

# Global flux of perfluoroalkyl acids from glaciers in a warming climate

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28 **SUMMARY**

29 Climate warming is accelerating glacier melting, releasing human-made chemicals that  
30 have been trapped in glaciers for decades. Among these are perfluoroalkyl acids  
31 (PFAAs), highly persistent and toxic pollutants that threaten aquatic ecosystems,  
32 fisheries, and human health. Despite global efforts to curb PFAA emissions, their  
33 continued release from melting glaciers represents a legacy source that remains  
34 unquantified on a global scale. Here we combine field and literature data with machine  
35 learning and glacial mass balance model to estimate current and future PFAA fluxes via  
36 both dissolved and particle-bound phases. We find that global glaciers release  
37 approximately 3,500 kg of PFAAs annually, with suspended particles contributing  
38 around 12% of this amount. Projected trends suggest future release potential will  
39 rapidly boom through 2040 under extreme climate warming. These findings fill a  
40 critical gap in the global PFAA budget and underscore the urgency of coordinated action  
41 on both legacy pollutant management and climate mitigation.

42

43 **KEYWORDS**

44 perfluoroalkyl acids; glacier melt; release flux; machine learning; future trend

## 45 INTRODUCTION

46 In the Anthropocene, chemical pollution has become a defining challenge as novel  
47 entities for planetary health.<sup>1-3</sup> Among these, per- and polyfluoroalkyl substances  
48 (PFAS), known as “forever chemicals” due to their extreme persistent, have attracted  
49 widespread attention.<sup>4,5</sup> PFAS are a large class of human-made chemicals that have  
50 been widely used in industrial and consumer products since the mid-20th century.<sup>6</sup>  
51 Perfluoroalkyl acids (PFAAs) are the best-studied subgroup of PFAS, including  
52 compounds listed for phase-out under Stockholm Convention due to a series of adverse  
53 effects.<sup>7,8</sup> Alarmingly, even in remote regions, the concentrations of perfluorooctanoic  
54 acid (PFOA) in rainwater greatly exceed health advisory levels (4 pg L<sup>-1</sup>),<sup>9</sup> indicting  
55 the planetary boundary for PFAA pollution has been exceeded worldwide.<sup>10</sup>

56 Glaciers in the remote cryosphere are critical reservoirs for PFAAs. PFAAs can undergo  
57 long-range atmospheric transport and subsequently deposit onto glacial surfaces  
58 through both dry and wet deposition processes due to cold-trapping effect.<sup>11</sup> Measurable  
59 concentrations of PFAAs have been detected in glaciers in the Arctic,<sup>12</sup> Antarctica,<sup>13</sup>  
60 and high-altitude alpine regions,<sup>14,15</sup> ranging from hundreds to thousands of picograms  
61 per liter. As forever chemicals, PFAAs can persist in glaciers for decades, even after  
62 their primary emissions are controlled,<sup>16,17</sup> posing long-term threats to fragile  
63 ecosystems, particularly in sensitive regions such as the Arctic and the Tibetan Plateau.

64 Previously trapped PFAAs in glaciers are being released under the warming climate,<sup>18,19</sup>  
65 creating an emerging threat to proglacial ecosystems. Signals of PFAA meltwater  
66 sources have been identified in the contaminant profiles of sediment cores and in the  
67 biota from proglacial environments,<sup>20,21</sup> indicating the re-release of these previously  
68 accumulated chemicals. Once released, PFAAs can undergo biomagnification via food  
69 chain from benthic invertebrates to top predators,<sup>22,23</sup> thereby threatening aquatic  
70 diversity, fisheries and even human health. For example, high concentrations (~500 ng  
71 mL<sup>-1</sup>) were found in blood serum from East Greenland Inuit who rely on traditional  
72 marine food, despite relatively low environmental concentrations (0.0005 ng mL<sup>-1</sup>).<sup>24</sup>

73 Despite growing recognition of this emerging threat, several knowledge gaps remain.  
74 Most notably, we lack a comprehensive estimate of the magnitudes and spatial patterns  
75 of PFAA fluxes from glaciers on a global scale. Existing studies are limited to a few  
76 sampling sites. To date, only two studies report the release flux of PFAAs to the  
77 downstream: one reporting an annual flux of  $1.6 \pm 0.7$  kg in the Canadian High Arctic,<sup>25</sup>  
78 and another estimating of about  $0.5 \text{ kg year}^{-1}$  from a single glacier on the central Tibetan  
79 Plateau.<sup>26</sup> The lack of global flux data limits our understanding of how glacial melt  
80 contributes to global PFAA budget and hinders the identification of pollution hotspots.

81 Furthermore, current assessments of PFAA transport focus on dissolved phases while  
82 overlooking the contribution of suspended particles generated through glacial erosion  
83 and permafrost thawing.<sup>17,18</sup> These suspended particles, mobilized at increasing rates  
84 under accelerated glacial melting, may serve as important vectors for PFAAs, especially  
85 for more toxic long-chain compounds.<sup>19,20</sup> As climate warming accelerates erosion and  
86 sediment mobilization, the omission of particle-bound transport may result in  
87 underestimation of total PFAA fluxes and associated health risks.

88 Here we address these critical knowledge gaps by quantifying current and future flux  
89 of PFAAs released from glaciers worldwide via both dissolved and particle-bound  
90 particles. Our analysis includes a global dataset of field data from Tibetan glaciers and  
91 literature data from global glaciers, the application of machine learning and glacial mass  
92 balance model, and the prediction of release fluxes through the end of the 21st century.  
93 Our findings reveal that approximately 3,500 kg of PFAAs are released from global  
94 glaciers per year with Arctic Canada and South and Central Asia identified as major  
95 export regions. Suspended particles contribute around 12% of the total flux,  
96 highlighting their non-negligible impact. The release flux is projected to increase  
97 rapidly through 2040, leaving a narrow time window for effective actions. Our  
98 estimates and predictions are crucial for a better understanding of the role of glacial  
99 melting on PFAA cycling in the global cryosphere.

## 100 **RESULTS**

### 101 ***Methods summary***

102 To evaluate the global flux of PFAAs from glaciers, we posed three research questions:

103 (1) What are the magnitudes and spatial patterns of PFAA release fluxes from glaciers

104 on a global scale? (2) What is the role of the particle-bound phase in total PFAA export?

105 (3) How might these fluxes change under future climate scenarios?

106 To answer these questions, we first conducted field sampling on seven glaciers across

107 the Tibetan Plateau, measuring 13 individual PFAAs in both dissolved phase and

108 suspended particles. These field data were integrated into a global dataset comprising

109 739 samples from 49 glaciers across eight major glacial regions worldwide. Due to the

110 scarcity of concentrations in suspended particles, we developed predictive models for

111 log-transformed particle–water partition coefficients ( $\log K_d$ ), using both traditional

112 curve-fitting and machine learning approaches.  $\log K_d$  were predicted based on

113 environmental parameters (e.g., water temperature, particulate organic carbon) and

114 molecular descriptors of PFAAs. The best-performing machine learning model was

115 finally selected from multiple candidates based on its accuracy and stability.

116 We then coupled the predicted  $\log K_d$  with glacial runoff volumes derived from glacial

117 mass balance model to quantify annual release flux of PFAAs via both dissolved phase

118 and suspended particles. For regions with high export fluxes, we further identified

119 pollution hotspots using threshold-based spatial analysis of PFAA concentrations in

120 water and sediment. Finally, we integrated climate projections under three Shared

121 Socioeconomic Pathways (SSP126, SSP245, and SSP585) to simulate global glacial

122 PFAA exports from 2020 to 2100. This integrative framework enables a comprehensive

123 assessment of current and future glacier-sourced PFAA fluxes on a global scale.

### 124 **PFAAs in glacial meltwaters of the Tibetan Plateau**

125 To characterize the current status of PFAAs in high-altitude glacial regions, we

126 measured 13 individual PFAAs ([Table S1](#)) in meltwater and associated suspended

127 particles from seven glacier regions across the Tibetan Plateau. [Figure 1](#) illustrates the  
128 upstream-to-downstream arrangement of sampling sites, along with average  
129 concentrations and compositions of  $\Sigma_{13}$ PFAAs. The highest concentration (5430  $\text{pg L}^{-1}$ )  
130 <sup>1)</sup> was observed in the Rongbuk glacial region of Mt. Everest ([Figure 1B](#)), while  
131 concentrations at the other sites were generally below 1200  $\text{pg L}^{-1}$ . Across all sites,  
132 short-chain PFAAs, consisting of <C8 perfluoroalkyl carboxylic acids (PFCAs) and  
133 <C6 perfluoroalkyl sulfonic acids (PFSAs), dominated the PFAA profiles, accounting  
134 for 68–96% of the total concentrations ([Figure 1C](#)).

