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1 **Passive sampler derived profiles and mass flows of Perfluorinated Alkyl Substances (PFAS)**
2 **across the Fram Strait in the North Atlantic**

3
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11
12 **ABSTRACT**

13 Per- and polyfluorinated alkyl substances (PFAS) are a family of pollutants of high concern due
14 to their ubiquity and negative human health impacts. The long-range marine transport of PFAS
15 was observed during year-long deployments of passive tube samplers in the Fram Strait across
16 three depth transects. Time weighted average concentrations ranged from 2.4-360 pg L⁻¹, and
17 10 different PFAS were regularly observed. PFAS profiles and concentrations were generally
18 similar to those previously characterized for polycyclic aromatic hydrocarbons (PAHs) at these
19 sites. The detection of several anionic PFAS in “old” water demonstrated that they are not
20 perfect water mass tracers, but are also transported to depth via settling particles. Mass flows
21 of PFAS through the Fram Strait in and out of the Arctic Ocean were basically similar (112 ±82

22 Mg year⁻¹ northward flow, 100 ±54 Mg year⁻¹ southward flow). For FOSA, export from the Arctic
23 Ocean via the Fram Strait exceeded import by Atlantic Water, likely due to preferential transport
24 and deposition in the Arctic Ocean. These observations suggest PFAS in the Arctic are governed
25 by the feedback loop previously described for PAHs in the region – with additional atmospheric
26 transport delivering volatile PFAS to the Arctic, which then get exported further.

27

28 **Synopsis:** Year-round passive sampling derived concentrations and mass flows suggest that the
29 Arctic Ocean is already exporting similar concentrations of PFAS to those are entering it via the
30 Fram Strait.

31

32 **Keywords:** PFAS, Fram Strait, PFAS Export, Long Term Monitoring, Arctic, POPs, Passive Sampling

33

34

35 **Introduction**

36 One of the defining characteristics of persistent organic pollutants (POPs) is their ability
37 to undergo long range transport.¹ Per- and polyfluorinated alkyl substances, or PFAS, are a
38 specific POP class of great concern due to their negative human health effects and global
39 presence, even in remote regions including the Arctic.²⁻⁸ Due to this long range transport and
40 ability to bioaccumulate, great effort has been put into characterizing the impact of PFAS in the
41 Arctic.^{5-7,9-14} While many previous studies have detected the most common PFAS,
42 perfluorooctanoic acid (PFOA) and perfluorooctanoic sulfonic acid (PFOS) in water, sediment,
43 and biota, there is new concern about their replacements as industry has phased out PFOA and

44 PFOS.^{6,9,13} Past research has shown increases in legacy PFAS deposition to the Arctic are tied to
45 increases in production, something that may be replicated again with replacement compounds
46 such as 6:2 fluorotelomer alcohol (FTOH), perfluorobutane sulfonate (PFBS), and other
47 compounds.^{5,8,9,12}

48 Understanding the fate and transport of PFAS to the Arctic is made further difficult by
49 the region's remoteness. However, the Fram Strait, which has been previously characterized for
50 other POPs, offers a unique opportunity to study PFAS dynamics through this "gateway of the
51 Arctic".¹⁵⁻¹⁷ Previous research has observed a feedback loop through the Fram Strait, where
52 similar mass flows of POPs are imported from Atlantic Water masses in the eastern Fram Strait
53 and exported from the Arctic Ocean to the North Atlantic in western parts of the strait.¹⁶
54 However, there has been observations of depletions with depth for all POPs in the Fram Strait,
55 potentially caused by degradation processes that govern some legacy POPs, or might reflect
56 changing emissions over time .^{15,16} Due to inherent persistence of perfluorinated acids, they
57 constitute an ideal tracer to further examine the complex interplay of the hydrological
58 processes intersect with chemistry and biology at depth.^{12,15-18}

