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# Profile of per- and polyfluoroalkyl substances, source appointment, and determinants in Argentinean postpartum women

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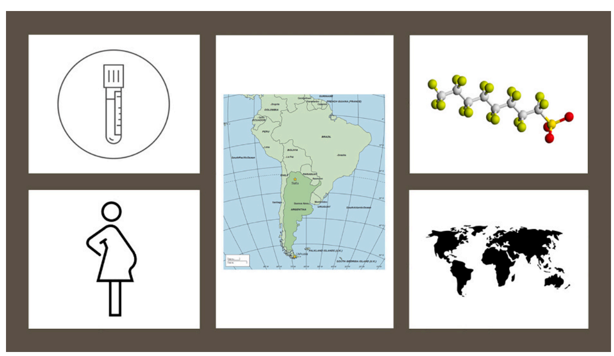
## HIGHLIGHTS

- Maternal serum PFAS from the world's southernmost city and the Andes mountain
- Low detection limits with a high percentage of detected samples for the 24 PFAS
- High detection frequencies of PFBA and PFHxA, which mainly are uncommonly in humans
- Source appointment indicates region-specific PFAS exposure sources despite low concentrations.

## GRAPHICAL ABSTRACT



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## ABSTRACT

*Background:* Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals with potential adverse health effects. Information concerning PFAS concentrations in relation to pregnancy is scarce in South America and non-existent in Argentina.
*Aim:* We aimed to investigate an extended maternal PFAS profile herein serum concentrations in a regional and global view, source appointment, and determinants in Argentinean women.
*Methods:* A cross-sectional study with a sampling period from 2011 to 2012 included 689 women from Ushuaia and Salta in Argentina. Serum samples collected two days postpartum were analyzed by ultra-high pressure liquid chromatography coupled to electrospray negative ionisation tandem-quadrupole mass-spectrometry. Principal Component Analysis (PCA) following absolute principal component score-multiple linear regression (APCS-MLR) was used for PFAS source appointments. Determinants of PFAS were explored through a MLR approach. A review of previous studies within the same period was conducted to compare with present levels.

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*Results:* Argentinean PFAS concentrations were the lowest worldwide, with PFOS (0.74 ng/mL) and PFOA (0.11 ng/mL) as the dominant substances. Detection frequencies largely aligned with the compared studies, indicating the worldwide PFAS distribution considering the restrictions. The PCA revealed region-specific loading patterns of two component groups of PFAS, a mixture of replaced and legacy substances in Ushuaia and long-chain in Salta. This might relate to a mix of non-diet and diet exposure in Ushuaia and diet in Salta. Region, age, lactation, parity, household members, migration, bottled water, and freshwater fish were among the determinants of various PFAS.

*Conclusion:* This is the first study to monitor human PFAS exposure in Argentina. Maternal PFAS concentrations were the lowest observed worldwide in the same period. Exposure contributions are suggested to be affected by restrictions and substitutions. Given the limited population-based studies and the emergence of PFAS, it is essential to conduct further monitoring of PFAS in Argentina and South America.

## 1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are synthetic chemicals widely used in industrial and consumer applications. They are environmentally persistent with a potential for global long-range transport and are bio-accumulative and toxic in living organisms (Buck et al., 2011). Thus, PFAS exposure raises health concerns (Fenton et al., 2021), with particular concern apparent to maternal and child issues, both from a short- and long-term perspective (Blake and Fenton, 2020; Rickard et al., 2022). Due to growing concerns, global regulation and phase-outs have been initiated from the 2000s for legacy PFAS, like perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) (Brennan et al., 2021), and lately on perfluorohexane sulfonic acid (PFHxS); all listed in the Stockholm Convention in 2009, 2019, and 2023, respectively (Stockholm Convention, 2023).

### Abbreviations

| | |
|---|---|
| AFFFs | aqueous film-forming foams |
| PFASs | *per*- and polyfluoroalkyl substances |
| PFCAs | perfluoroalkyl carboxylic acids |
| PFSAs | perfluoroalkyl sulfonic acids |
| FTSAs | fluorotelomer sulfonic acids |
| PFBS | perfluorobutane sulfonic acid |
| PFPeS | perfluoropentane sulfonic acid |
| PFHxS | perfluorohexane sulfonic acid |
| PFHpS | perfluoroheptane sulfonic acid |
| PFOS | perfluorooctane sulfonic acid |
| PFNS | perfluorononane sulfonic acid |
| PFDS | perfluorodecane sulfonic acid |
| PFDoDS | perfluorododecane sulfonic acid |
| PFBA | perfluorobutanoic acid |
| PFPeA | perfluoropentanoic acid |
| PFHxA | perfluorohexanoic acid |
| PFHpA | perfluoroheptanoic acid |
| PFOA | perfluorooctanoic acid |
| PFNA | perfluorononanoic acid |
| PFDA | perfluorodecanoic acid |
| PFUnDA | perfluoroundecanoic acid |
| PFDoDA | perfluorododecanoic acid |
| PFTrDA | perfluorotridecanoic acid |
| PFTeDA | perfluorotetradecanoic acid |
| FOSA | perfluorooctane sulfonamide |
| 4:2 FTSA | 4:2 fluorotelomer sulfonic acid |
| 6:2 FTSA | 6:2 fluorotelomer sulfonic acid |
| 8:2 FTSA | 8:2 fluorotelomer sulfonic acid |
| 10:2 FTSA | 10:2 fluorotelomer sulfonic acid |

The maternal body burden of PFAS has been investigated worldwide, mostly in developed regions, and detected as ubiquitous in varying concentration ranges with PFOS and PFOA as predominant substances (Bjerregaard-Olesen et al., 2017; Liu et al., 2020). Restrictions on production and use have reflected declining human temporal trends of PFOS and PFOA. From the 1990s until 2018, in the USA, Canada, Germany, Sweden, and Norway, human PFOS has declined from around 60 % to 75 %, also for PFOA, although with more variations across nations (Fan et al., 2022). Similar declines were observed in Australia from 2002 to 2011 (Toms et al., 2014). Also, maternal declining trends were observed in Denmark (2008–2013) (Bjerregaard-Olesen et al., 2016), Japan (2003−2011) (Okada et al., 2013), and in most studies in a global review (1972–2016), except an upward effect for Inuits in Alaska (Liu et al., 2020).

Simultaneous, various trends have been observed in human exposure to replacement PFAS, which include both long-chain substances (with nine or more carbon-fluor chains) and short-chain substances (less than C8) (Fan et al., 2022; Liu et al., 2020; Okada et al., 2013). For example, mostly delayed downward trends were reported for PFHxS, but an upward trend in Sweden for PFHxS and perfluorodecanoic acid (PFDA) (1972–2016) (Liu et al., 2020). Increasing trends were also seen for PFNA perfluorononanoic acid (PFNA), PFDA, and perfluoroundecanoic acid (PFUnDA) in Inuits in Canada (2004–2017) (Caron-Beaudoin et al., 2020) and PFNA and PFDA in Japan, but solely declining trends in Denmark (Bjerregaard-Olesen et al., 2016).

Paralleling the global consensus of regulations, with exemptions, the production of PFOS and PFOA has continued in Asia, particularly in China (Wang et al., 2015). Additionally, PFOS has also been imported into Brazil for Sulfluramid manufacturing (Barbosa Machado Torres et al., 2022). Consequently, human exposure trends in China increased for PFOS until 2009 and PFOA until 2015 (Fan et al., 2022). Trends for South America are less pronounced. To our knowledge, PFAS status is limited to maternal studies from Brazil in 2011 (Souza et al., 2020) and 2017 (Espindola Santos et al., 2021) and global studies with small samples from Latin America (Fiedler and Sadia, 2021) and Brazil (Kannan et al., 2004).