135 PFAAs released through glacier ablation can be transported downstream either in the  
136 dissolved phase or adsorbed onto suspended particles. We found that concentrations of  
137 most short-chain PFAAs in the dissolved phase increased from upstream to downstream  
138 sites ([Figure S1](#)), likely due to their higher solubility and mobility in water. In contrast,  
139 long-chain PFAAs ( $\geq$ C8 PFCAs and  $\geq$ C6 PFSAs) adsorbed onto suspended particles  
140 exhibited the opposite trend, with their concentrations decreasing from upstream to  
141 downstream ([Figure S1](#)). In other words, long-chain PFAAs associated with suspended  
142 particles tend to accumulate in the headwater regions. The opposing spatial trends of  
143 PFAA concentrations during transport ([Figure S1](#)) indicate the crucial need for accurate  
144 PFAA concentrations in both dissolved and particle-bound phase in glacier-fed systems.

### 145 **Occurrence of PFAAs in global glacial regions**

146 To place Tibetan data in a broader context, we compiled a global database of  $\Sigma_{13}$ PFAA  
147 concentrations from more than 49 glaciers, including field data from this study ( $n=59$ )  
148 and previous research ( $n=680$ ) ([Table S2](#)). [Figure 2](#) presents the locations of these  
149 glacial sites across eight major glacial regions, and corresponding average  $\Sigma_{13}$ PFAA  
150 concentrations (bold numbers) at each region. Pie charts depict the relative composition  
151 of individual PFAAs at each region.

152 Across the global dataset, PFAA concentrations in glacial meltwater ([Table S3](#)) ranged  
153 from several hundred to several thousand picograms per liter, exhibiting distinct

154 regional variation (Figure 2). Although the data exhibited fluctuations, the average  
155 concentrations of  $\Sigma_{13}$ PFAAs were relatively high (4486 pg L<sup>-1</sup>) in glacial regions in  
156 Arctic Canada (ACG) (Figure 2) [i.e., Lake Hazen watershed, see Table S2], and those  
157 near the Greenland Sea exhibited much lower levels (297 pg L<sup>-1</sup>). Similar to the Tibetan  
158 Plateau, short-chain PFAAs predominated in most regions, accounting for 62–80% of  
159 the total concentrations in areas such as ACG, Central Europe (CEG), and South and  
160 Central Asia (SCAG) (Figure 2, pie charts).

161 However, field data on PFAAs associated with suspended particles remain scarce in  
162 global glacial regions. To overcome this limitation, we applied particle-water partition  
163 coefficient ( $K_d$ ) to bridge the field-measured dissolved PFAA concentrations to particle-  
164 bound concentrations.

### 165 **Predicting particle–water partition coefficient**

166 The  $K_d$  describes the distribution of PFAAs between the dissolved and particle-bound  
167 phases. However,  $K_d$  values are highly variable and influenced by environmental  
168 parameters and molecular descriptors of PFAAs. Therefore, we selected key influencing  
169 factors, including temperature ( $T_w$ ), particulate organic carbon (POC),  $\log K_{ow-ionic}$ , and  
170 PFAA type, for predicting  $\log K_d$ . Details on selecting these factors and model validation  
171 procedures are provided in the Methods section.

172 Initial attempts to predict  $\log K_d$  using curve-fitting models yielded limited performance,  
173 with coefficients of determination ( $R^2$ ) ranging from 0.48 to 0.64 (Note S1). To improve  
174 prediction accuracy, we developed several machine learning (ML) models, including  
175 support vector machines (SVM), decision trees (DT), random forests (RF), and extreme  
176 gradient boosting (XGBoost). Among these, the XGBoost model (XGBoost-ML)  
177 outperformed others in both explanatory power and stability ( $R^2=0.95$  for the training  
178 set and  $Q^2=0.85$  for the test set). The best-performing XGBoost-ML showed strong  
179 agreement with field-measured  $\log K_d$  for all individual PFAAs in the training set  
180 (Figure 3A). Almost all of the predicted  $\log K_d$  in the test set fell within an order of

181 magnitude ( $\pm 1$  range) of the measured values, except one outlier (Figure 3B, white  
182 squares), which was measured with high POC values (up to 10%) (Table S4).

183 In summary, the XGBoot-ML outperforms traditional curve-fitting methods and other  
184 ML models in predicting  $\log K_d$ . With the combination of model-predicted  $\log K_d$  with  
185 field-measured dissolved PFAA concentrations (Table S2), the concentrations of  
186 PFAAs on suspended particles across the eight global glacial regions were calculated.  
187 The results are provided in Table S5.

### 188 **Global fluxes of PFAAs from glaciers**

189 Using dissolved and estimated particle-bound concentrations (Tables S3 and S6),  
190 combined with modeled glacial runoff volumes, we quantified the annual release fluxes  
191 of individual PFAAs from eight major glacial regions worldwide, with data listed in  
192 Table S6. Table 1 summarizes the release fluxes of  $\Sigma_{13}$ PFAAs via the dissolved phase  
193 and suspended particles for each region.

194 The total estimated global flux of  $\Sigma_{13}$ PFAAs was  $3,547 \pm 375$  kg year<sup>-1</sup>, with the  
195 dissolved phase contributing  $3,117 \pm 165$  kg year<sup>-1</sup> (~88%) and suspended particles  
196 accounting for the remaining  $430 \pm 210$  kg year<sup>-1</sup> (~12%) (Table 1). In most glacial  
197 regions, the contribution of suspended particles remained less than 15%. However, in  
198 sediment-rich systems such as the Greenland ice sheet (GIS), suspended particles  
199 contributed up to 51% of the total flux, likely due to high particle loads in glacial  
200 outflows.

201 Differences in the chain length of PFAAs influenced release fluxes in dissolved and  
202 particle-bound phases. In the dissolved phase, short-chain PFAAs dominated, with an  
203 estimated flux of  $1,823 \pm 97$  kg year<sup>-1</sup>, compared to  $1,129 \pm 61$  kg year<sup>-1</sup> for long-chain  
204 compounds (Table 7). In contrast, the particle-bound phase was enriched in long-chain  
205 PFAAs, which contributed  $238 \pm 63$  kg year<sup>-1</sup>, nearly three times the flux of short-chain  
206 compounds ( $87 \pm 14$  kg year<sup>-1</sup>) (Table 7), consistent with their stronger sorption  
207 affinities.

208 Substantial regional variations in total PFAA fluxes were also observed (Table 1). The  
209 SCAG region contributed the highest load, with an estimated flux of  $1394 \pm 86$  kg  
210 year<sup>-1</sup>, accounting for nearly 40% of global glacial PFAA export. This was followed by  
211 the ACG ( $698 \pm 48$  kg year<sup>-1</sup>), Arctic Europe (AEG) ( $564 \pm 25$  kg year<sup>-1</sup>), Western  
212 Canada and the USA (WCUG) ( $426 \pm 15$  kg year<sup>-1</sup>), and GIS ( $316 \pm 190$  kg year<sup>-1</sup>).  
213 Collectively, these five regions represented over 95% of the total global flux. High  
214 glacial runoff volumes, ranging from 212 to 547 km<sup>3</sup> year<sup>-1</sup> (Table 1), and relatively  
215 high PFAA concentrations (Figure 1) contributed to their dominant role.

216 In contrast, smaller glaciers such as those in North Asia (NAG) and Central Europe  
217 (CEG) exhibited limited annual PFAA fluxes ( $< 20$  kg year<sup>-1</sup>), due to both lower glacial  
218 runoff volumes (Table 1) and moderate PFAA levels (Figure 2). The Antarctic and  
219 Subantarctic Ice Sheet (AIS) also contributed modestly ( $118 \pm 10$  kg year<sup>-1</sup>), consistent  
220 with its relatively isolated location from PFAA emission sources.