59 Ample evidence suggests the dominant sources of PFAS in the remote northern
60 hemisphere are via long range transport from emissions to air, and subsequent atmospheric
61 deposition, coupled with oceanic long-range transport .^{12,13,19-21} Prior studies have highlighted
62 the presence and changing composition of PFAS contamination in the Atlantic and Arctic Ocean.
63 For example, a modeling study by Zhang et al. (2017) suggested that the import of PFAS into the
64 Arctic Ocean with Atlantic water was greatest around 2000, and has since decreased, with a few
65 tons PFOS being imported annually, though another study suggests transport to the Arctic for

66 PFAS remains consistent or increasing.^{10,22} One way to address these dynamic conditions is to
67 rely on detection tools, such as passive sampling, that can be used for long deployments in
68 these remote regions to derive representative concentrations and profiles.^{9,20,23,24} In particular,
69 a previously validated tube passive sampler design containing a microporous membrane filled
70 with sorbent was deployed.²⁵⁻²⁷ Hence, the aims of our study were to (i) test passive samplers
71 as a sampling tools across different sites and water depths in the Fram Strait; (ii) compare PFAS
72 profiles and concentrations to legacy POPs from the same sites, and (iii) infer import and export
73 of PFAS from the Arctic Ocean, and assess whether the underlying sources are changing.

74

75 **Methods and Materials**

76

77 **Chemicals and reagents.** Liquid chromatography-grade methanol (LC-MeOH), and water
78 (LC-water) were purchased from Fisher Scientific (New Hampshire, USA) along with ammonium
79 hydroxide (NH₄OH), ammonium acetate (C₂H₇NO₂), ACS-grade ethanol (EtOH) and ACS-grade
80 methanol (MeOH). Analytical standards were used to create native compound standards from
81 the Wellington PFAC-30PAR mix plus an additional four analytical compounds (Table S1). Mass
82 labeled surrogate solutions were comprised the Wellington Laboratories (Canada) MPFAC-24ES
83 plus an additional three of their mass labeled compounds (Table S1).

84

85 **Long-Term Deployments and Sampling Details.** Single tube passive samplers that have
86 been previously validated were deployed at 3 different monitoring locations of the LTER (Long-
87 Term Ecological Research) observatory HAUSGARTEN in the Fram Strait at 4 depths each (Figure

88 1).^{25,28} Passive samplers were deployed for just over one year (392-413 days) and discrete water
89 samples were collected alongside the recovery of these passive during *RV Polarstern* Cruise
90 PS131 in the summer of 2022. Water properties including average salinity, directional velocity,
91 and temperature with depth were reported from long term monitoring mooring sites as
92 described in previous research.¹⁶ Sites HGIV and F4 were deeper transects on the Eastern end of
93 the Fram Strait (>1000 m), while site EGC sampled shallower (<900 m) waters of the East
94 Greenland Current. For more site details, see Table S2.
95



Figure 1. Sampling locations within the Fram Strait monitored from 2021 – 2022 using passive samplers.

97 **PFAS Sample Analysis.** Discrete water samples of approximately 1000 mL were shaken,
98 weighed, and spiked with 4 ng of isotopically labeled internal standard followed by offline weak
99 anion exchange (WAX) solid phase extraction (SPE), in accordance with previously published
100 methods.^{25,29} A matrix spike experiment was performed to assess the recovery of a native PFAS
101 spike in tap water (Table S3). Passive samplers were spiked with internal standards and
102 extracted with MeOH following previously published methods with slight modifications,
103 including the addition of a third 24-hour extraction sequence, to yield a final extract volume of
104 18 mL that was then concentrated to 0.5 mL under a gentle stream of nitrogen at 40 °C. The
105 passive sampler assembly has been reported earlier during the sampler's validation.²⁵ For
106 calculation of time weighted averages from passive sampler profiles, see SI section S2 and tables
107 S9-10.

108 **Quality control and instrumental analysis.** Method detection limits (MDLs) were
109 calculated from laboratory and travel blanks collected for passive sampler (10-3300 pg sampler⁻¹)
110 and solid phase (10-3900 pg L⁻¹) extraction procedures. The median and three times standard
111 deviation of the blank concentrations were summed to determine MDLs (see Table S4). Method
112 recoveries was evaluated using mass labeled surrogates purchased from Wellington, including
113 their MPFAC24 mixture and 3 additional single compounds (Table S5-6). Recoveries of mass
114 labeled standards ranging from 60-140% were accepted, with an exception made for
115 Perfluoroundecanoic acid (PFUnDA) which had an average recovery of 55% due to losses during
116 the extraction procedure (for details, see SI). The instrumental analysis was performed using a
117 SCIEX Exion LC AC UHPLC system coupled to a SCIEX X500R quadrupole time-of-flight tandem
118 mass spectrometer (QTOF MSMS). For additional details on instrumentation, see SI section S1.