Human PFAS profiles are influenced by several factors, with diet being the primary human exposure. This is followed by drinking water, while dermal absorption, dust, and air inhalation – especially personal care products and indoor environments – also contribute (Jian et al., 2017; Pérez et al., 2014; Wee and Aris, 2023). Pregnancy and lactation are regulators of PFAS through placental and lactation transfer to the baby (Olsen et al., 2009). Race, region, country of origin, and socio-economic status are other relevant determinants (McAdam and Bell, 2023). Age is reported with an inconclusive relationship to PFAS, mainly explained by production, emission, regulation, year of peak, and half-life (McAdam and Bell, 2023; Quinn and Wania, 2012). Exposure contamination grade and the properties of each substance affect the body burden. Humans are exposed to a mixture of PFAS and with variety within and across nations (EFSA, 2020). Profiles of human PFAS exposure might be helpful to understand sources and exposure pathways (Hu et al., 2018), and thus, it is essential to describe the population status beyond legacy substances such as PFOS and PFOA (Sunderland et al., 2019).

This study aimed to enhance understanding of PFAS exposure in South America by providing a comprehensive serum PFAS profile of postpartum women in two Argentinean regions. We include a global comparison and identify sources and predictors of PFAS levels in our study.

## 2. Material and methods

### 2.1. Description of the study area and data collection

The EMASAR study (Estudio del Medio Ambiente y la Salud Reproductiva; Study on the Environment and Reproductive Health) aimed to investigate contaminants related to pregnancy in Argentina. Study areas were designated to Ushuaia, the world's southernmost city, and Salta in the northwestern highland of Argentina. Responsible for the project were UiT, The Arctic University of Norway, Tromsø, and Stavanger University Hospital, both in Norway. Local partners were the private institution Clínica San Jorge in Ushuaia and the Hospital Público Materno Infantil in Salta.

The study period ranged from April 2011 to November 2011 in Ushuaia and June 2011 to March 2012 in Salta and included 698 women (200 from Ushuaia and 498 from Salta). For this present PFAS study, nine participants were excluded due to damaged serum vials. Non-fasting blood samples were obtained at a median of one day with a range of 0–3 days following delivery. Information collected through personal interviews and medical records covered socio-economy, pregnancy, health and lifestyle conditions, diet, and environmental aspects. A detailed description of the study has been given elsewhere (Okland et al., 2017).

Local approvals of the EMASAR study were given by the Ethics Committee of the Salta Medical Association and the Ministries of Health in both the Province of Salta and the Province of Tierra del Fuego (#2010/7317). The Norwegian Regional Committee for Medical and Health Research Ethics (REC North) approved the study (#2011/706). The study was conducted in accordance with the Helsinki Declaration. Informed consent was obtained from the participating woman.

### 2.2. Chemical analysis of PFAS

Maternal blood was collected in BD Vacutainers® (BD SST II Plus Advance 10/8.5 mL), sampled and treated according to the instruction leaflet, and centrifuged at 2000 relative centrifugal force (RCF) for 10 min. The serum was subsequently apportioned into n-hexane/acetone pre-rinsed glass vials. Samples were shipped in a frozen state to the Biobank at the UiT The Arctic University of Norway for storage at minus 30 degrees Celsius. In 2019, samples were analyzed for 24 PFAS and 6 accompanying linear substances.

The analytical work was conducted at The Environmental Pollution Laboratory, Department of Laboratory Medicine, University Hospital of North Norway, Norway. Aliquotes of 50 μL serum samples were extracted by solid-phase microelution on an Oasis WAX-μElution plate (30 μm, Waters, Milford, USA) and analyzed by ultra-high pressure liquid chromatography coupled to electrospray negative ionisation tandem-quadrupole mass-spectrometry (Waters Acquity UPLC Xevo TQ-S system, Milford, MA, USA) as described previously (Huber and Brox, 2015). The limit of detections (LODs) was set as concentrations calculated by the Targetlynx-software for each sample (LODi) and each PFAS with a signal-to-noise ratio of 3 divided by the related sample amount. For quality assurance, four blank samples, four SRM 1958 (NIST, Gaithersburg, MD, USA), and three bovine serum samples (Sigma Aldrich, Steinheim, Germany) were prepared and analyzed within each batch of 96 samples to control for background and carry-over effects. All the quality controls were within the acceptance limits. Correlation coefficients of variation were $< 10$ % for all measured PFAS. The limit of detection was in the range of 0.0034 ng/mL to 0.031 ng/mL for analyzed PFAS; five substances were not detected (Table S3). For PFBA and PFOA, a batch-wise blank subtraction was performed. Concentrations detected in the blank samples of each individual sample preparation batch were used and subtracted from the concentration measured in the samples from the EMASAR study as follows: sample concentration – (average blank concentration + 3 × standard deviation blank concentration). Subtracted blank concentrations were around 0.50 ng/mL for PFBA and 0.11 ng/mL for PFOA, respectively. All PFAS analyses were within the acceptable ranges of the international quality control program: the Arctic Monitoring and Assessment (AMAP) Ring Test for Persistent Organic Pollutants in Human Serum (organized by Centre du Toxicologie du Québec (CTQ), Institut National de Santé Publique du Quebec, Canada.

### 2.3. Statistical analysis

Statistical analyses were done using the IBM SPSS Statistics for Windows statistical package version 28 (SPSS Inc. Chicago, IL, USA). Figures were performed in R. Significant levels were set at $p < 0.05$. PFAS concentrations below LOD were replaced by individual LOD divided by the square root of 2 (Hornung and Reed, 1990). According to the Kolmogorov-Smirnov test, most substances deviated from a normal distribution and were harmonized during $\log_{10}$ transformation. Descriptive analyses described raw data, and the *t*-test, chi-square, or Mann-Whitney test explored variations. The Pearson correlation (r) assessed correlations between the PFAS.

Through the dimensional reduction method principal component analysis (PCA), we explored the linear relationship between the log-PFAS by region. Restricted to substances detected above 60 %, only one cluster was revealed for Salta, and rotation was not performed. Thus, the detection frequencies were extended. Initially, ten substances were included, followed by the exclusion of perfluorobutanoic acid (PFBA) for Ushuaia only and perfluoroheptanoic acid (PFHpA) due to a correlation below 0.3 (Table S5). Orthogonal (varimax) rotation was used. Both sampling adequacy measured by Kayser-Meyer-Olkin (KMO 0.8) and Bartlett's` test of sphericity ($p < 0.001$) met the correlation criteria for PCA. Kaiser's criterion ≥1 was set in selecting important factors. Factor loading was evaluated as strong (> 0.75), moderate (0.5–0.75), or weak <0.05–0.3) (Cho et al., 2022). Next, the absolute principal component score-multiple linear regression (APCS-MLR) model was performed as described in detail by others (Cho et al., 2022; Wallis et al., 2023). The individual PFAS contribution (dependent variable) in percentage (%) of each factor loading (independent variable) from the PCA was explored in a multiple linear regression (MLR) model. Percentage calculations were based on the equation: i (%) = 100 * (Bi/ΣniBi), where Bi is the beta coefficient for the individual PFAS (Wallis et al., 2023). To account for the contribution of negative values, negative beta coefficients were treated as positive values (Haji Gholizadeh et al., 2016).