### 221 **Priority locations of concern**

222 As mentioned above, the SCAG and Arctic regions present hotspot export zones for  
223 PFAA releases from melting glaciers. To further identify the locations of concern that  
224 should be prioritized for protection, we compiled the data on PFAAs in water and  
225 sediment from these regions (Tables S7 and S8).

226 The average concentration of  $\Sigma_{13}$ PFAAs in water (2 ng L<sup>-1</sup>) and sediments (1 ng g<sup>-1</sup>)  
227 were obtained and served as baselines for assessing priority locations of concern. Levels  
228 of concern are based on exceeding the average by 1, 2, and 3 times, respectively. Figure  
229 4 illustrates priority locations of concern for PFAAs with relatively high concentrations  
230 in water or sediment in the Arctic and SCAG regions. Red circles on the map indicate  
231 sites influenced by water, while purple circles represent those influenced by sediment.  
232 The higher the concentration of PFAAs, the larger the buffer zone. Detailed locations  
233 of each site are available in Tables S7 and S8.

234 In the Arctic, locations of concern are categorized into four main regions: Svalbard,

235 Arctic Europe, the Canadian Arctic basins, and the Chukchi Sea (Figure 4A). Svalbard  
236 exhibits elevated PFAA levels in both water and sediment, indicating a significant  
237 overlap in environmental risk. In Arctic Europe, areas of high concern are identified in  
238 coastal sediments along the Norwegian Sea and the Barents Sea. Additionally, Northern  
239 Swedish glaciers are classified as high-concern sites due to high PFAA concentrations  
240 in glacial runoffs. In the Canadian Arctic basins, such as Lake Hazen and Lake B35,  
241 impacts on local wildlife, including polar bears and Arctic foxes, have been observed  
242 in surrounding areas like Grise Fjord and Arviat (Figure 4A, orange triangle).  
243 Meanwhile, sediments near the coast of the Chukchi Sea indicate relatively high  
244 environmental risk.

#### 245 **Future release potential under climate warming**

246 Apart from regional impacts, understanding how the release of PFAAs from glaciers  
247 change under future climate warming is also imperative. Figures 5A and 5B illustrate  
248 the future release potential of  $\Sigma_{13}$ PFAAs from global glaciers in the 21<sup>st</sup> century via  
249 dissolved and suspended-particle phases under three shared socioeconomic pathway  
250 (SSP) scenarios, i.e., SSP126 (low emissions), SSP245 (intermediate emissions) and  
251 SSP585 (high emissions). The release potential refers to a predicted possibility of PFAA  
252 fluxes under future climate scenarios.

253 Under SSP126, the release potential of  $\Sigma_{13}$ PFAAs is expected to remain relatively stable  
254 by the end of the 21<sup>st</sup> century (Figures 5A and 5B, SSP126 scenario). In contrast, under  
255 SSP245—and especially under SSP585—the potential  $\Sigma_{13}$ PFAA fluxes exhibit  
256 increasing trends throughout the century. Under SSP585, the average release fluxes of  
257  $\Sigma_{13}$ PFAAs via the dissolved phase could reach up to  $\sim 13,000 \text{ kg year}^{-1}$ , and those via  
258 suspended particles may reach  $\sim 2,500 \text{ kg year}^{-1}$ , which represents a 6-fold increase  
259 compared to 2020 levels (Figures 5A and 5B, SSP585 scenario). By 2040, the release  
260 potential is expected to be less than double those observed in 2020; however, after 2040,  
261 the rate of increase is projected to boom rapidly. These results identify the next 15 years  
262 as a critical window for global efforts to mitigate PFAA pollution and combat climate

263 change.

264 Regionally, under SSP 585, ACG and GIS are predicted to experience a nearly 10-fold  
265 increase in the release potential of dissolved PFAAs by the end of this century. In  
266 contrast, WCUG, AEG, SCAG and AIS are expected to have about a twofold increase  
267 (Figure S2). For the suspended-particle phase, the release potential in ACG and GIS is  
268 projected to increase by ~8-fold, while WCUG, AEG, SCAG and AIS are anticipated  
269 to experience about a twofold increase (Figure S3).

270 However, these estimates may be conservative, as they assume that the rate of sediment  
271 transport—and thus particle-associated PFAA flux—remains constant under future  
272 climate warming. Actually, glacial sediment export has already doubled over the past  
273 30 years in regions such as the Arctic coastline and the Himalayas.<sup>27</sup> If this trend  
274 continues, doubling every 30 years, the release potential of  $\Sigma_{13}$ PFAAs associated with  
275 suspended particles in the Arctic (Greenland) region is expected to undergo an  
276 exponential increase, reaching approximately 50-fold above 2020 levels by 2100 under  
277 the SSP 585 scenario (Figure 5C). In the SCAG region, particle-bound PFAA fluxes are  
278 also projected to rise substantially, with a nearly tenfold increase over the same period  
279 (Figure 5D).

280

## 281 **DISCUSSION**

282 Although glaciers are increasingly recognized as secondary sources of pollutants, the  
283 magnitude and mechanisms of PFAA release, particularly via the particle-bound phase,  
284 have remained largely unresolved on a global scale. Our study addresses this gap by  
285 providing the first quantitative estimate of both dissolved and particle-bound PFAA  
286 releases from glaciers worldwide, revealing that glacial melt can export thousands of  
287 kilograms of legacy PFAAs to downstream ecosystems each year, with significant  
288 regional and chemical partitioning patterns.

289 In the following context, we first evaluate limitations of uncertainty in our global PFAA  
290 flux estimates. We then assess whether glaciers currently act as net sources or sinks of  
291 PFAAs by comparing the global patterns of release and depositional fluxes derived  
292 from the GEOS-Chem model. These glacial fluxes were further compared to global  
293 PFAA emission inventories to clarify their relative contribution and to identify locations  
294 of high concern. Beyond quantification, we explore the potential adverse effects of  
295 increased PFAA outflows, particularly the role of particle-bound transport. Finally, we  
296 discuss strategies for controlling future PFAA releases from glaciers under climate  
297 warming.

### 298 **Limitations**

299 Several sources of uncertainty may influence the accuracy of our global PFAA flux  
300 estimates. First, the limited number of sampled glaciers within each region may not  
301 fully capture spatial heterogeneity, potentially introducing bias when extrapolating to  
302 the regional scale. Temporal uncertainty also arises due to the seasonal variability of  
303 both meltwater runoff<sup>28</sup> and PFAA concentrations,<sup>19</sup> as most field campaigns were  
304 conducted during limited time windows. In some Arctic regions, the scarcity of glacial  
305 meltwater data led to the use of PFAA concentrations in adjacent seawater (Table S2),  
306 which may underestimate fluxes due to dilution effects. These factors likely contributed  
307 to the large standard deviations observed in both PFAA concentrations (Figure 2) and  
308 flux estimations (Table 1).

309 Second, data gaps in key regions remain. In particular, no observational data is currently  
310 available for Patagonia and the southern Andes. Glaciers in these regions are among the  
311 most rapidly retreating on Earth.<sup>29</sup> This lack of data would lead to an underestimation  
312 of global glacier-sourced PFAA fluxes. Moreover, geopolitical dynamics, such as  
313 international regulations on PFAA production, industrial substitution of legacy PFAAs,  
314 and the geographic relocation of manufacturing facilities, could alter emission patterns  
315 over time. However, due to the lack of reliable global models and transparent reporting,

316 the influence of these political and economic factors could not be quantitatively  
317 assessed.

318 Finally, analytical uncertainties also exist. Perfluorobutanoic acid (PFBA), a prevalent  
319 short-chain PFAA, has been inconsistently reported in previous studies due to analytical  
320 challenges.<sup>30,31</sup> To assess the impact of its exclusion, we recalculated fluxes for  $\Sigma_{12}$   
321 PFAAs (excluding PFBA), with total fluxes of  $2250 \pm 306$  kg year<sup>-1</sup>, with 18%  
322 transported via the suspended particles (Table S6). The regional flux ranking remained  
323 consistent—SCAG ( $536 \pm 39$  kg year<sup>-1</sup>), AEG ( $449 \pm 24$  kg year<sup>-1</sup>) and WCUG ( $426 \pm 15$   
324 kg year<sup>-1</sup>)—but the total global flux was substantially lower, underscoring the  
325 importance of including PFBA in future assessments. Improved analytical techniques,  
326 such as high-resolution mass spectrometry,<sup>32</sup> are essential for accurately quantifying  
327 low-mass PFAAs and ensuring comparability across studies in the future.

