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1 **Effect of soil organic matter on the plant uptake of perfluorooctanoic acid**
2 **(PFOA) and perfluorooctanesulfonic acid (PFOS) in lettuce on granular**
3 **activated carbon applied soil**

4

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21

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27 Korea.

28 **Abstract**

29 The presence of perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) in
30 crops is an important consideration for food safety. The soil organic matter (SOM) content may
31 affect the adsorption potential of PFOA and PFOS in water and soil and their subsequent uptake
32 in crops. To better understand these dynamics, the adsorption and uptake of PFOA and PFOS
33 in lettuce was investigated using granular activated carbon (GAC)-treated soils with varying
34 SOM content. The adsorption potential of GAC was investigated, with maximum adsorption
35 capacities for PFOA and PFOS calculated to be 9.091 mg g⁻¹ and 27.778 mg g⁻¹, respectively.
36 These values decreased to 5.208 mg g⁻¹ and 17.241 mg g⁻¹, respectively, after the addition of
37 0.04 wt.% humic acid. The average plant uptake factor (PUF) in low and high perfluoroalkyl
38 and polyfluoroalkyl acid (PFAA)-contaminated soils with 4.0 wt.% SOM was restricted to
39 0.353 for PFOA and 0.108 for PFOS. The PUFs were approximately two times lower than
40 those for soil with 2.6 wt% SOM. Addition of 1 wt% GAC to the soil successfully reduced the
41 PUF by up to 99.4%, with values of 0.006 (PFOA) and 0.005 (PFOS) in 2.6 wt% SOM treated
42 soil and 0.079 (PFOA) and 0.023 (PFOS) in 4.0 wt% SOM treated soil. Although the PUF in
43 the GAC-treated soil was drastically decreased, the PUF of the soil with 4.0 wt% SOM was at
44 least four times higher than that with 2.6 wt% SOM. Therefore, SOM content is an important
45 consideration in the remediation of PFOA and PFOS contaminated farmland soil using
46 carbonaceous adsorbent.

47 **Key words:** Soil organic matter (SOM), carbonaceous adsorbent, perfluorooctanoic acid
48 (PFOA), perfluorooctanesulfonic acid (PFOS), lettuce, plant uptake

49

50 **Introduction**

51 Perfluoroalkyl and polyfluoroalkyl acids (PFAAs) are widely used in industrial and consumer
52 products, as they are anti-static, flame retardant, and waterproof (Jian et al., 2017; Mahinroosta
53 and Senevirathna, 2020). Long-chained PFAAs such as perfluorooctanoic acid (PFOA) and
54 perfluorooctanesulfonic acid (PFOS) have reportedly demonstrated high physicochemical
55 stability and bioaccumulation rates with potent toxicity. PFOA, PFOS, and their precursors
56 were listed as persistent organic pollutants (POPs) by the Stockholm Convention; hence, their
57 use is being phased out globally (Ghisi et al., 2019; Kim et al., 2015; Seong et al., 2019; Weber
58 et al., 2019; Stockholm Convention, 2019). However, being highly persistent, PFAAs are
59 frequently detected even in remote regions (Guo et al., 2016; Jian et al., 2017; Perez et al.,
60 2014; Sungur, 2018). Recent research has indicated that PFAAs are present in sub part-per-
61 billion (ppb) concentrations of 88.7% in farmland soil in Korea (Choi et al., 2017). Furthermore,
62 for humans, the major sources of exposure to PFAAs are the PFOA and PFOS residues present
63 in food, particularly protein-rich foods, such as meat, dairy, and fish (Brambilla et al., 2015;
64 Ghisi et al., 2019; Heo et al., 2014; Xiang et al., 2020). In addition, the uptake and accumulation
65 of PFAAs in crops could be an important pathway for their accumulation in animal feed and
66 food (Choi et al., 2018; Eun et al., 2020; Ghisi et al., 2019; Liu et al., 2017; Luo et al. 2019;
67 Sungur, 2018; Wen et al., 2018; Xiang et al., 2020). Bioaccumulation factors (BAFs) of PFAAs
68 in crops have been widely reported. Reported BAFs for PFOA in different crops are: 0.004-
69 0.006 in rice_{grain}, 0.435 in rice_{whole}, 0.02 in corn_{fruit}, 0.111-2.821 in spinach, 0.12 in wheat, 0.154
70 in parsley, 0.252-6.6 in lettuce, 0.26-1.349 in carrot, 0.256-0.962 in Welsh onion, 0.355 in
71 plum, and 0.592 in white cabbage. Similar BAFs have been reported for PFOS: 0.002-0.003 in
72 rice_{grain}, 0.057 in rice_{whole}, 0.017-0.181 in Welsh onion, 0.048-3.175 in spinach, 0.067 in parsley,
73 0.086 in white cabbage, 0.181-1.071 in carrot, 0.286-0.65 in lettuce, and 0.96 in corn

