using PIGE (Table S1). Foundations produced the highest median total fluorine concentration, while mascaras produced

the largest range of total fluorine measurements. Table S8 and Figure S2 provide additional analysis of the PIGE data in each category including the $\mu\text{g F/cm}^2$ distribution for all 231 products analyzed. We characterized the concentration of total fluorine for each product as containing fluorine \leq LOD ($0-0.127 \mu\text{g F/cm}^2$, 32%), fluorine between LOD and LOQ ($0.127-0.384 \mu\text{g F/cm}^2$, 16%), and high fluorine ($>0.384 \mu\text{g F/cm}^2$, 52%) as shown in Figure 1. For the subset of products analyzed by targeted analysis, concentrations ranged from LOD greater than 42.3 to 2360 $\mu\text{g F/g}$ product (Table S9). The cosmetic categories that had the highest percentage of high fluorine products were foundations (63%), eye products (58%), mascaras (47%), and lip products (55%).

A comparison of the total fluorine concentrations with those reported by Schultes et al.⁷ revealed similar ppm measurements of PFAS for overlapping cosmetic product categories such as foundations. The nine foundations tested in ref 14 had total fluorine concentrations determined using CIC from LOD of less than 91.1 to 3120 $\mu\text{g/g}$ which is a similar range to that reported here. Total fluorine reported by Brinch et al.⁸ ranged from 3.3 to 740 $\mu\text{g/g}$, lower than ranges reported here and by Schultes et al.⁷

Analysis of the total fluorine concentrations from PIGE revealed interesting trends within the selected categories. High fluorine levels were found in products commonly advertised as “wear-resistant” to water and oils or “long-lasting” including foundations, liquid lipsticks, and waterproof mascaras.^{19–22} These terms align with the functionality given for the use of many PFAS in cosmetics in industrial literature.^{23–25} The percentage of products with high fluorine concentrations in foundations, mascaras, and lip products suggested a possible link to the use of fluorinated ingredients in their manufacturing. Hence, foundations, mascaras, and lip products were selected for targeted MS analyses. Additional analyses of these trends determined from PIGE analysis are given in Table S9.

Targeted Analyses Using GC-MS and LC-MS/MS. All 29 cosmetic products contained detectable levels of at least four PFAS as shown in Table 1, with a maximum of 13 individual PFAS detected in a single product (Tables S11 and S12). Fluorotelomer alcohols (FTOHs), methacrylates (FTMA), and phosphate esters (PAPs) were the most frequently detected PFAS and also those contributing the most to total PFAS (Table S13). All of the collected products were used without homogenizing the entire mass or volume of the product before analysis. Differences in both the total fluorine concentrations from PIGE and targeted analyses results, which were not significantly correlated, suggests (1) numerous PFAS are not on the target list, (2) the presence of inorganic or polymeric fluorine, or (3) an effect due to the lack of homogenization or different shades/colors being chosen or all of the above.

The 6:2 species of these groups were the most commonly detected. Here, 6:2 FTOH was detected at a frequency of 100% and 94%, and 6:2 FTMA was detected at a frequency of 100% and 82%, in U.S. and Canadian products, respectively. The 6:2 monoPAP was detected at a frequency of 58% and 71% in U.S. and Canadian products, respectively. The high frequency of the 6:2 species presumably illustrates the overall industrial shift from longer chains, 8:2 and 10:2 PFAS, to shorter-chain variants in the market.²⁶ However, 8:2 FTOH, 8:2 FTMA, and 8:2 monoPAP were commonly found in U.S. and Canadian products signaling the continued use of or carryover contamination of longer-chain PFAS. Previous work on the biodegradation in mammals of both 8:2 FTOH and 6:2 FTOH has found that a fraction of FTOHs can undergo a series of metabolic transformations that result in the formation of terminal PFCAs, including perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), and perfluoroheptanoic acid (PFHpA), as well as longer-chain PFAS such as PFOA (in the case of 8:2 and longer FTOHs).^{26,27} Meta-analyses point to high toxicity and potentially bioaccumulative properties of some metabolites of 6:2 and 8:2 FTOHs.^{28,29} Of the samples subjected to targeted analyses, PFPeA was detected in 33% and 35% of the U.S. and Canadian products, respectively. The concentrations of PFCAs detected were substantially lower than those of FTOHs, PAPs, and FTMAcs.