### 328 ***Global patterns of release and depositional fluxes***

329 Based on the field measurements, predicted  $\log K_d$  and modeled glacial runoffs, we  
330 estimated that glaciers globally release approximately  $3,547 \pm 375$  kg year<sup>-1</sup> of  
331  $\Sigma_{13}$ PFAAs into downstream environments, with about 12% transported via suspended  
332 particles. While previous studies have only quantified dissolved-phase fluxes at the  
333 regional scale,<sup>25,26</sup> our study extends this assessment to the global scale and incorporates  
334 the often-overlooked particle-bound phase.

335 To evaluate whether PFAA release is in balance with atmospheric inputs, we compared  
336 regional glacial output fluxes with deposition fluxes estimated by the GEOS-Chem  
337 global atmospheric chemistry model (Table 1),<sup>33</sup> and the estimates derived from ice  
338 core records in the same glacial regions (Table S9).<sup>14,15,34-36</sup> Deposition fluxes showed  
339 substantial regional variability, with  $\text{AIS} \approx \text{GIS} \approx \text{SCAG} > \text{ACG} \approx \text{WCUG} > \text{CEG} \approx$   
340  $\text{NAG}$  (Table 1). The high depositional fluxes in AIS and GIS were primarily due to their  
341 large glacial surface areas. In contrast, the comparable flux in SCAG can be linked to  
342 the high deposition concentration of PFAAs from nearby emission sources.<sup>37,38</sup>

343 Since GEOS-Chem model only include PFCAs, we focused on the comparison on  
344  $\Sigma_{10}$ PFCAs (Table 1). Our results shows that the global average glacial release fluxes of  
345  $\Sigma_{10}$ PFCAs ( $\sim 2900 \text{ kg year}^{-1}$ ) were about 3 to 9 times higher than the modeled  
346 atmospheric deposition inputs ( $329\text{--}900 \text{ kg year}^{-1}$ ). Regionally, the glacial release flux  
347 exceeds the maximum depositional flux in all glacial regions, except for the AIS. This  
348 was obvious in ACG and SCAG, where the export fluxes of  $\Sigma_{10}$ PFCAs were 6 to 8 times  
349 higher than the maximum import fluxes (Table 1), followed by CEG and WCUG, due  
350 to rapid glacier ablation.<sup>39</sup> These findings indicated that under climate warming most  
351 glaciers were experiencing a net loss of PFAAs.

352 By contrast, the maximum depositional fluxes for the AIS were about twofold higher  
353 than the current release fluxes of  $\Sigma_{10}$ PFCAs, suggesting that the AIS may still act as a  
354 temporary sink for PFAAs.<sup>36</sup> Therefore, accumulation of PFAAs in the AIS should be  
355 continuously monitored,<sup>36,40</sup> particularly concerning their adverse effects on sensitive  
356 Antarctic wildlife like penguins.<sup>41</sup>

357 Intriguingly, based on results from the Devon, Meighen, Melville and Agassiz ice caps  
358 in 2005,<sup>42</sup> the total depositional flux of just four PFCAs, including PFOA,  
359 perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic  
360 acid (PFUdA), to the Arctic (north of  $65^\circ \text{ N}$ ) was approximately  $2603 \text{ kg year}^{-1}$ . This  
361 value is almost an order of magnitude higher than the estimated depositional fluxes for  
362 the Arctic regions (ACG+GIS+AEG =  $128\text{--}390 \text{ kg year}^{-1}$ ) derived from the GEOS-  
363 Chem model for the period 2013–2015 (Table 1). The substantial decline in deposition  
364 flux during 2013–2015 may be attributed to the gradual phase-outs of PFAAs.<sup>43</sup> This  
365 discrepancy suggests that large amounts of PFAAs had already been deposited and  
366 stored in Arctic glaciers during earlier decades.

367 Known as “forever chemicals”, PFAAs deposited on snow/ice are likely to undergo  
368 minimal degradation,<sup>11</sup> resulting in their long-term storage within the glaciers for  
369 decades to centuries.<sup>44</sup> With continuous global warming, the previously stored PFAAs

370 in glaciers (aged ice) would be largely released at an increasing rate.<sup>45,46</sup>

### 371 ***Glacial fluxes compared to global PFAA emissions***

372 While glaciers have been identified as secondary sources of PFAAs, their contribution  
373 remains limited in the context of global emissions. PFAAs have been widely used in  
374 industrial and household applications since the mid-20<sup>th</sup> century.<sup>6</sup> Under a high-  
375 emission scenario, global annual emissions of  $\Sigma_{10}$ PFCAs around 2020 were estimated  
376 at approximately 400 tonnes.<sup>47</sup> In comparison, the total glacial release fluxes of  
377  $\Sigma_{10}$ PFCAs are approximately 2.9 tonnes per year (Table 1), representing only about 0.7%  
378 of annual global emissions and indicating a limited contribution. Similarly, the input of  
379 PFAAs to the Arctic via glacial meltwater is also minor compared to the hundreds of  
380 tonnes transported by sea spray aerosols.<sup>31</sup>

381 Nevertheless, the local impact of glacial release on PFAA distribution and wildlife in  
382 glacial regions remains significant. As observed in Figure 4A, PFAA concentrations in  
383 Arctic fjords are higher than in other locations. The deep and narrow structure of fjords  
384 can restrict water circulation, leading to slower mixing and flushing, which causes  
385 PFAAs to accumulate in the fjord waters for extended periods. Similarly, PFAA levels  
386 in high-latitude Arctic fjord sediments are higher than in lower latitudes.<sup>20</sup> The unique  
387 geomorphology of Arctic fjords, characterized by deep valleys and significant sediment  
388 deposits, allows for more effective trapping and accumulation of PFAAs. Importantly,  
389 many of these fjords also sustain vital fisheries.<sup>48</sup> Therefore, the Arctic fjords should be  
390 of high concern in the Arctic regions.

391 In the SCAG region, high level PFAA areas are predominantly located south of 33°N  
392 latitude (Figure 4B). The Khumbu and Rongbuk Glaciers have the largest PFAA risk  
393 buffer zones, followed by the Yulong Baishui No.1 glacier (Figure 4B). These glaciers  
394 are located in the monsoon-affected southern Tibetan Plateau (south of 33°N), where  
395 glacier melt rates are higher compared to the north.<sup>49</sup> Proglacial river sediments have  
396 been proven to be substantial sinks for PFAAs released by glacial meltwater,<sup>18</sup> acting

397 as important sources for the benthic food web. The export of PFAA by meltwater and  
398 suspended particles has led to PFAA being enriched in cold-water fish species in the  
399 Tibetan rivers (south of SCAG).<sup>50</sup> Consequently, particular attention should be given to  
400 headwater regions at glacier termini and adjacent catchment, as they are likely hotspots  
401 for PFAA contamination.

#### 402 ***Adverse effects of increased PFAA outflows***

403 With ongoing glacial retreat, the particle-bound fraction of PFAAs, especially  
404 bioaccumulative long-chain compounds, is expected to increase substantially. Our  
405 estimates suggest that fluvial suspended particles currently deliver  $238 \pm 63 \text{ kg year}^{-1}$   
406 of long-chain PFAAs globally (Table S6), a figure likely to grow as glacier erosion and  
407 sediment fluxes intensify under warming conditions<sup>27,51,52</sup>. For example, projections for  
408 Greenland indicate that sediment transport could double every 30 years,<sup>27</sup> leading to a  
409 20- to 60-fold increase in particle-bound  $\Sigma_{13}$ PFAAs releases by 2100 under different  
410 SSPs scenarios (Figure 5C). This sediment enrichment may eventually surpass  
411 meltwater as the dominant pathway of PFAA outflows, posing increasing risks to  
412 proglacial ecosystems.