74 (Bizkarguenaga et al. 2016; Choi et al. 2018; Choi et al. 2020; Lee et al. 2020; Ghisi et al. 2019;
75 Liu 2017; Navarro et al. 2017; Xiang et al. 2018). Although BAFs in crops are generally lower
76 than those in animals, the dietary exposure of PFAAs from crops was high enough to almost
77 reach the revised reference doses (RfD) for PFOA and PFOS as stipulated by European Food
78 Safety Authority (EFSA) (Choi et al. 2020). Therefore, PFAA residues in crops should be
79 strictly controlled and monitored to reduce human health risks from translocation and
80 bioaccumulation of PFAAs in animal feed and food.

81 Remediation techniques including conventional thermal and physicochemical treatments, have
82 been successfully applied to remove PFOA and PFOS residues from farmland soils. However,
83 incineration, sonication with water-extraction, and chemical treatments (oxidation/reduction),
84 are difficult to apply cost-effectively on large agricultural scales (Mahinroosta and
85 Senevirathna, 2020; Santos et al., 2016). Recently, carbonaceous adsorbents such as activated
86 carbon (AC) and biochar have been considered as alternative physicochemical treatment agents.
87 They have helped to immobilise organic pollutants with AC shown to adsorb over 90% of the
88 leaching PFAAs in soil (Choi et al., 2018; Hale et al., 2017; Kupryianchyk et al., 2016; Silvani
89 et al., 2019; Zhang et al. 2019). Carbonaceous adsorbents can also bring added benefits such
90 as carbon sequestration and improvement of physicochemical soil properties (Jakob et al. 2012;
91 Mahinroosta and Senevirathna, 2020; Xia et al. 2020). While adsorption properties of
92 carbonaceous adsorbents for PFAAs in water and soil have been investigated, studies regarding
93 the relationship between adsorption of carbonaceous adsorbents and crop-uptake of PFAAs are
94 limited (Askeland et al., 2020; Hale et al., 2017; Kupryianchyk et al., 2016; Xiao et al., 2017;
95 Zhang et al., 2019).

96 The plant uptake of PFOA/PFOS depends on crop cultivars, soil properties, and environment
97 parameters such as humidity, light-conditions, and temperature (Blaine et al., 2013; Eun et al.,