119 **Estimation of PFAS mass flows.** Estimation of PFAS mass flows (Mg per year)
120 across the Fram Strait were calculated as described previously, with water masses assigned
121 based on location and depth to Polar Water (PW), Atlantic Water (AW), and Re-circulating
122 Atlantic Water (RAW).¹⁶ This approach was employed at the same three sites and depth profiles
123 characterized in Zhang et al. (2023).¹⁶ Detected PFAS profiles were then integrated along the
124 depth transect of the water mass to derive a mean concentration for each compound using the
125 `castr` package in R from depths of 0 to 400 m for PW, and 0 to 840 m for RAW and AW, as
126 described previously.¹⁸ For further details on the calculation and method, see the SI section S3.

127

128 **Results**

129 **Passive Sampler Uptake.** A total of 10 PFAS were detected at least once in passive
130 samplers above MDLs, at all 3 sites. These compounds displayed a general decrease in
131 accumulated mass with depth, ranging from 0.01-1.9 ng sampler⁻¹ for each individual
132 compound (Fig S1). PFOA and PFOS were present at the highest amounts across all sites (0.14-
133 1.9 ng sampler⁻¹), but all compounds were generally detected at the same frequency across
134 depth and space (Table S7). No HFPO-DA (“Gen X”) was detected in the passive samplers or
135 water grabs from this study, despite previous detection.¹² Passive sampler percent of
136 equilibrium reached (i.e. progress for PFAS to reach thermodynamic equilibrium between the
137 passive sampler and the surrounding seawater) ranged from 1.0 - 54% across all 10 compounds,
138 decreasing with chain length (Table S9). Other compounds aside from the 10 reported had no
139 peaks detected or were below MDLs. When one of these 10 PFAS was found to be <MDL, a
140 replacement value of the $MDL/\sqrt{2}$ was used instead for calculating sum PFAS or integrated

141 concentration with depth. As displayed in a previous study, it is unlikely that biofouling in the
142 shallower sites impacted uptake.²⁵

143 **Seawater grab results.** Most PFAS in the grab samples were below the MDL. The only
144 compound detected above its MDL greater than 75% of the time was perfluorooctane
145 sulfonamide (FOSA), for which concentrations ranged from 18 - 140 pg L⁻¹ (Table S8). The next
146 most prevalent compound was the long chain nonanoic carboxylate, PFNA, which was detected
147 in 42% of samples above MDL, and ranged from 13 - 17 pg L⁻¹. Overall, calculated MDLs ranged
148 from 10 pg L⁻¹ to 3900 pg L⁻¹, displaying how low PFAS concentrations are in these remote
149 waters (Table S4).

150 **Time Weighted Average Concentration and Profiles.** Derived time weighted average
151 concentrations ranged from 2.4 - 360 pg L⁻¹, with an average propagated uncertainty of ±39%
152 (for details see SI section S2). Detection was dominated by C5-C8 perfluorocarboxylic acids
153 (PFCA), and C4-C8 perfluorosulfonic acids (PFSA) including PFOA and PFOS (Fig 2). There was a
154 general decrease in concentration with depth aside from a few exceptions, similar to prior
155 observations for the same compounds previously studied in Arctic waters.¹² Detection of PFAS
156 decreased in particular for passive samplers deployed below 250 m, though PFOS and PFOA
157 remained detectable (Fig 2). For reference, the MDL values for detection in passive samplers
158 equate to limits near 20 pg L⁻¹ when converted to time weighted average concentrations.