413 Sediments serve as an important pathway for the entry of PFAAs into the food web (i.e.,  
414 sediment – phytoplankton – zooplankton – Arctic char – seals – polar bears).<sup>22-24</sup>  
415 Bioaccumulation factors for long-chain PFAAs are approximately 30-50 fold from  
416 sediment to Arctic char,<sup>23</sup> 10-30 fold from char to ringed seals,<sup>53</sup> and up to 10 fold from  
417 seal to polar bear.<sup>54</sup> Under the worst-case warming scenario (SSP585), PFAA  
418 concentrations in polar bear liver tissues could rise from current levels of several  $\mu\text{g g}^{-1}$   
419 to potentially  $\text{mg g}^{-1}$  concentrations (Figure S4). Although no official safe thresholds  
420 have been established for PFAAs in polar bears, such extreme high concentrations  
421 would exacerbate their health risks.

422 Polar bears are already listed on the International Union for Conservation of Nature  
423 (IUCN) Red List.<sup>55</sup> Rising PFAA concentrations under future warming scenarios could

424 accelerate the decline of this keystone species, disrupt Arctic ecosystems, and  
425 contribute to broader biodiversity loss. Although the Arctic Monitoring and Assessment  
426 Programme (AMAP) has already warned of increasing PFAA-related toxicity in polar  
427 organisms,<sup>56</sup> our work provides a quantitative amplification of this trend, specifically  
428 in the increased concentrations found in the livers of polar bears (Figure S4). This is  
429 critical for understanding and mitigating the environmental impact of PFAAs released  
430 from glacial sources.

431 Compared to the Arctic, the Tibetan Plateau (southern SCAG) presents a lower risk  
432 profile. The aquatic food chain in the plateau is generally shorter than that of the Arctic  
433 marine ecosystem, which limits the biomagnification of PFAAs.<sup>50</sup> However, glaciers  
434 on the Tibetan Plateau serve as the headwaters of major Asian rivers that support the  
435 livelihoods of over two billion people downstream.<sup>49</sup> Under future warming scenarios,  
436 accelerated glacial melt and enhanced sediment transport may increase the release of  
437 particle-bound PFAAs into these headwater systems. This could pose adverse impacts  
438 on ice-dwelling headwater species at the base of the food web, such as algae, copepods,  
439 and amphipods.<sup>57</sup> Such upstream ecological disturbances may cascade downstream,  
440 threatening water quality and aquatic biodiversity in major Asian rivers.

#### 441 ***Controlling future PFAA releases from glaciers***

442 To mitigate adverse impacts of future PFAA releases from glaciers, coordinated and  
443 immediate global actions are essential. Reducing greenhouse gas emissions remains the  
444 most critical step, as slowing the rate of climate warming directly reduces glacial melt  
445 rates and the remobilization of legacy PFAAs stored in ice. Lower emissions will not  
446 only stabilize climatic systems but also protect ecosystems from accelerated  
447 degradation due to rising temperatures.

448 In parallel, stronger global regulations on PFAS production and use are urgently needed.  
449 Current policies have largely targeted long-chain PFAAs and their precursors, but short-  
450 chain and emerging PFAS—many of which are poorly regulated or still unknown—are

451 now dominant in the environment. Expanding regulatory frameworks to include these  
452 compounds is essential for protecting sensitive ecosystems, particularly polar and  
453 alpine regions where bioaccumulation risks remain high.

454 Policy oversight must also account for unintended consequences. For instance, the  
455 global transition from chlorofluorocarbons (CFCs) to short-lived substitutes under the  
456 Montreal Protocol has inadvertently accelerated atmospheric deposition of ultra-short-  
457 chain PFAAs, such as trifluoroacetic acid (TFA).<sup>44,58</sup> These substitutes, while beneficial  
458 for ozone protection, now contribute both to climate forcing and chemical  
459 contamination.<sup>59</sup> This underscores the need for integrated environmental governance,  
460 where solutions to one crisis do not amplify another.

461 As shown in [Figures 5A and 5B](#), the world faces a narrow window of approximately 15  
462 years to effectively curb glacier-derived PFAA release. A combined strategy of  
463 emissions reduction, PFAS regulation, and careful chemical substitution is essential.  
464 Without coordinated intervention, PFAA discharges from glaciers are projected to rise  
465 sharply, with long-term consequences for freshwater security, fisheries, biodiversity,  
466 and global pollutant burdens.

467 Finally, establishing long-term environmental and wildlife monitoring programs is  
468 crucial. These systems serve as early-warning networks, enabling the detection of rising  
469 PFAA levels and other ecological stress signals in fragile regions. Proactive monitoring  
470 not only informs risk assessment and response but also supports evidence-based policy  
471 decisions aimed at preventing irreversible ecological damage to the most fragile  
472 cryosphere on our planet.

473

## 474 **METHODS**

### 475 ***Study area and field sampling***

476 Seven glaciers on the Tibetan Plateau (affiliated to part of SCAG) were investigated in

477 the field, including the Rongbuk (RB), Qiangyong (QY) and Rije Cojia (RC) glaciers  
478 in the Himalayan Mountains, the Korchung Gangri (KG) and Zhadang (ZD) glaciers in  
479 the Gangdise–Nyainqêntanglha Mountains, and the Galongla (GL) and Parlung No. 4  
480 (PL) glaciers in the Hengduan Mountains (Figure 1). Among them, RB, ZD, KG, QY,  
481 and RC are continental valley glaciers, whereas the GL and PL are typical temperate  
482 glaciers. Details of these glacial regions, among which there are significant  
483 geographical, hydrological, and meteorological differences, are summarized in Table  
484 S10.

485 Meltwater sampling was conducted from July to August 2020, corresponding to the  
486 peak melting season. At each glacier, surface runoff samples (0–30 cm) were collected  
487 at three sites from the upstream to the downstream (Figure 1A). Approximately 20 L of  
488 water was collected at each site using pre-cleaned polypropylene bottles. Prior to use,  
489 all sampling bottles were rinsed three times with methanol and Milli-Q water. To  
490 minimize contamination, sampling materials containing fluoropolymer coatings or  
491 PFAS-related components were strictly avoided. *In situ* measurements of basic water  
492 quality parameters, including water temperature ( $T_w$ ), pH, conductivity ( $\sigma$ ), and  
493 dissolved oxygen (DO), were conducted during the sampling at the same site.

#### 494 **Analysis of PFAAs**

495 Glacial runoff samples from seven glacial catchments on the Tibetan Plateau were  
496 analyzed for 13 target PFAAs. Upon analysis, all water samples were filtered through  
497 Whatman 0.7  $\mu\text{m}$  glass fiber filters (GFFs) to separate suspended particles. The filtrates  
498 (2L each) were subjected to solid-phase extraction (SPE) using Waters Oasis WAX  
499 cartridges (150 mg, 6  $\text{cm}^3$ , 30  $\mu\text{m}$ ), while the retained GFFs were extracted by ultrasonic  
500 agitation using methanol to measure particle-bound PFAAs. Detailed extraction  
501 procedures are described in Notes S2 and S3.

502 Quantification of 13 PFAAs and internal standards (Table S11) were performed using  
503 high-performance liquid chromatography coupled with tandem mass spectrometry

504 (HPLC–MS/MS; ThermoFisher UltiMate 3000 and TSQ Quantiva triple quadrupole  
505 system), as described in [Note S4](#). POC on the GFFs (the suspended particle samples)  
506 was measured using a Shimadzu 5000-A total organic carbon analyzer to support  
507 subsequent partitioning analysis.

508 All analytical procedures were conducted under strict quality assurance and quality  
509 control measures ([Note S5](#)). Field and procedural blanks prepared with Milli-Q water  
510 yielded concentrations below the method quantification limits ([Table S12](#)), confirming  
511 that sample contamination was negligible. Matrix spiked recoveries, obtained by  
512 spiking with 2000 pg of internal standards, ranged from 52% to 81% for water samples  
513 and 42% to 70% for suspended particle samples, with detailed values listed in [Table](#)  
514 [S13](#).

### 515 ***Identification of glacial regions and data compilation***

516 The identification of glacial regions was undertaken using version 6.0 of the RGI  
517 (<http://www.glims.org/RGI/>). Eight major glacial regions were included in this study  
518 ([Figure 2](#)): ① West Canada and USA (WCUG), ② Arctic Canada (ACG), ③ Greenland  
519 Ice Sheet (GIS), ④ Arctic Europe (AEG), ⑤ North Asia (NAG), ⑥ Central Europe  
520 (CEG), ⑦ South and Central Asia (SCAG), and ⑧ Antarctic and Subantarctic Ice Sheet  
521 (AIS).