Table 1. Summary of Total Fluorine Concentration ($\mu\text{g F/cm}^2$) Measured by PIGE, Total PFAS Concentrations Measured with GC/MS and LC/MS/MS, and Number of Individual PFAS Detected in Each Sample from the 53 Compounds Targeted

U.S. Products			
Sample ID	PIGE ($\mu\text{g F/cm}^2$)	Sum of 53 PFAS concentrations (ng/g)	Number of PFAS detected
Foundation 1	0.466	147	8
Foundation 2	5.82	10,500	6
Foundation 3	4.20	860	11
Lips 1	5.74	445	13
Lips 2	<0.127	1560	9
Lips 3	1.72	263	7
Lips 4	<0.127	216	7
Mascara 1	6.42	894	8
Mascara 2	<0.127	568	4
Mascara 3	<0.127	318	4
Mascara 4	2.97	264	6
Mascara 5	3.97	215	6
Canadian products			
Sample ID	PIGE ($\mu\text{g F/cm}^2$)	Sum of 53 PFAS concentrations (ng/g)	Number of PFAS detected
Foundation 1	0.895	70	10
Foundation 2	5.87	77	13
Foundation 3	7.07	1930	12
Foundation 4	2.32	1000	10
Lips 1	9.91	215	7
Lips 2	<0.127	1100	13
Lips 3	3.76	3820	11
Lips 4	<0.384	1610	9
Lips 5	1.16	235	8
Lips 6	34.4	3090	10
Mascara 2	<0.127	88	10
Mascara 3	<0.127	98	6
Mascara 4	12.7	192	9
Mascara 5	9.17	95	8
Mascara 6	21.7	161	10
Mascara 7	<0.127	365	6
Mascara 8	3.03	22	7

The measurement of both PAPs and PFCAs in these samples agrees with previous studies. Fujii et al.⁶ hypothesized that the measured levels of PFCAs in products in the Republic of Korea and Japan were attributed to PAPs disclosed on the products' ingredient lists. Measurement of PFCAs was expected to be from the use of PAPs, either as carryover contamination from their manufacturing or as degradation products since PAPs are formed from the condensation of FTOH with phosphate.^{6,30} Brinch et al.⁸ and Schultes et al.⁷ generated similar results for PFCAs, and the latter identified various mono- and di-PAPs in 9 of the 31 products tested. Three foundations from ref 7 which disclosed PAPs on their ingredient labels reported concentrations of PAPs in the parts-per-million ($\mu\text{g/g}$) range. However, products that did not disclose a PAP had concentrations from 11.2 to 282 ng/g, similar to our reported values of 0.05–244 ng/g for various mono- and di-PAPs.

Another potential source of PFCAs is the FTMAc detected here in both high frequency and concentration. Here, 8:2 FTMAc can breakdown into 8:2 FTOH, leading to increases in measurable PFCA concentrations, as previously de-

scribed.^{26,31,32} We speculate that the high concentrations of FTOHs measured here are likely due to both the impurities, as well as the breakdown, of both FTMAcs and PAPs. High levels of these nonpolymeric PFAS in products applied to the skin are a cause of concern due to potentially serious health risks as described above.^{28,29} Detection of FTMAcs in cosmetics is reported here for the first time. Their potential use in cosmetics as a film-forming agent was noted in 2018 by the CIR who reported insufficient data to enable an assessment of the safety of four FTMAc-based side-chain fluorinated polymers but that none of the cosmetic products surveyed were found to contain these four compounds.³³