MLR, using a stepwise procedure with backward elimination, described the relationships between seven frequently detected log 10 transformed substances and selected independent factors according to McAdam and Bell (2023). Initial variables were age (year), parity (parous - multiparous), lactation (interval), region (Ushuaia - Salta), people in the household (number), education (primary/secondary - tertiary/university), migration inland/abroad (no-yes), bottled water almost daily (no-yes), dietary factors during pregnancy (never/seldom - weekly/daily): freshwater fish, saltwater fish, seafood, meat, poultry, processed meat, egg, dairy products (milk), butter/cheese, vegetables (bean, tomato, garlic), fruit, bread. Other vegetables and items were not included due to intake below 3 %. Finally, the models were evaluated through diagnostic plots, variance inflation factor (VIF), Mahalanobis and Cook's distance, and casewise diagnostics of standardized residuals with exclusions of the most influential outliers (Field, 2009). The modified Breusch-Pagan test checked for abruption of homogeneity ($p < 0.05$) and with robust standard errors corrected for heteroskedasticity (Mansournia et al., 2020).

For a global comparison of PFAS concentrations, a review of publications published after 2010 was performed on PubMed. The search string was [*PFAS per-and polyfluoroalkyl substances pregnancy*] in addition to *blood, whole blood, serum, or plasma* with 438, 383, 174 or 22 matches, respectively. Additionally, reference lists in review publications were searched. After searching the sampling period, 16 studies collected between 2010 and 2013 were identified. In the aftermath, a recent publication confirmed the included studies (Kuo et al., 2023).

## 3. Results

### 3.1. Background characteristics

Compared to those in Salta, women living in Ushuaia were notably older, had fewer children, and had shorter lifetime breastfeeding but the proportion of first-time mothers was similar. Ushuaian women also had higher education and more household members (Table 1). Regarding diet during pregnancy, those from Ushuaia had a significantly higher intake of bottled water, dairy, and marine food, but a lower intake of processed meat, eggs, bread, pasta/cereals, and sugar. The consumption of freshwater fish, meat, vegetables, or fruits did not differ between locations (Supplementary Table S2). Characteristics have been described in detail elsewhere (Okland et al., 2017).

### 3.2. Serum PFAS detection frequencies and concentrations

The distributions of PFAS in maternal serum samples varied fairly between Ushuaia and Salta (Tables 2, S3). Of the 24 PFAS analyzed, seven substances had a detection frequency above 80 %. Notably, PFOS was detected in all samples of the study group. In Ushuaia, PFOA showed a similar universal presence, followed by the frequency order of PFHxS = PFNA > PFBA > PFDA. In Salta, the order was PFHxS > perfluorohexanoic acid (PFHxA) > PFNA > PFOA > PFDA, with the highest detection frequencies in Ushuaia. Striking were the detection differences between PFBA and PFHxA with alternating detection between the cities. Substances with substantially lower detection are reported in Tables 2, S3, and S4.

**Table 1**
Characteristic of the study population by region. The EMASAR study in Argentina, 2011–2012.

| | Ushuaia $n = 193$ Mean, SD or n (%) | Salta $n = 496$ Mean, SD or n (%) | *P*-value |
|---|---|---|---|
| Age, year | 28.8 ± 6.5 | 24.7 ± 6.2 | <0.001[a] |
| Parity, number | 1.9 ± 0.97 | 2.2 ± 1.5 | 0.005[b] |
| para 1 | 78 (40.4) | 221 (44.6) | 0.001[c] |
| para 2 | 72 (37.3) | 120 (24.2) | |
| para >2 | 43 (22.3) | 155 (31.3) | |
| Breastfeeding, months, overall | 12.3 ± 18.2 | 18.6 ± 27.3 | <0.001[b] |
| Breastfeeding, months, multipara | 20.6 ± 19.7 | 34.0 ± 29.0 | <0.001[b] |
| BMI, postpartum | 28.0 ± 4.1 | 26.1 ± 4.3 | <0.001[a] |
| Smoking during last year | 54 (28.0) | 127 (26.3) | 0.525[c] |
| Education | | | |
| Primary | 7 (3.6) | 161 (32.5) | 0.001[c] |
| Secondary | 93 (48.2) | 283 (57.2) | |
| Tertiary | 55 (28.5) | 39 (7.9) | |
| University | 38 (19.7) | 12 (2.4) | |
| Marital status | | | |
| Married/cohabitant | 172 (89.1) | 337 (73.9) | <0.001[c] |
| Single/divorced | 21 (10.9) | 159 (32.1) | |
| Permanent job, yes | 128 (66.3) | 86 (31.2) | <0.001[c] |
| People living in your home, number | 4.2 ± 1.4 | 6.6 ± 3.5 | <0.001[b] |
| Country/province born | | | |
| Born current province | 63 (32.6) | 437 (88.1) | |
| Born other provinces | 124 (62.3) | 44 (8.9) | |
| Born other nations | 6 (3.1) | 15 (3.0) | |

SD, standard deviations.
[a] T-test.
[b] Welch test.
[c] Chi-square test.

**Table 2**
Maternal serum concentrations (ng/mL) of per- and polyfluoroalkyl substances by region. The EMASAR study in Argentina, 2011–2012[a].

| Region | | PFBA | PFHxA | PFHpA | PFOA | PFNA | PFDA | PFUnDA | Linear PFHxS | Sum PFHxS |
|---|---|---|---|---|---|---|---|---|---|---|
| Ushuaia ($n$ = 193) | GM | 0.07 | 0.04 | 0.008 | 0.24 | 0.06 | 0.02 | 0.02 | 0.06 | 0.18 |
| | AM | 0.09 | 0.10 | 0.01 | 0.29 | 0.07 | 0.03 | 0.02 | 0.09 | 0.23 |
| | Median | 0.08 | 0.09 | 0.007 | 0.24 | 0.05 | 0.03 | 0.01 | 0.05 | 0.18 |
| | Min | 0.01 | 0.004 | 0.004 | 0.04 | 0.006 | 0.005 | 0.003 | 0.004 | 0.006 |
| | 25%ile | 0.05 | 0.007 | 0.006 | 0.15 | 0.040 | 0.02 | 0.008 | 0.04 | 0.12 |
| | 75%ile | 0.11 | 0.17 | 0.01 | 0.38 | 0.08 | 0.03 | 0.03 | 0.09 | 0.27 |
| | Max | 0.29 | 0.47 | 0.06 | 1.28 | 0.42 | 0.20 | 0.18 | 3.20 | 3.30 |
| | % > LOD | 88.1 | 58.5 | 19.7 | 100 | 99.5 | 85.5 | 53.9 | 98.4 | 99.5 |
| Salta ($n$ = 498) | GM | 0.04 | 0.06 | 0.01 | 0.08 | 0.03 | 0.02 | 0.007 | 0.04 | 0.21 |
| | AM | 0.06 | 0.08 | 0.01 | 0.11 | 0.04 | 0.03 | 0.009 | 0.06 | 0.26 |
| | Median | 0.05 | 0.07 | 0.008 | 0.09 | 0.04 | 0.02 | 0.006 | 0.04 | 0.22 |
| | Min | 0.008 | 0.002 | 0.002 | 0.004 | 0.004 | 0.004 | 0.001 | 0.002 | 0.002 |
| | 25%ile | 0.02 | 0.04 | 0.006 | 0.05 | 0.027 | 0.016 | 0.006 | 0.026 | 0.147 |
| | 75%ile | 0.09 | 0.10 | 0.02 | 0.14 | 0.48 | 0.03 | 0.008 | 0.67 | 0.32 |
| | Max | 0.23 | 0.27 | 0.10 | 2.90 | 1.14 | 0.55 | 0.13 | 0.81 | 1.28 |
| | % > LOD | 60.3 | 96.4 | 32.1 | 89.7 | 92.7 | 85.5 | 19.2 | 97.2 | 98 |
| | P-value[b] | <0.001 | 0.693 | | <0.001 | <0.001 | 0.616 | | <0.001 | <0.001 |