522 To construct a global dataset of PFAA concentrations, we compiled published data for  
523 13 PFAAs from 680 samples across 49 glaciers in these eight glacial regions ([Figure 2](#)).  
524 The dataset includes samples from glacial runoff, snow/ice, and seawater neighboring  
525 the glaciers. In addition, we combined our field measurements ( $n=59$ ) from seven  
526 glaciers on the Tibetan Plateau with the above published data to produce a global dataset  
527 ( $n=739$ , [Table S2](#)). This integrated dataset offers the basis for the global-scale  
528 assessment of PFAA fluxes from glaciers.

## 529 *Drivers of particle-water partition coefficient ( $K_d$ )*

530 The  $K_d$  is defined as the ratio of a chemical's concentration in the particle-bound phase  
531 ( $C_p$ ) to that in the dissolved phase ( $C_w$ ) (i.e.,  $K_d = C_p/C_w$ ). The  $\log K_d$  data obtained from  
532 measurements of 13 PFAA concentrations in runoff water and suspended particles from  
533 the Tibetan glaciers is presented in [Table S4](#). While  $K_d$  values are variable, they often  
534 respond to environmental factors and chemical properties of PFAAs.

535 To explore the key drivers of PFAA partitioning, we analyzed the relationships between  
536  $\log K_d$  and several environmental parameters, including pH,  $T_w$ , POC,  $\sigma$  and DO, as well  
537 as molecular descriptors such as perfluorinated chain length and functional group type  
538 (i.e., carboxylates for PFCAs and sulfonates for PFSAs). Statistically significant  
539 correlations were observed between  $\log K_d$  and both POC and  $T_w$  ( $p < 0.05$ ), while no  
540 significant correlations were found for pH, DO or  $\sigma$  ([Table S14](#)). These findings are  
541 consistent with previous studies highlighting the importance of organic carbon and  
542 water temperature in governing PFAS partitioning.<sup>60</sup>

543 In addition, we examined the influence of molecular hydrophobicity using theoretical  
544 ionic octanol–water partition coefficients ( $\log K_{ow-ionic}$ ), as reported by Hidalgo and  
545 Mora-Diez (2015).<sup>61</sup> Given that glacial meltwaters are commonly alkaline<sup>62</sup> and PFAAs  
546 possess low  $pK_a$  values (typically  $< 1.0$ ), these compounds are expected to be fully  
547 dissociated to their anionic forms.<sup>63</sup> The  $\log K_{ow-ionic}$  values, which vary by carbon chain  
548 length but are fixed for each compound, were significantly correlated with  $\log K_d$  ( $p$   
549  $< 0.01$ , [Figure S5](#)), which suggests that  $\log K_{ow-ionic}$  can be a useful model predictor of  
550  $\log K_d$ , especially if combined with the abovementioned POC and  $T_w$ .

551 To support model development, we compiled a comprehensive dataset ( $n=771$ ),  
552 containing  $\log K_d$ ,  $T_w$ , POC,  $\log K_{ow-ionic}$ , and PFAA type. These data were sourced from  
553 both glacial regions ( $n=191$ ) and non-glacial environments (e.g., inland watersheds and  
554 coastal areas;  $n=580$ ). All values were either directly measured or extracted from the  
555 previous literature, as listed in [Table S4](#). Notably,  $T_w$  values were all below 19°C to

556 represent relatively low-temperature environments, with data screening criteria detailed  
557 in [Note S1](#).

### 558 ***Prediction of $K_d$ with machine learning models***

559 Traditional curve-fitting methods showed limited performance in predicting  $\log K_d$ . To  
560 overcome this limitation, we developed machine learning (ML) models to estimate  
561  $\log K_d$ , including the support vector machine (SVM), decision tree (DT), random forest  
562 (RF), and extreme gradient boosting algorithm (XGBoost).

563 All ML models were implemented in Python 3.9.7 using the *scikit-learn* module. Model  
564 performance was evaluated using multiple metrics: the coefficient of determination for  
565 the training set ( $R^2$ ), the predictive  $Q^2$  for the test set, and the mean absolute error (MAE)  
566 and mean squared error (MSE) for both sets. To ensure robustness and reproducibility,  
567 the dataset was randomly divided into training and test sets using  
568 `sklearn.model_selection.train_test_split`, with varying split ratios ranging from 5:95 to  
569 50:50, and 10 replicates for each ratio.

570 Among all models, the XGBoost-ML (denoted as Xgb\_A) exhibited the best overall  
571 performance, achieving the highest  $R^2$  ( $0.95 \pm 0.01$ ) and  $Q^2$  ( $0.85 \pm 0.04$ ), along with the  
572 lowest MAE and MSE for both the training set ( $0.138 \pm 0.016$  and  $0.034 \pm 0.008$ ,  
573 respectively) and test set ( $0.241 \pm 0.032$  and  $0.105 \pm 0.030$ , respectively). Additionally,  
574 statistical analyses using Student's t-tests showed no significant variation in predictive  
575 accuracy when increasing the proportion of the test set from 5% to 45% of the total  
576 dataset (raw significant level  $\alpha=0.05$ , Bonferroni-corrected significant level  $\alpha=0.0011$ ,  
577 number of tests =45).

578 Based on these results, the XGBoost-ML (Xgb\_A) was selected as the best-performing  
579 model for predicting  $\log K_d$ . Further details regarding ML model development and  
580 comparative performance are provided in [Note S1](#).

581 **Glacial mass balance model**

582 Glacial runoff was calculated using a water balance approach adapted from Bliss et al.<sup>39</sup>  
583 Within each glacial region, there are several subregions characterized by different  
584 meteorological and geological characteristics. In addition, there are several different  
585 landforms in each subregion, which might influence the melting rate of snow or ice, i.e.  
586 the degree-day factor for snow/ice. Therefore, the estimation for the monthly glacier  
587 runoff needs to account for variations among different subregions and landforms. The  
588 monthly average glacier runoff of each glacial region ( $Q$ ) was calculated using Equation  
589 1 below.

590 
$$Q = \sum_{i=1}^n S_i \times (a_i + P_{liq,i} - R_i) \quad (\text{Equation 1})$$

591 where  $S_i$  (km<sup>2</sup>) is the effective area of the glacial region, which is computed by the  
592 annual average area of subregion  $i$ , included by the glacial region.  $a_i$  is the snow/ice  
593 melt of each month,  $P_{liq,i}$  (mm) is the net accumulated liquid precipitation of each month  
594 and  $R_i$  (%) is the potential of refreezing during melting of each month.

595 It was assumed that melting had a linear correlation with the monthly air temperature,  
596  $a_i$ , if  $> 0^\circ\text{C}$ , and thus  $a_i$  was calculated using Equation 2 below.

597 
$$a_i = f_{snow/ice} \times \int \max(T_i, 0) dt \quad (\text{Equation 2})$$

598 where  $f_{snow/ice}$ , the degree-day factor for snow/ice (mm w.e. d<sup>-1</sup> °C<sup>-1</sup>), is a constant that  
599 depends on the sample types, and  $T_i$ , the monthly air temperature (°C) above the glacier  
600 surface, is a random variable, which was computed using Equation 3 below:

601 
$$T_i = T_{\text{annual average}} + (T_{\text{summer}} - T_{\text{annual average}}) \times \sin((i - 4) \times \frac{2\pi}{12}) + \delta T_i \quad (\text{Equation 3})$$

602 where the  $T_{\text{annual average}}$  and  $T_{\text{summer}}$  were provided from previous studies.<sup>39</sup> The  $\delta T_i$  is a  
603 random variable following normal distributions, with a mean value of 0°C. When  $i$   
604 referred to winter (December to February) or summer (June to August) time, the  $\delta T_i$   
605 was the standard deviation of summer temperatures. Vice versa, when  $i$  referred to

606 Spring (March to May) or Autumn (September to November) time, the  $\delta T_i$  was the  
607 standard deviation of annual average temperatures.

608 Fluxes contributed by liquid precipitations,  $P_{liq,i}$ , were calculated using the following  
609 Equation 4.

$$610 \quad P_{liq,i} = P_i \times \theta_{(T_i,0)} \quad (\text{Equation 4})$$

611 where  $P_i$  is a random parameter following a normal distribution, with its mean value  
612 and standard deviation cited from previous publications,<sup>62</sup> and  $\theta_{(T_i,0)}$  is a Heaviside  
613 function, which equals to 1 when  $T_i$  is greater than 0, and equals to 0 when  $T_i$  is  
614 smaller or equals to 0.