159 Site HG-IV, in the eastern Fram Strait, displayed a unique depth profile of PFAS
160 concentrations: higher PFAS concentrations were present at a depth of 415 m than were found
161 at the shallowest (198 m) depth, likely due to the sinking of warmer Arctic waters already
162 enriched in PFAS, when they encounter colder Polar water (Fig 2).¹⁶ Another exception to the

163 decrease with depth was the observation of PFHxS at site F4, which reached a maximum
164 concentration at a depth of 498 m, potentially liberated from settling particles. A similar profile
165 has been previously observed at this site for polycyclic aromatic hydrocarbons (PAH).¹⁶ The PFAS
166 observed in this study suggest that sources of PFAS to the Fram Strait and Arctic are increasingly
167 influenced by replacement compounds including shorter-chain FTOHs and PFBS. While legacy
168 compounds such as PFOA, PFNA, PFHxS, PFOS were all still present at high concentrations
169 relative to other studies in the region, PFBS (a short chain PFSA) as well as PFPeA, PFHxA, and
170 PFHpA (short chain PFCA) were present at similar concentrations (Table S11).^{12,22,23,30,31} Principal
171 component analysis was used to examine if water parameters such as depth, temperature, or
172 salinity influenced PFAS profiles (Fig S2). Only depth strongly impacted PFAS profile
173 composition, with deep (>800 m) samples clustering away from the 64-500 m samples (Fig S2).
174 Results from passive and discrete grab samples are discussed in the SI section S4. In addition,
175 the PFHpA/PFOA ratios derived and whether it can indicate atmospheric deposition can be
176 found in SI section S5.

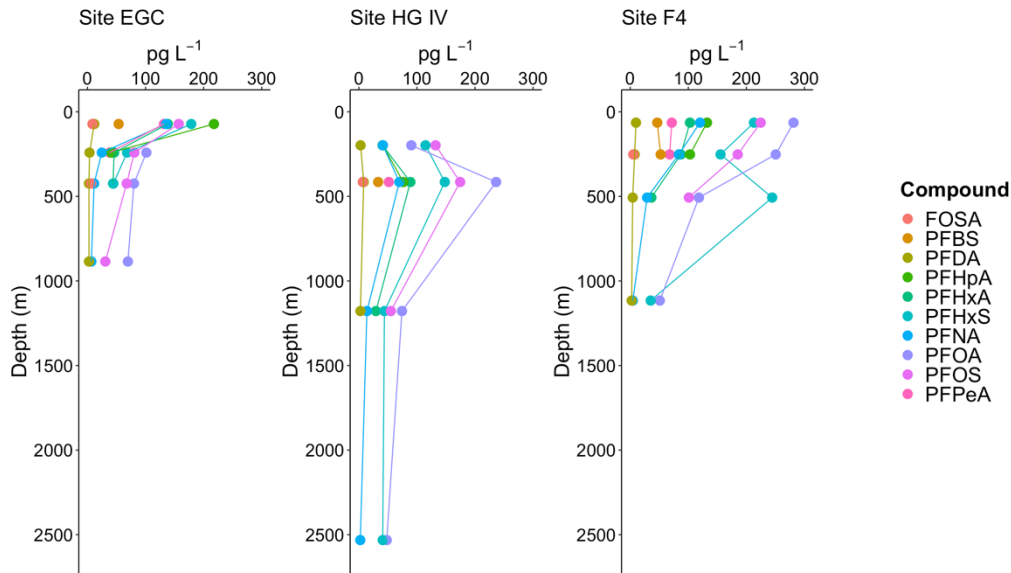


Figure 2. Time weighted average concentration profiles across depth in the Fram Strait.

178 **Comparison to Legacy POP Profiles.** Passive sampler-derived PFAS concentrations and
 179 depth profiles were generally similar to those reported for PAHs at the same study sites, despite
 180 different physio-chemical properties and reactivities between the two groups.¹⁶ Unlike PAHs,
 181 PFAS do not undergo the same depth-depletion via microbial degradation that has been
 182 suggested previously.^{16,32} PFAS profiles, like those reported for PAHs, differ from depth profiles
 183 for polychlorinated biphenyls (PCBs) and organochlorine pesticides, which have displayed higher
 184 concentrations within the intermediate and deep waters of Fram Strait, likely due to their
 185 earlier ban/ phase outs and possible settling on particles (Fig S3).³³ For further discussion,
 186 please see SI section S6.