| Region | | Linear PFHpS | Sum PFHpS | Linear PFOS | Sum PFOS | Sum PFAS | Sum PFSA | Sum PFCA | Sum PFSA and PFCA |
|---|---|---|---|---|---|---|---|---|---|
| Ushuaia ($n$ = 193) | GM | 0.007 | 0.01 | 0.42 | 0.84 | 1.73 | 1.07 | 0.60 | 1.70 |
| | AM | 0.01 | 0.02 | 0.51 | 1.02 | 1.97 | 1.28 | 0.66 | 1.94 |
| | Median | 0.01 | 0.02 | 0.41 | 0.84 | 1.75 | 1.08 | 0.58 | 1.73 |
| | Min | 0.001 | 0.001 | 0.14 | 0.28 | 0.69 | 0.38 | 0.23 | 0.67 |
| | 25%ile | 0.003 | 0.003 | 0.28 | 0.58 | 1.26 | 0.76 | 0.42 | 1.23 |
| | 75%ile | 0.02 | 0.034 | 0.55 | 1.13 | 2.23 | 1.44 | 0.80 | 2.20 |
| | Max | 0.19 | 0.28 | 6.02 | 14.4 | 15.90 | 15.2 | 2.37 | 15.85 |
| | % > LOD | 55.4 | 55.4 | 100 | 100 | | | | |
| Salta ($n$ = 498) | GM | 0.004 | 0.004 | 0.40 | 0.70 | 1.37 | 0.97 | 0.35 | 1.35 |
| | AM | 0.007 | 0.010 | 0.56 | 0.97 | 1.66 | 1.24 | 0.39 | 1.64 |
| | Median | 0.004 | 0.004 | 0.39 | 0.70 | 1.38 | 0.97 | 0.37 | 1.36 |
| | Min | 0.001 | 0.001 | 0.06 | 0.11 | 0.24 | 0.12 | 0.07 | 0.22 |
| | 25%ile | 0.002 | 0.002 | 0.27 | 0.50 | 1.09 | 0.74 | 0.28 | 1.07 |
| | 75%ile | 0.008 | 0.013 | 0.54 | 0.92 | 1.74 | 1.25 | 0.48 | 1.72 |
| | Max | 0.45 | 0.53 | 33.1 | 59.8 | 60.97 | 60.5 | 5.03 | 60.95 |
| | % > LOD | 24.8 | 25 | 100 | 100 | | | | |
| | P-value[b] | | | 0.326 | <0.001 | <0.001 | 0.025 | <0.001 | <0.001 |

[a] The limit of detection was in the range of 0.0034 ng/mL to 0.026 ng/mL for all PFAS; Substances with lower detection frequency (<12 %) are reported in the Supplementary file.
[b] Group comparison for PFAS detected >50 %, Mann-Whitney test; AM, arithmetic mean; GM, geometric mean; max, maximum; min, minimum; %ile, percentile; abbreviations of the substances, see Table S1.

Median serum PFOS, PFOA, PFBA, and PFNA were significantly highest in Ushuaia women, while PFHxS were highest in Salta women (Table 2). Of the total PFAS, the highest sum concentration was observed in Ushuaia ($p < 0.03$, Table 2). The seven dominating substances accounted for 93 % of the total PFAS in both places. Further, the perfluoroalkyl sulfonic acids (PFSAs; PFOS, PFHxS and perfluoroheptane sulfonic acid (PFHpS)) were the leading fraction of total PFAS, comprising 64.9 % in Ushuaia and 75 % in Salta. The fraction of linear PFOS to the total PFOS was 49.8 % and 56.9 %, and linear PFHxS to the total PFHxS was 40.9 % and 21.2 % in Ushuaia and Salta, respectively ($p < 0.001$, data not shown).

### 3.3. *Comparison of Argentinean PFAS profile to global studies*

Covering worldwide maternal blood PFAS biomonitoring around a similar period, primarily representing the Northern Hemisphere, present Argentinean concentrations of PFAS were of the lowest reported (Table 3a). In all studies, PFOS was the dominating substance, followed by PFOA. The Argentinean PFOS and PFOA were comparable to Tanzania. Argentinean PFOS were ten times lower than neighboring Brazil, 3–7 times lower than North America (USA > Canada), 4–10 times lower than Europe (Denmark > France), 5 to 15 times lower than both Asia (China > South Corea > Japan) and Inuit women (Greenland > Canada). For PFOA, our levels were half of Brazil, 10–15 times lower than North America and Europe, and >12 times lower than Asia but 350 times lower than China. The remaining PFAS varied across nations. Long-chain PFAS were highest in Inuit women in Greenland and Canada but comparable to China. In general, PFOS and PFOA were detected in all study groups except for PFOA, with detection of 68 % in Brazil and 90 % in Tanzania and Salta. Other PFAS were highly detected, although with more variations across the countries (Table 3b).

### 3.4. *PFAS loading profile and source contribution across regions*

In the PCA, two loading components satisfying the Kaisers criterion ≥1 and explained around 40 % and 20 % of the variance for their respective component groups. We observed distinct PFAS rotated loading patterns between the two regions (Fig. 1, Table S6). The contribution of each substance's loadings in the APCS-MLR model is presented in brackets (Fig. 2, Table S7). In Ushuaia, component 1 was strongly influenced by PFHxA (85.5 %), PFHpS (78.6 %), and moderately by PFOS (74.0 %), PFHxS (90.5), and PFOA (59.3 %) and PFHpS (86.3 %). Component 2 was clustered strongly by PFDA (88.4 %) and PFUnDA (89.6 %) and moderately by PFNA (53.2 %). In Salta, component 1 was moderately controlled by PFDA (76.4 %), PFUnDA (82.9 %), PFOS (65.8 %), and PFHpS (86.3 %), and component 2 was strongly influenced by PFHxA (82.8 %) and moderately by PFBA (88.8 %), PFOA (57.7 %), and PFHxS (74.9 %). Additionally, The MLR analysis of moderate to strong PFAS included in the factor loadings to each APCS score supported the clustering profile with an explained variance of 92–97 % (Table S7).

### 3.5. *Determinants of maternal PFAS concentrations*

Fig. 3 demonstrates the determinants of PFAS discovered through MLR. Compared to Ushuaia, living in Salta was associated with higher concentrations of PFHxA (+74 %) and PFHxS (37 %), but similar lower concentrations of PFOA (−61 %), PFBA (−43 %) and PFNA (−27 %). Age elevated PFOS, PFHxA, PFOA, and PFNA by up to 24 % by five years. Advancing parity from mono- to multiparous reduced all substances up to 45 % except for PFBA and PFDA. Lactation decreased PFOS, PFOA, and PFHxA by around 6 % per half year. Elevating household members lowered PFOA, PFOS, and PFDA by 2 %. Concerning diet, a swift from never/seldom to weekly/daily intake, freshwater fish contributed to PFOS by 12 % and PFDA by 19 %, and fruit and egg elevated PFHxA up to 30 %. Inverse associations were revealed for food

**Table 3a**
Global comparison of PFAS (ng/mL) in maternal serum, plasma, or whole blood in selected studies during 2010–2013.