615  $R_i$  was estimated according to the linear relationship between  $R_i$  and air temperature ( $T_i$ )  
616 at different elevations, assuming that the snow meltwater frozen on the surface of the  
617 glacier ( $R_i$ ) does not flow away. The estimation was conducted using Equation 5 below.

$$618 \quad R_i = -0.69 \times T_i + 0.0096 \quad (\text{Equation 5})$$

619 For each of eight glacial regions, 2500 Monte Carlo simulations were conducted (Note  
620 S6). For each simulation, glacier runoff fluxes in 12 months were computed using the  
621 method stated above and then summed over to obtain the annual glacier runoff fluxes.  
622 One glacial region may be composed of several glaciers, while each of them might have  
623 a different composition of landforms: 3 types of landforms, namely the sample types,  
624 were considered, having different degree-day factors:  $f_{\text{snow/ice}} = 1$  for *glacier runoff*,  
625  $f_{\text{snow/ice}} = 2$  for *snow/ice*, and  $f_{\text{snow/ice}} = 4$  for *seawater neighboring the glaciers*. Each  
626 sample type was chosen randomly for the simulation of the whole year, without bias.

627 **Released flux estimation of PFAAs (dissolved part).** The annual fluxes of 13 PFAAs  
628 in the dissolved phase were estimated for eight glacial regions using Monte Carlo  
629 simulations (Note S6). For each region and each PFAA, 2500 simulations were

630 conducted. Because a glacial region can consist of multiple subregions of varying sizes,  
631 the number of simulations allocated to each subregion is determined based on the  
632 respective area sizes within the larger region. For each simulation, the PFAA  
633 concentrations discharged in a subregion ( $C_w$ ) were those of a sample randomly chosen  
634 from one of three sample types (ice/snow, glacier runoff, and seawater) (Table S2), with  
635 equal probability.

636 The glacial meltwater fluxes of the subregion from each sample type in each month  
637 were determined in the above-mentioned glacial mass balance model, and the PFAA  
638 fluxes in dissolved phase of that month were the concentration of PFAAs times the  
639 corresponding runoff volume. Annual fluxes for each region were derived by summing  
640 monthly fluxes across all subregions.

641 ***Released flux estimation of PFAAs (suspended particle part)***. The annual fluxes of 13  
642 PFAAs partitioned on suspended particles of the eight glacial regions were also  
643 estimated using Monte Carlo simulations (Note S6). Based on the predictions for  $\log K_d$   
644 by the XGBoost-ML and the reported mean PFAA concentrations in water of each  
645 glacial region, we obtained the concentration of PFAAs on suspended particles ( $C_p$ )  
646 (Table S5). The annual particulate PFAA fluxes from different glacial regions were then  
647 estimated by multiplying the mean  $C_p$  by the regional average glacial runoff and the  
648 mean suspended particle concentration (Table 1). The final flux estimates for all regions  
649 are presented in Table S6.

### 650 ***Future Release Potential of PFAAs***

651 The project future release fluxes of 13 PFAAs in both dissolved and particle-bound  
652 phases were estimated with the support of shared socioeconomic pathways (SSPs),  
653 which provided estimates of precipitation and air temperature from 2020 to 2100  
654 (Figures S6 and S7). The modeling assumed there would be sufficiently large amounts  
655 of historical storage of PFAAs in glaciers.

656 Three SSP scenarios, namely SSP126, SSP 245, and SSP585,<sup>64,65</sup> were utilized to  
657 represent low, intermediate, and high global warming scenarios, respectively. For each  
658 scenario, the annual and summer average air temperatures in each subregion were  
659 calibrated using the 2020 values as a baseline. This ensured that the projected  
660 temperature increases under different SSPs, relative to 2020, were maintained.  
661 Similarly, monthly precipitation values for each region were calculated based on the  
662 recorded data from 2020, such that changes in annual precipitation projected by the  
663 SSPs were consistently applied while preserving the seasonal distribution. Detailed  
664 calibration procedures are provided in [Note S6](#).

665 The potential release fluxes of 13 PFAAs, both in the dissolved phase and on suspended  
666 particles, were estimated annually from 2020 to 2100 under the three climate warming  
667 scenarios ([Figures 5A and 5B](#)). For each year, 2,500 Monte Carlo simulations were  
668 conducted to account for variability and uncertainty. The modeling approach for 2021–  
669 2100 followed the same framework used for the baseline year 2020, ensuring  
670 methodological consistency across the entire simulation period.

### 671 ***Deposition and budget estimates of $\Sigma_{10}$ PFCAs***

672 Atmospheric deposition is the primary input pathway of PFAAs to glacier surfaces. As  
673 the GEOS-Chem model does not simulate PFSAs,<sup>33</sup> our analysis focuses on ten PFCAs  
674 with carbon chain lengths from C4 to C13 ( $\Sigma_{10}$ PFCAs). Using the atmospheric  
675 depositional fluxes of  $\Sigma_{10}$ PFCAs generated by the GEOS-Chem model,<sup>33</sup> along with  
676 glacier surface area data ([Table 1](#)), we estimated the deposition fluxes (input) of  
677  $\Sigma_{10}$ PFCAs to global glaciers. By comparing this input with the predicted release fluxes  
678 (output) of  $\Sigma_{10}$ PFCAs, we evaluated the input-output budget of  $\Sigma_{10}$ PFCAs across the  
679 eight different glacial regions ([Table 1](#)).

680 The GEOS-Chem model (version 12.2) is a widely used, community-developed  
681 atmospheric chemistry model that incorporates reanalysis meteorological data to  
682 simulate the transport and deposition of pollutants.  $\Sigma_{10}$ PFCAs were included in the

683 GEOS-Chem model with a detailed set of chemical reactions, emission inventories,  
684 prescribed meteorology, and wet and dry deposition schemes.<sup>33</sup> The model was then  
685 run at a horizontal resolution of  $4^{\circ}\times 5^{\circ}$ , with 46 vertical levels, using assimilated GEOS-  
686 FP meteorology data. This produced the modeled spatial distribution of the annual  
687 deposition of  $\Sigma_{10}$ PFCAs in 2013–2015 resulting from the inventory of degradation of  
688 precursor molecules and direct global emissions. Due to the observed long-term  
689 stability of atmospheric PFAA concentrations, e.g. perfluorooctane sulfonic acid  
690 (PFOS), in polar regions over recent decades (Note S7)<sup>12,66</sup>, we assumed the annual  
691 deposition fluxes of  $\Sigma_{10}$ PFCAs from 2013 to 2020 are the same.

692 The GEOS-Chem estimation contains a few uncertainties. These include incomplete  
693 emission inventories of short-chain PFAA replacements<sup>67,68</sup> and precursors,<sup>17,47</sup>  
694 unquantified contribution of sources such as sea spray aerosols,<sup>69,70</sup> and limited  
695 understanding of atmospheric transformation mechanism.<sup>71</sup> Despite these limitations,  
696 GEOS-Chem model offers a valuable basis for estimating large-scale deposition  
697 patterns of PFAAs to compare their input-output budget in remote glacial environments.

698

## 699 **RESOURCE AVAILABILITY**

### 700 ***Lead contact***

701 Xiaoping Wang ([wangxp@itpcas.ac.cn](mailto:wangxp@itpcas.ac.cn)).

### 702 ***Materials availability***

703 This study did not generate new unique materials.

### 704 ***Data and code availability***

705 The code and data needed for Monte Carlo simulation of PFAA glacial runoff volumes,  
706  $\log K_d$  estimation for each situation based on trained XGBoost model, and the estimated  
707 release fluxes of PFAAs via dissolve phase and suspended particles are available in

708 [https://github.com/EssexHUBOYUAN/Glacial\\_runoff\\_PFAS\\_sims](https://github.com/EssexHUBOYUAN/Glacial_runoff_PFAS_sims).