187

188 **PFAS and water mass age.** Prior work established mean estimated water mass ages
 189 across the Fram Strait, which ranged from modern (last 50 years) for the top 500m to around

190 50-100 years at 1000 m to about 250 years for waters at 2,000 m depth.¹⁸ The detection of
191 several PFAS at depth clearly shows that even the anionic PFAS, which have been produced for
192 several decades, are not perfect water mass tracers, but are also transported to depth via
193 settling particles.³¹

194

195 **PFAS mass flows through the Fram Strait.** In the Fram Strait, the highest export
196 of Σ_{10} PFAS was observed in the Atlantic Water (AW) with a mass flow of 112 Mg year⁻¹ (± 82)
197 northwards into the Arctic Ocean (Fig 3). The Recirculating Water and Arctic Atlantic Water
198 (RAW/AAW) and Polar Water (PW) both had southward PFAS mass flows to the North Atlantic
199 Ocean of -66 (± 36) Mg year⁻¹ and -33 (± 19) Mg year⁻¹ respectively (Fig 3). These values are
200 larger than mass flows reported earlier for sum PAHs, sum OPEs, and sum PBDEs in the Fram
201 Strait.^{15,16,33} While no PCB mass flows have been reported for these same sites in the literature,
202 their concentrations were, at times, multiple orders of magnitude below the PFAS reported in
203 this study, suggesting PFAS may constitute the largest contaminant mass flows across the Fram
204 Strait that is currently known.¹⁷ There are likely seasonal fluctuations in PFAS concentration and
205 water mass volumes in the Fram Strait, that are difficult to quantify. We hence relied to annual
206 mean concentrations, and literature estimates of water mass fluxes.

207

208 Σ_{10} PFAS mass flows across the Fram Strait were very similar in both directions, with +112
209 and -100 Mg year⁻¹ estimated in this study. While there was good agreement for sum PFAS
210 between Arctic import and export (within 20%), the difference ranged from 1-74% between
211 inputs and exports for most individual PFAS (Table S12). This balance between inputs and

212 outputs was also noted in other studies for both PFAS and PAHs in the Fram Strait.^{12,16} Once
213 again, this overall balance between inputs and outputs was also observed when using a
214 traditional mass flux approach, albeit with more discrepancies between individual compounds'
215 North and South fluxes as these results are uncorrected for total water volume (Table S13). Only
216 FOSA displayed a wider discrepancy (74% difference) with approximately twice as much being
217 exported from the Arctic Ocean via the Fram Strait as was entering with Atlantic Water (0.56 Mg
218 year⁻¹ versus -1.0 Mg year⁻¹). This likely represents the preferential transport, deposition and re-
219 mobilization of the volatile FOSA in the Arctic Ocean. For the anionic PFAS, in contrast, very
220 similar mass flows were estimated (Fig 3). The estimated PFAS mass flows were similar to prior
221 estimates in the Fram Strait derived from grab samples.¹² The slight increase in export/input
222 estimated in this study relative to Joerrs et al. (2020) could be due to the time weighted average
223 nature of the data collected by these passive samplers.¹² Furthermore, these mass flow
224 estimates rely on yearly average water concentrations derived from passive samplers, with their
225 39% uncertainty, as well as the uncertainty associated with the transport volumes of each water
226 mass reported in literature, which ranged from 54– 73%.¹⁸