| Year | Country | n | PFBA | PFHxA | PFHpA | PFOA | PFNA | PFDA | PFUnDA | PFHxS | PFHpS | PFOS | Central tendency | Material | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 2013 | France (Toulouse) | 100 | | | | 1.05 | 0.43 | ND | ND | 0.62 | ND | 3.07 | Median | S | Cariou et al., 2015 |
| 2012 | China (Tanjin) | 141 | | 0.45 | 0.18 | 4.0* | 1.05 | 1.33 | 0.61 | 0.38 | | 6.7[a] | GM | S | Jiang et al., 2014 |
| 2012 | China (Shanghai) | 981 | | | | 19.6 | 1.79 | 2.05 | 1.54 | 2.73 | | 10.8 | GM | P | Tian et al., 2018 |
| 2012 | Canada (Nunavik) | 111 | ND | ND | | 0.67 | 2.00 | 0.45 | 0.44 | 0.35 | | 3.80 | GM | S | Caron-Beaudoin et al., 2020 |
| 2012 | Tanzania (Arusha) | 48 | | ND | ND | 0.21 | 0.17 | 0.14 | NC | NC | | 0.50 | Median | P | Müller et al., 2019 |
| 2011–2012 | Argentina (Ushuaia) | 193 | 0.07 | 0.01 | 0.008 | 0.24 | 0.03 | 0.02 | 0.02 | 0.18 | 0.007 | 0.84 | GM | S | Present study |
| 2011–2012 | Argentina (Salta) | 496 | 0.04 | 0.008 | 0.01 | 0.08 | 0.06 | 0.02 | 0.007 | 0.21 | 0.004 | 0.70 | GM | S | Present study |
| 2011–2012 | USA (Colorado) | NA | | | | 1.80 | 0.50 | 0.80 | | 1.00 | | 5.30 | Median | S | Zell-Baran et al., 2023 |
| 2011 | Canada (10 cities) | 95 | | | | 1.38 | | | | 1.04 | | 3.65 | GM | P | Fisher et al., 2016 |
| 2011 | France (national) | 277 | ND | ND | ND | 1.50 | 0.52 | 0.26 | NC | 0.74 | NC | 3.1 | GM | S | Dereumeaux et al., 2016 |
| 2011 | Japan (Hokkaido) | 30 | | <0.1 | <0.1 | 1.27 | 1.26 | 0.69 | 1.3 | 0.33 | | 3.52 | Median | P | Okada et al., 2013 |
| 2011 | South Korea (Gyeongbuk) | 70 | | | | 2.62 | | | | 1.21 | | 9.37 | Median | S | Lee et al., 2013 |
| 2010–2013 | China (Shandong) | 369 | | | 0.06 | 39.3 | 0.78 | 0.52 | 0.46 | 0.32 | | 4.25 | GM | S | Wang et al., 2019 |
| 2010–2013 | Greenland (national) | 207 | | NC | NC | 1.19 | 1.30 | 0.72 | 1.60 | 0.7 | 0.19 | 10.2 | Median | S | Long et al., 2015 |
| 2010–2012 | Denmark (Odense) | 1436 | | | | 1.68 | 0.64 | 0.29 | | 0.36 | | 7.5 | Median | S | Birukov et al., 2021 |
| 2010–2011 | Brazil (Ribeirão Preto) | 243 | | ND | ND | 0.46 | NC | NC | NC | NC | | 7.8 | GM | WB[b] | Souza et al., 2020 |
| 2010–2011 | Canada (Winnipeg) | 247 | | ND | ND | 0.89 | 0.37 | 0.13 | 0.069 | 0.44 | | 2.2 | Median | P | Workman et al., 2019 |
| 2010 | Canada (10 cities) | 943 | | | | 1.62 | | | | 1.07 | | 4.3 | GM | P | Fisher et al., 2016 |

GM, geometric mean; med., median; n, number of participants; NA, not available; ND; not detectable; NC, quantified, but GM or median concentration not reported/calculated due to low detection.
[a] Median.
[b] Whole blood concentration multiplied by two for comparison with serum or plasma concentrations as described in Ehresman et al., 2007.

**Table 3b**
Global comparison of detection frequencies of PFAS (ng/mL) in maternal serum, plasma, or whole blood in selected studies during 2010–2013.

| Country | Year | Detection | PFBA | PFHxA | PFHpA | PFOA | PFNA | PFDA | PFUnDA | PFHxS | PFHpS | PFOS | Material | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| France (Toulouse) | 2013 | % > LOD | | | | 100 | 98 | 93 | 79 | 99 | 50 | 100 | S | Cariou et al., 2015 |
| China (Tanjin) | 2012 | | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | | 100 | S | Jiang et al., 2014 |
| China (Shanghai) | 2012 | % > LOD | | | | 100 | 100 | 100 | 99.9 | 100 | | 100 | P | Tian et al., 2018 |
| Canada (Nunavik) | 2012 | % > LOD | ND | ND | | 100 | 100 | 98 | 92 | 92 | | 100 | S | Caron-Beaudoin et al., 2020 |
| Tanzania (Arusha) | 2012 | % > LOD | | ND | ND | 90 | 81 | 85 | 27 | 38 | | 100 | P | Müller et al., 2019 |
| Argentina (Ushuaia) | 2011–2012 | % > LOD | 88.1 | 58.5 | 19.7 | 100 | 99.5 | 85.5 | 53.9 | 99.5 | 55.4 | 100 | S | Present study |
| Argentina (Salta) | 2011–2012 | % > LOD | 60.3 | 96.4 | 32.1 | 89.7 | 92.7 | 85.5 | 19.2 | 98 | 24.8 | 100 | S | Present study |
| USA (Colorado) | 2011–2012 | % > LOD | | | | 99.7 | 98.2 | 64.8 | | 98.5 | | 99.3 | S | Zell-Baran et al., 2023 |
| Canada (10 cities) | 2011 | % > LOD | | | | 100 | | | | 97.9 | | 100 | P | Fisher et al., 2016 |
| France (national) | 2011 | % > LOQ | ND | ND | 0.4 | 100 | 100 | 67.9 | 30.3 | 99.6 | 7.2 | 100 | S | Dereumeaux et al., 2016 |
| Japan (Hokkaido) | 2011 | % > LOD | | 20 | 50 | 100 | 100 | 100 | 100 | 76.7 | | 100 | P | Okada et al., 2013 |
| South Korea (Gyeongbuk) | 2011 | | | | | 100 | | | 100 | | | 100 | S | Lee et al., 2013 |
| China (Shandong) | 2010–2013 | % > LOD | | | 85.1 | 100 | 100 | 100 | 100 | 99.7 | | 100 | S | Wang et al., 2019 |
| Greenland | 2010–2013 | % > LOQ | | ND | 16.3 | 100 | 100 | 100 | 100 | 100 | 86.8 | 100 | S | Long et al., 2015 |
| Denmark (Odense) | 2010–2012 | % > LOQ | | | | 100 | 100 | 100 | | 96 | | 100 | S | Birukov et al., 2021 |
| Brazil (Ribeirão Preto) | 2010–2011 | % > LOD | | ND | ND | 67.9 | 11.1 | 1.65 | 0.41 | 0.41 | | 100 | WB[b] | Souza et al., 2020 |
| Canada (Winnipeg) | 2010–2011 | % > LOQ | | ND | ND | 100 | 96 | 94 | 85 | 89 | | 100 | P | Workman et al., 2019 |
| Canada (10 cities) | 2010 | % > LOD | | | | 99.7 | | | | 97.1 | | 99.7 | P | Fisher et al., 2016 |

LOD, limit of detection; LOQ; limit of quantification; ND, not detectable; S, serum; P, plasma; WB, whole blood.

items to mostly all substances, except PFOA and PFHxS, with no dietary relationship. Daily bottled water intake elevated PFHxA by 27 % and PFOA by 16 %. Table S9 shows the detailed MLR results.