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## 715 **AUTHOR CONTRIBUTIONS**

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717 L.W., M.C., and S.R.; formal analysis, B.H. and M.C.; visualization, Y.Z., B.H., and  
718 M.C.; writing—original draft: Y.Z. and B.H.; writing—review and editing, X.W., J.F.,  
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## 721 **DECLARATION OF INTERESTS**

722 The authors declare no competing interest.

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## 1018 **Figures and tables**

1019 **Figure 1. Sampling sites from upstream to downstream (A) and average concentrations (B)**  
1020 **and compositions (C) of 13 perfluoroalkyl acids (PFAAs) in seven glacial regions of the Tibetan**  
1021 **Plateau.** The seven glacial regions are: RB, the Rongbuk glacier; KG, the Korchung Gangri glacier;  
1022 ZD, the Zhadang glacier; GL, the Galongla glacier; PL, the Parlung No. 4 glacier; QY, the  
1023 Qiangyong glacier; and RC, the Rije Cojia glacier. U, M, and D in part (A) represent upstream,  
1024 midstream, and downstream sites, respectively.

1025 **Figure 2. Average concentrations and compositions of 13 perfluoroalkyl acids (PFAAs) in (A)**  
1026 **eight glacial regions and sampling sites in the (B) Arctic and (C) Asian regions.** The dots  
1027 represent sample locations in different glacial regions. Glaciers are shown in blue, ice sheets in  
1028 white, and the black lines indicate the outlines of glaciers from the Randolph Glacier Inventory. The  
1029 eight glacial regions are: ①West Canada and USA (WCUG), ②Arctic Canada (ACG), ③Greenland  
1030 ice sheet (GIS), ④Arctic Europe (AEG), ⑤North Asia (NAG), ⑥Central Europe (CEG), ⑦South  
1031 and Central Asia (SCAG), ⑧Antarctic and Subantarctic ice sheet (AIS). The pie charts represent  
1032 the composition of PFAAs in each glacial region, and bold numbers present the average  
1033 concentration and standard deviation of PFAAs. The 13 PFAAs are perfluorobutanoic acid (PFBA),  
1034 perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid  
1035 (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid  
1036 (PFDA), perfluoroundecanoic acid (PFUDA), perfluorododecanoic acid (PFDoA),  
1037 perfluorotridecanoic acid (PFTrDA), perfluorobutane sulfonic acid (PFBS), perfluorohexane  
1038 sulfonic acid (PFHxS), and perfluorooctane sulfonic acid (PFOS).

1039 **Figure 3. Comparison of the measured  $\log K_d$  for perfluoroalkyl acids (PFAAs) in glacial and**  
1040 **non-glacial regions with the predicted  $\log K_d$  driven from the XGBoost algorithm: (A) Training**  
1041 **set and (B) Test set.** The red and green squares indicate the ratios of measured  $\log K_d$  and predicted  
1042 values that fall within  $\pm 1$  log unit for samples from glacial and non-glacial regions, respectively.  
1043 The white square represents the ratio of the measured  $\log K_d$  and predicted  $\log K_d$  that falls outside  
1044  $\pm 1$  log unit, with only one data observed in the non-glacial regions.

1045 **Figure 4. Geographical distribution of perfluoroalkyl acids (PFAAs) concerned areas in the**  
1046 **(A) Arctic and (B) the Tibetan Plateau.** The red circle and purple circle represent the concerned  
1047 sites for water and sediment, respectively. The orange triangles mark areas of potential biodiversity  
1048 loss due to high PFAA levels found in polar bears and Arctic foxes, as reported in previous studies.<sup>72</sup>  
1049 Detailed data for the buffer zone are provided in [Tables S7 and S8](#), with average concentrations for  
1050 water and sediment being approximately 2000 pg/L and 1000 pg/g, respectively. Buffer zones I, II,  
1051 and III represent areas where PFAA concentrations in water and sediment exceed the regional  
1052 average concentration by 1 time, 2 times, and more than 3 times, corresponding to impact zones of  
1053 100 km, 200 km, and 300 km, respectively.

1054 **Figure 5. Annual future release potential (kg year<sup>-1</sup>) of perfluoroalkyl acids (PFAAs) from glaciers**  
1055 **in eight glacial regions via the (A) dissolved phase and (B) suspended particles in 2020-2100 under**  
1056 **three climate change scenarios (SSP126, SSP 245 and SSP 585). And the release potential (kg year<sup>-1</sup>)**  
1057 **of PFAAs via suspended particles from glaciers in the (C) Greenland Ice Sheet (GIS) and (D) South**  
1058 **and Central Asia (SCAG).** The release potential refers to a predicted possibility of PFAA fluxes under  
1059 future climate scenarios. The solid line in parts (A) and (B) presents the average value with a window of  
1060 one year and the shaded area indicates the range of minimum and maximum values. The line in the box of  
1061 part (C) and (D) is median line, the edges of the boxes are the 5th and 95th percentile, and the whisker is  
1062 range within the minimum and maximum values. The difference in the time scales on the horizontal axis  
1063 in part (C, D) compared with (A, B) is due to the assumption that the release of glacier meltwater sediment  
1064 doubles every 30 years.<sup>27</sup>

1065 **Table 1. Release fluxes of perfluoroalkyl acids (PFAAs) via the dissolved phase and suspended particles from glaciers in eight glacial regions globally**  
 1066 **compared with the depositional fluxes, with the glacial area, glacial runoff and average concentration of suspended particles.**

Glacial region	Glacial area (km <sup>2</sup> )	Glacial runoff (km <sup>3</sup> year <sup>-1</sup> )	ACSP (mg L <sup>-1</sup> )	Release fluxes (kg year <sup>-1</sup> )		$\Sigma_{13}$ PFAAs	$\Sigma_{10}$ PFCAs	Depositional fluxes of $\Sigma_{10}$ PFCAs estimated by the GEOS-Chem model (kg year <sup>-1</sup> ) <sup>33</sup>	
				Dissolved phase <sup>a</sup>	Suspended particles <sup>a</sup>			Minimum	Maximum
WCUG	101,249	400	182	380 ± 8.6 (89%)	46 ± 6.0 (11%)	426 ± 15	388 ± 13	16.9	79.6
ACG	146,016	212	97	683 ± 47 (98%)	14 ± 1.3 (2%)	698 ± 48	683 ± 47	17.5	81.8
GIS	1,718,000	547	881	153 ± 8.4 (49%)	162 ± 181 (51%)	316 ± 190	179 ± 42	96.2	206
AEG	88,498	146	91	520 ± 18 (92%)	44 ± 7.3 (8%)	564 ± 25	330 ± 11	14.2	102
NAG	2410	7	91	9.1 ± 0.5 (85%)	1.7 ± 0.5 (15%)	11 ± 0.9	8.3 ± 0.6	0.39	2.89
CEG	2075	9	356	18 ± 0.3 (88%)	2.6 ± 0.1 (12%)	21 ± 0.4	20 ± 0.3	3.32	3.32
SCAG	97,604	352	500	1244 ± 73 (89%)	151 ± 13 (11%)	1394 ± 86	1159 ± 60	151	205
AIS	13,746,463	48	73	109 ± 9.0 (93%)	8.2 ± 1.1 (7%)	118 ± 10	116 ± 10	28.9	220
Total				3117 ± 165 (88%)	430 ± 210 (12%)	3547 ± 375	2884 ± 184	329	900

1067 Notes: WCUG, Western Canada and USA; ACG, Arctic Canada; GIS, Greenland ice sheet; AEG, Arctic Europe; NAG, North Asia; CEG, Central Europe; SCAG,  
 1068 South and Central Asia; AIS, Antarctic and Subantarctic ice sheet; ACSP, average concentration of suspended particles in each glacial region reported previously.  
 1069 The areas of the GIS and AIS were obtained from Pfeiffer et al.<sup>73</sup> and Liston et al.<sup>74</sup>, respectively. The other data are available from the Randolph Glacier Inventory  
 1070 version 6.0 (<http://www.glims.org/RGI/>). The glacial runoffs of the GIS and AIS are available from Tedesco et al.<sup>75</sup> and Liston et al.<sup>74</sup>, respectively, with the other data

1071 collected from Bliss et al.<sup>39</sup>. The depositional fluxes of  $\sum_{10}$ PFCAs estimated by the GEOS-Chem model represent the average value for 2013-2015, the historical  
1072 depositional fluxes might be much higher.

1073 <sup>a</sup> Data presented as release fluxes (% of total fluxes).