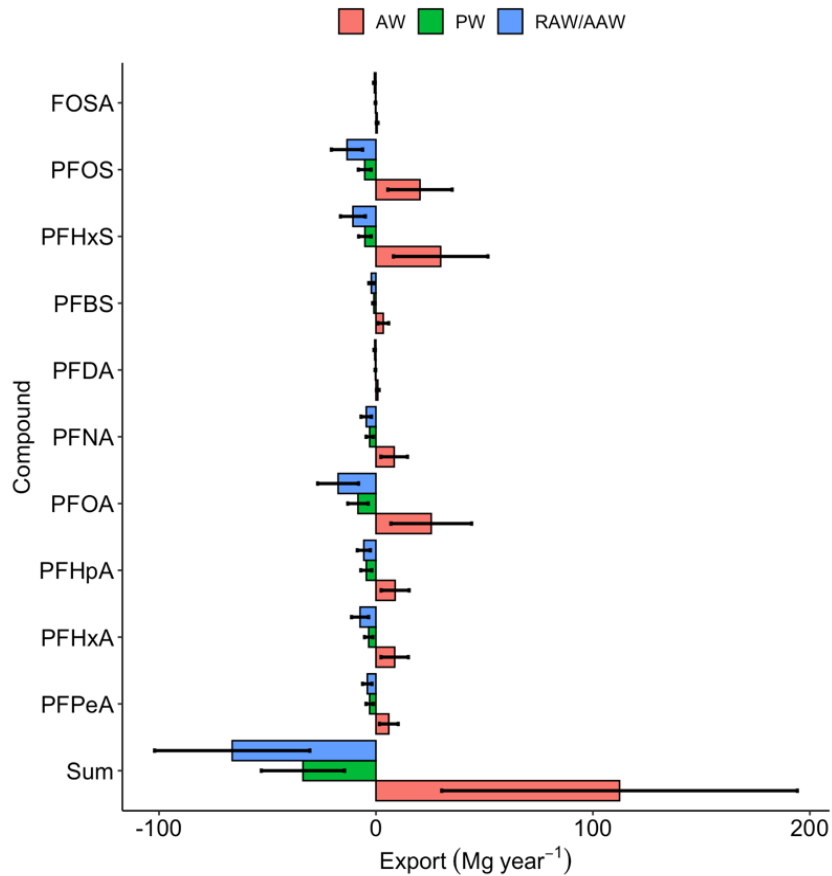


Figure 3. Estimated mass export of individual PFAS compounds in Atlantic Water (AW), Polar Water (PW), and Recirculating Atlantic Arctic Water (RAW/AAW).

227

228

229 **Outlook.** This study demonstrated the ability of passive samplers to overcome detection

230 limits of PFAS in remote, low concentration marine environments. The time weighted average

231 concentration derived in this study show that PFOA and PFOS were still present at high

232 concentrations in Arctic water masses, but shorter chain compounds (PFPeA, PFHxA, PFBS) were

233 also routinely present across the Fram Strait.

234 The detection of shorter chain PFCAs in the Arctic water masses may reflect industry's
235 switch to shorter chain replacements (i.e., PFBS replacing PFOS or 6:2 FTOH replacing 8:2
236 FTOH).^{5,8,21,34} A comparison of PFAS to legacy POP profiles indicated similar trends across the
237 Fram Strait with depth and across sites, implying that similar oceanographic processes are
238 mostly responsible. Given the persistence of perfluorinated acids, this would also imply that
239 biodegradation was of limited importance as a removal mechanism for those legacy POPs and
240 PAHs.

241 Overall, our results demonstrate that PFAS are circulating in the Arctic Ocean and North
242 Atlantic water masses in a feedback loop, as was previously suggested for PAHs.¹⁶ This feedback
243 loop consisted of the continued release of volatile PFAS at mid-latitudes that are transported to
244 high latitudes, where they are buoyed by smaller, local secondary sources, and are then re-
245 exported back to the North Atlantic Ocean.¹⁶ In the case of PFAS, this transport is more
246 complicated, as the volatile polyfluoroalkyl compounds may degrade into stable anionic
247 perfluoroalkyl acids in the high northern latitudes, and are then exported back to the Atlantic
248 Ocean.

249

250 **Supporting Information**

251 The Supporting Information is available free of charge on the ACS Publications website at DOI:
252 It contains a word document with additional analytical details, methods, standards, quality
253 control and assurance, LC/MS conditions, details on field deployment sites and conditions, and
254 calculation of time weighted averages. In a separate excel sheet, tables containing information

255 on partition coefficients for passive samplers, and further details on calculations of mass flows,
256 time weighted average concentrations, and discrete results can be found.

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275

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