## 4. Discussion

To our knowledge, this is the first study observing PFAS profiles in populations living in Argentina. Analyzing a broad spectrum of PFAS in the context of source contribution brings further novelty to understanding the regional human exposure to PFAS.

### *4.1. Global comparison of PFAS profile*

Present maternal serum concentrations of PFAS were low. PFOS and PFOA were far below the limit with no risk of adverse health effects (5 μg/L and 2 μg/L, respectively) according to Germany Environment Agency (Umweltbundesambandt, 2023). Less than a handful exceeded the action limit for women of childbearing age (10 μg/L and 5 μg/L, respectively).

Although the lowest maternal PFAS observed, the Argentinean detection frequencies aligned with the above-referred global studies and reflected the widespread distribution of PFAS. Clearly, the findings in our Argentinean samples demonstrate low maternal exposure. Variations in detection frequencies and concentrations across the countries likely reflect the production, regulations, and phase-out. In contrast to the Western world, but equal to Tanzania (Müller et al., 2019), Argentina has the status of a developing country, and to our knowledge, historically, there has been no PFAS production. North America was the first to introduce PFOS restrictions and phase-out from 2001, and Europe from around five years later (Brennan et al., 2021). Meanwhile, PFAS production has been shifted from the USA, Europe, and Japan to an extended production of PFOS and PFOA in China (Zhang et al., 2012) and Sulfluramid in Brazil (Guida et al., 2023; Löfstedt Gilljam et al., 2016). This might explain the lower profile in the USA (Zell-Baran et al., 2023) and Canada (Fisher et al., 2016; Workman et al., 2019) to Europe (Birukov et al., 2021; Cariou et al., 2015; Dereumeaux et al., 2016), and Asia (Lee et al., 2013; Wang et al., 2019), and the PFOA concentrations in China likely explained by local contamination (Jiang et al., 2014; Wang et al., 2015; Wang et al., 2019), and the10-fold higher PFOS in Brazil (Souza et al., 2020) than in Argentina. Further, the distinct PFAS profile in Inuits living in remote Greenland and Canada has been explained by contamination of the marine diet (Caron-Beaudoin et al., 2020; Long et al., 2015).

Considering the short half-life of days to one month for PFBA and PFHxA (EFSA, 2020; Luz et al., 2019), our high PFBA and PFHxA detection frequencies were unexpected. As in the comparison, both substances have, to a small degree, been analyzed in human populations and with infrequent detection, primarily undetected, as reviewed by others (Anderson et al., 2019; EFSA, 2020; Lee and Mabury, 2011). Given the low bioaccumulation, our findings indicate ongoing but low exposure in agreement with the dominance of aquatic biota in Argentina (Llorca et al., 2012).

### *4.2. Source appointment and contributing factor of PFAS concentrations*

Consequences of restriction and phase-out partly align with findings

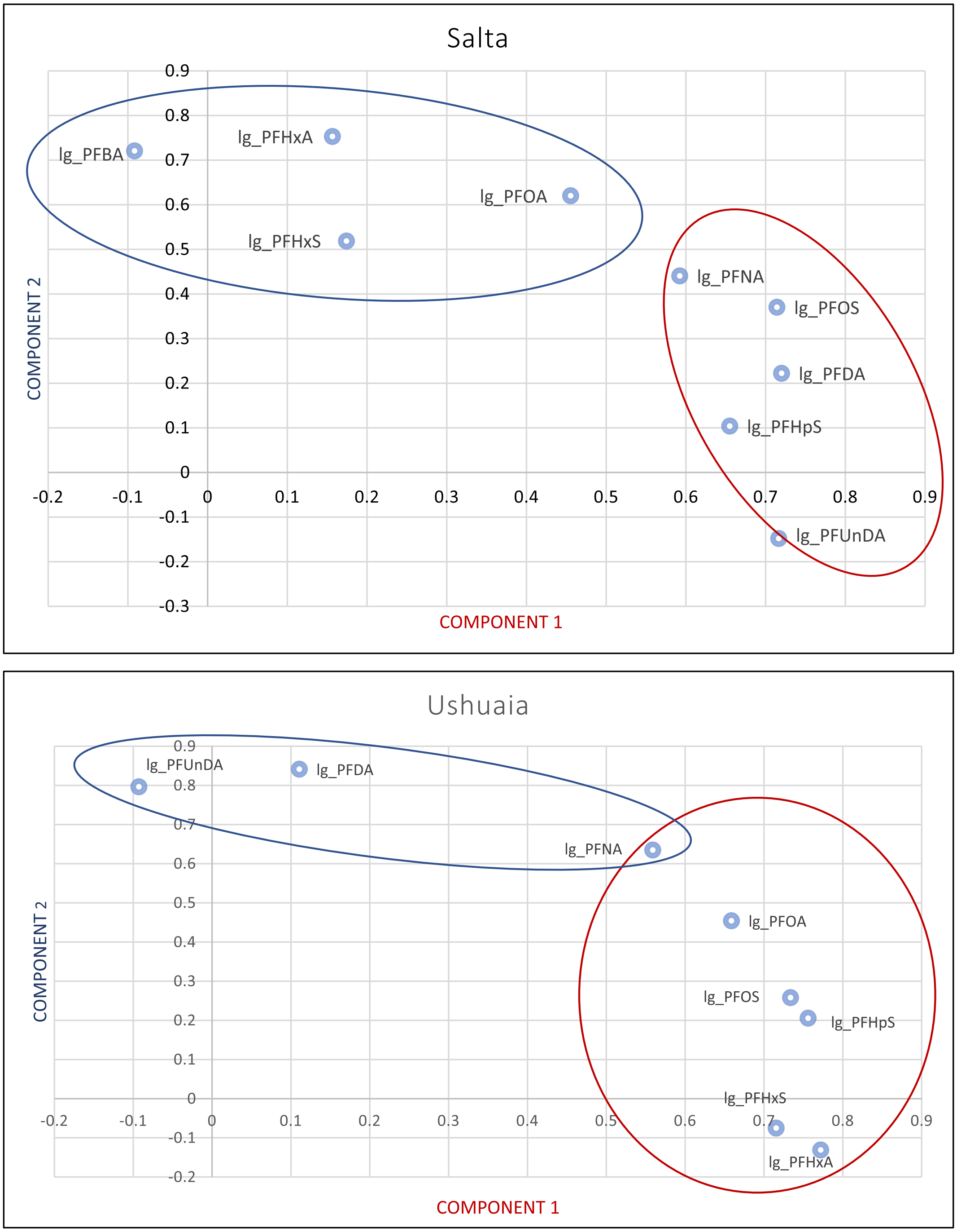


**Fig. 1.** Rotated component plot of log-transformed PFAS by region in the EMASAR study, 2011–2012.
Extraction Method: Principal Component Analysis. Rotation Method: Varimax with Kaiser Normalization. Rotation converged in 3 iterations and strong or moderate factor loadings >0.5 are visible in the plot. For details of the analyses, see Table S5. For abbreviations of the PFAS, see Table S1.