Comparison of Total Fluorine and Targeted Analyses to Ingredient Labeling. Only 8% of the 231 cosmetics screened for total fluorine had any PFAS listed as ingredients, and only 3% of the 29 cosmetics in which targeted PFAS were measured had any PFAS listed as ingredients. Thus, we compiled a list of ingredients to try to determine the likely ingredient(s) responsible for PFAS in the 12 U.S. products chosen for targeted analysis. A total of 166 ingredients were listed across all 12 cosmetic products with 37% of these ingredients being listed on multiple product's ingredient lists. Tables S14–S16 provide the full lists of labeled ingredients and a heatmap relating ingredient frequency to total fluorine concentration from PIGE. Inorganic sources of fluorine found on ingredient labels included both natural and synthetic forms of minerals and clays used as bulking agents and/or colorants, such as synthetic fluorphlogopite and disteardimonium hectorite. Additional bulking agents and/or colorants include mica and talc, which are known to contain inorganic fluoride. As described by Fujii et al.⁶ and in various patents filed by cosmetic manufacturers, both mica and talc can be treated with PAPs to provide hydrophobic properties that improve the durability and wear of applied cosmetics.^{34,35} The same is true of other ingredients including silica, Nylon-12, and color additives.

Other potential sources of PFAS measured here include fluorinated versions of methicone and dimethicone, acrylate and methacrylate, and silicone polymers.^{36–41} These ingredients can be purchased under trade names from chemical suppliers, for example, Fluorosil products from Siltech or Pecosil fluorinated products from Phoenix Chemical, Inc., among others.^{23,25} We speculate that PFAS detected were from these ingredients described on the labels using only their generalized name, for example, methicone, acrylate. This interpretation is supported by documents suggesting that fluorinated ingredients and ingredients treated with PFAS were used in concentrations ranging from 0.1%–6.5% of the total cosmetic product weight.^{34,35,41} These concentrations are similar to the highest concentrations of the total fluorine measured here.

The measurements of high total fluorine concentrations in a variety of cosmetic products in combination with the detection of several PFAS classes using targeted analysis indicate widespread use of fluorinated ingredients in cosmetics. Of particular concern are the high levels of 6:2 and 8:2 FTOHs, which are precursors to PFCAs that are known to be environmentally mobile and can cause adverse effects in both humans and the environment. Results from the analyses of ingredient labels confirm that the use of fluorinated ingredients is poorly disclosed and contributes to the difficulty in estimating the magnitude of PFAS use in cosmetics. These estimates are important for understanding the kinds and

amounts of PFAS in relation to direct human exposure during product use. For example, PFAS could be ingested when lipstick containing PFAS is inadvertently ingested, and some PFAS in mascara could be absorbed through tear ducts. Without requirements for labeling, consumers cannot choose to avoid such exposures to PFAS in products they purchase. The use of PFAS in products also contributes to human and ecosystem exposures throughout the lifecycle of the product from PFAS manufacturing to product end of life. Cosmetic use contributes to PFAS entering wastewater streams leading to environmental accumulation of PFAS. This can lead to additional human exposure.

The high detection frequency found by PIGE and concentrations of PFAS measured by targeted analyses in this study suggest the need for better government oversight of PFAS, including the labeling of all cosmetic products containing these chemicals. Furthermore, given the considerable potential for human and ecosystem exposure and health harm, we question the use of any member of the highly persistent, potentially toxic, and very large class of PFAS in cosmetics. Better labeling and government oversight of harmful chemicals in personal care products are needed.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.1c00240>.

Additional details and parameters used for the instrumental analyses, including PIGE analysis and targeted LC-MS/MS and GC-MS analyses. List of categories used to categorize the cosmetics analyzed, as well as a list of cosmetic brands sampled. Full results of targeted LC-MS/MS and GC-MS analyses, as well as a summary of various features of the total fluorine analyses. Detection frequencies of various PFAS classes for targeted analysis. Summary of label information for all ingredients used in the targeted products. (PDF)

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Notes

The authors declare no competing financial interest.

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