from the PCAs with source compounds dominated by short-chain substances used as replacements for legacy PFAS. The region-specific loading patterns of PFAS had distinct contributions (80–90 %) to the specific sources. In Ushuaia, compound 1 was a mixture of replaced and legacy substances across functional groups and chain length (Buck et al., 2011). The dominance of PFHxA, PFHpS, and PFHxS might indicate exposure from consumer products. As indicated by others, the dominance of short-chain substances likely reflects the shift from legacy PFAS to its replacements (Pérez et al., 2014). Short-chain PFAS have been substituted in consumer products, which significantly affects human uptake through ingestion of dust and drinking water (EFSA, 2020; Zheng et al., 2023). Also, due to the legislative control of PFOS and PFOA and the lower bioaccumulation of PFOA, these two substances may indicate the preference for terrestrial animal food. Thus, dietary contribution seems plausible. The secondary source with solely long-chain PFCAs is suggested to fish intake. Bioaccumulation increases by chain length, with the highest bioaccumulative ability in aquatic species (i.e., freshwater > marine origin) compared to terrestrial species (Augustsson et al., 2021). Similarly, PFCA clusters with C9–11 were revealed in a North Atlantic population with high seafood consumption (Hu et al., 2018). In Salta, compound 1, with the dominance of long-chain substances, is likely to reflect diet as the source and preferably diet with freshwater fish due to the long-chain PFCAs. For cluster 2, the strong dominance of short-chain PFBA, PFHxA, and PFHxS combined with

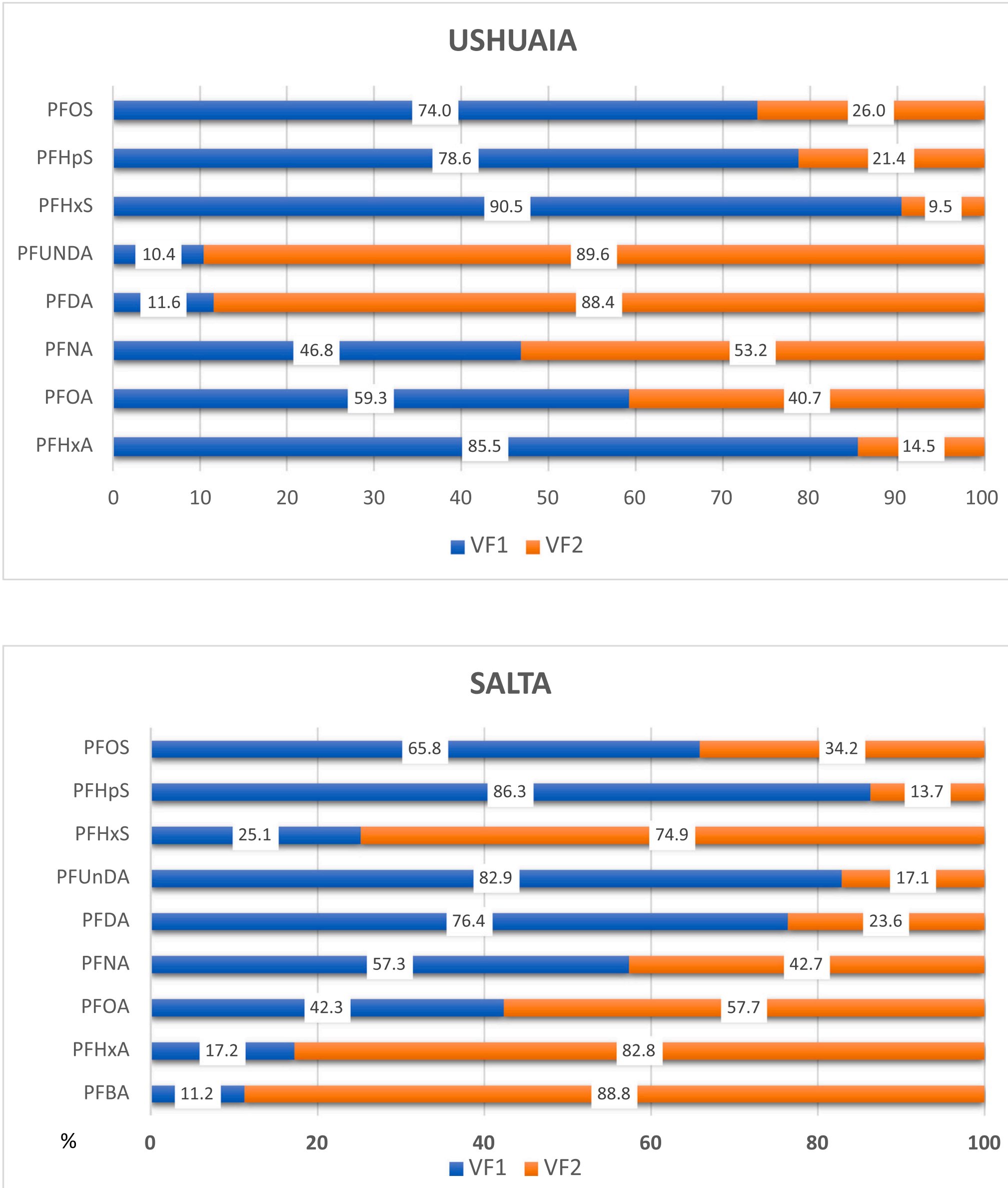


**Fig. 2.** Individual PFAS contributions in % of each factor loading from the PCA analyses through a multiple linear regression model by region. Percentage calculation based on the equation: i (%) = 100 * (Bi / ΣniBi).

PFOA have a shared affinity to water, and they are linked to consumer products (Domingo and Nadal, 2019; Zheng et al., 2023). Thus, with reservations, this distinct profile of component 2 might suggest low-grade polluted drinking water.

Contributing beyond suggested sources needs to be clarified. Considering the low concentrations, local pollution is of less importance. Known from industrialization and urbanization and pollutions elsewhere are e.g., industry, releases of aqueous film-forming foams (AFFFs) from airport firefighting activities, wastewater discharges, and pesticides from agriculture – i.e., leading to polluted drinking water as well as freshwater fish (Kurwadkar et al., 2022; Wee and Aris, 2023). As previously described, there have been identified general risks to water safety in Salta (Seghezzo et al., 2013), and growing urbanization with expanding activities in Ushuaia (Diodato et al., 2020; Ferreira et al., 2021).

### 4.3. Determinants of PFAS concentrations

After controlling for potential influential factors, regional differences for several PFAS exposures remained. Also, as previously described, there are variations between Ushuaia and Salta concerning latitudes and climate, human activity, economy, and demography (Okland et al., 2017).

Dietary fish intake is considered a major human source to PFAS, with freshwater fish dominating marine origin (Augustsson et al., 2021). In our model, freshwater fish increased PFOS and PFDA. As reviewed by Sunderland et al. (2019), the contribution of fish intake to PFAS is related to the significance of a population's diet. Relative to the high meat consumption, the freshwater fish found as a predominantly PFAS source likely reflects polluted aquatic habitat (van Asselt et al., 2011). The fact that marine intakes were restricted to Ushuaia might have attenuated the results as we observed marine products with borderline association for PFNA and PFHxA. Also, self-reported food frequency questionnaires (FFQ) and their form could be susceptible to bias and

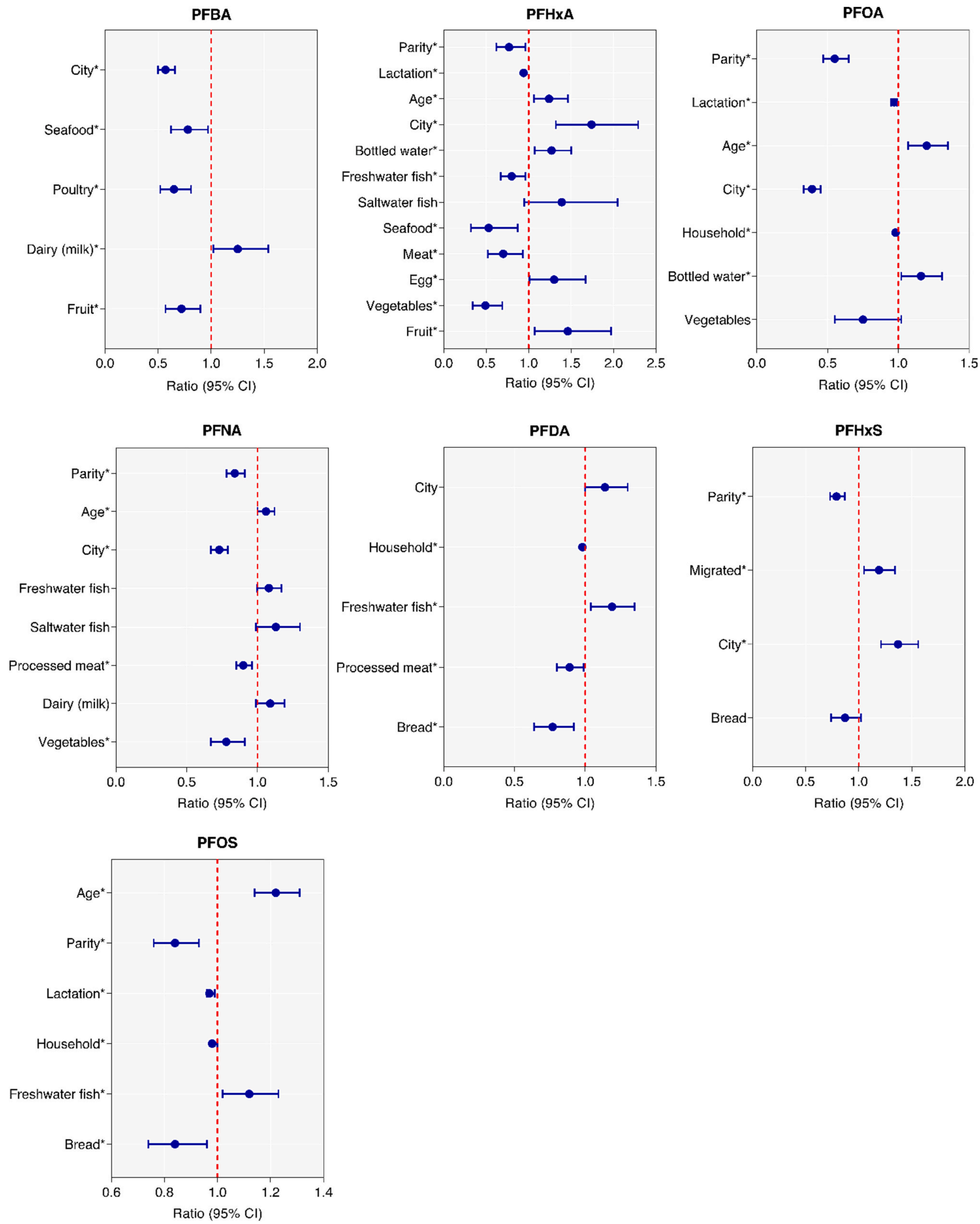


**Fig. 3.** Results of linear regression model with robust standard errors of maternal log10 transformed-PFAS (ng/mL) serum concentrations with region, socio-demography, obstetric history, and diet as predictors. The EMASAR study in Argentina, 2011–2012.

Backward regression with initial variables: age (10 years), parity (monoparous (ref) - multiparous), lifetime lactation (6 months), region (Ushuaia (ref) – Salta), migration (no-yes), people in household (number), education (primary/secondary – tertiary/university), bottled water daily (no-yes), dietary factors during pregnancy (never/seldom ((ref) – weekly/daily): freshwater fish, saltwater fish, seafood, meat, poultry, processed meat, egg, dairy products (milk), butter/cheese, fruit, bread, vegetables.

* The p-value of the selected predictor is $< 0.05$

limited accuracy (Poothong et al., 2020), as also observed in populations with high fish intake (Caron-Beaudoin et al., 2020). Moreover, the distinct pattern revealed in PCA-MLR analyses with the cluster of PFNA, PFDA, and PFUnDA might underpin the importance of marine fish intake for Ushuaia.

The contribution of specific dietary items to PFAS body burden is reported with dependency on carbon chain length and dietary patterns within geographical variations (Jian et al., 2017; Kärrman et al., 2009; van Asselt et al., 2011), which was observed in our models. Also, lower tropic items were inversely related to long-chain PFAS, as animal origin was due to more short-chain substances detected. Besides, our inverse dietary effect on PFAS may reflect a specific pattern with less contamination of the particular substance (Halldorsson et al., 2008; Tian et al., 2018).

Regarding the associations with bottled water, PFAS has infrequently been detected in bottled water, explained by the grade of contamination in water sources, and plastic, (Kaboré et al., 2018; Wee and Aris, 2023), or due to specific production/filling processes (Eschauzier et al., 2013). The underlying factor of household members` negative impact on PFOA, PFOS, and PFDA is unclear. However, it might reflect low socioeconomic status over the years with less access to consumer products or food items (Buekers et al., 2018), while others have linked to frequent cleaning (DeLuca et al., 2023). Migration is explained by pre-exposure or habits related to origin. Age and pregnancy-related factors align as known predictors (McAdam and Bell, 2023).

### *4.4. Strengths and limitations*

Our study encompassed a broad range of PFAS, providing a comprehensive exposure assessment in a relatively large study group. An expansive suite of PFAS is rarely present in human studies (De Silva et al., 2021; EFSA, 2020). The chemical analyses conducted in the study were highly quality and validated through participation in the Arctic Monitoring and Assessment Programme (AMAP) Ring Test (INSPQ, 2023). Standard sampling time and procedures were followed during this study by trained health-care professionals, ensuring consistency and comparability of results. The present study also has some limitations. Results for PFBA must be interpreted with care due to a limitation in analysis and only one available transition for identification (Huber and Brox, 2015). However, since the detection frequency for PFBA was outstandingly different compared to other previously analyzed and reported cohorts, this substance was worth mentioning in this publication. Next, the study was limited to two specific regions with unique socioeconomic characteristics, which may restrict the generalizability of the findings to other populations in Argentina. Moreover, the statistical analyses in the MLR may be subject to biased standard errors due to the violation of homogeneity of variance. However, robust common errors were implemented to mitigate this concern. Regarding the study design and the weakness mentioned, the findings revealed are indicators of associations, not causalities.

## 5. Conclusion

This study addresses a knowledge gap by investigating maternal PFAS exposure in pregnant women from two distinct geographical areas in Argentina. Low PFAS concentrations were detected, representing the lowest observed in a global comparison within the same period. Regional diversity in PFAS exposure was observed, and potential exposure sources were suggested to reflect the global regulations. In line with previous research, dietary predictors were compound-specific, and age and pregnancy-related factors were regulators. Considering the need for human PFAS information and temporal trends in South America, further maternal monitoring in Argentina and South America is warranted.

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## CRediT authorship contribution statement

**Solrunn Hansen:** Writing – original draft, Visualization, Validation, Methodology, Formal analysis, Conceptualization. **Shanshan Xu:** Writing – review & editing, Visualization. **Sandra Huber:** Writing – review & editing, Formal analysis. **Marisa Viviana Alvarez:** Writing – review & editing. **Jon Øyvind Odland:** Writing – review & editing, Methodology, Investigation, Funding acquisition, 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.

## Data availability

The authors do not have permission to share data.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.170096.

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