98 2020; Ghisi et al., 2019; Navarro et al. 2017; Xiang et al., 2018; Xiang et al., 2020). Among
99 them, soil organic matter (SOM) and humic acid (HA) content both affect the adsorption on
100 soil, and the crop-uptake of PFAAs (Blaine et al., 2014; Ghisi et al., 2019; Wang et al., 2015).
101 Organic matter helps adsorb PFAAs in soil and reduce their uptake in crops; similarly,
102 carbonaceous adsorbents in soils with high natural SOM content may inhibit the mobilisation
103 of PFAAs (Hale et al. 2017; Yu et al., 2012). However, to the best of our knowledge, studies
104 demonstrating the effect of varying SOM content on plant uptake upon the application of a
105 carbonaceous adsorbent to farmland soils have not been reported so far. Hence, to better
106 understand the effect of SOM and carbonaceous adsorbents on the PFOA and PFOS uptake by
107 plants, we performed three experiments. The adsorption and immobilisation of PFAAs by
108 granular activated carbon (GAC) was analysed to understand the effect of powdered AC on
109 crop-growth (Jakob et al., 2012). First, the adsorption kinetics of PFOA and PFOS on GAC
110 (with and without 0.04 wt.% HA) were investigated to understand the inhibitive effects of
111 organic matter on the adsorption of PFAAs on GAC. Second, the effect of SOM on the uptake
112 of PFAAs in lettuce was investigated by comparing the uptake in soils with two different SOM
113 concentrations (2.6 and 4.0 wt.%). Finally, the role of SOM was further investigated by
114 analysing the plant uptake factor (PUF) in GAC-treated soils with different SOM
115 concentrations (2.6 and 4.0 wt.%).

116

117 **Materials and Methods**

118 *Chemicals and standards*

119 Technical grades of PFOA and PFOS were purchased from Tokyo Chemical Industry Co. Ltd.
120 (Tokyo, Japan). Analytical standards of PFOA and PFOS including the isotope labelled
121 standards were obtained from Wellington Laboratories Inc. (ON, Canada). Humic acid (HA)

122 was obtained from Sigma Aldrich (Buchs, Switzerland). ENVI-Carb™ (Supelco, Bellefonte,
123 PA, USA), hydrophilic lipophilic balance (HLB) solid phase extraction (SPE) cartridges (0.5
124 g, 6 mL, Waters Co., Ireland), and nylon filter for solvent and sample (0.20 µm, Silicycle Inc.
125 Quebec, Canada) were purchased. Acetic acid, acetone, acetonitrile, methanol,
126 tetrahydrofuran, and water were HPLC grade and obtained from Merck KGaA (Darmstadt,
127 Germany). Polypropylene (PP) bags were used for soil and lettuce sampling.

128

129 *Physico-chemical property analysis of granular activated carbon (GAC)*

130 The GAC was purchased from Samchully Activated Carbon Co. Ltd. (Seoul, Korea). 1 - 2 mm
131 of GAC was used for these adsorption and plant uptake experiments. The chars were dried at
132 140 °C for 72 h before the application to soil. The pH and EC were measured using a pH meter
133 (Jenway 3540, Bibby Scientific Ltd., UK) and EC meter (WalkLAB, Trans Instrument,
134 Singapore) with a suspended GAC (5 g) solution in 100 mL dH₂O. The total pore volume and
135 surface areas of the GAC were analyzed by ASAP2020 (Micromeritics Instrument Co., GA,
136 USA). The elemental analysis of chars was performed by dry combustion (CHN628, Leco Co.,
137 MI, USA). The physico-chemical properties of the GAC are described in Table 1.

138

139 **Table 1** Physico-chemical properties of GAC

pH (1:20)	EC (1:20, dS m ⁻¹)	H/C	O/C	Surface area (m ² g ⁻¹)	Pore volume (cm ³ g ⁻¹)	Pore size (nm)
10.2	7.47	0.42	0.14	1230.0	0.452	24.29

140

141

142 *PFCs adsorption kinetics of GAC*

143 The adsorption experiments were performed in 250 mL PP bottles. Twenty milligrams of GAC
 144 was added to 100 mL PFOA and PFOS aqueous solutions (0.1-100 mg L⁻¹) w/wo 0.04 wt%
 145 HA in a PP bottle. The mixed solution was shaken at 150 rpm in room temperature for 6 days
 146 to reach an equilibrium condition. The solution was filtered and diluted with methanol to fit
 147 within the calibration range for instrumental analysis. The residual concentrations of PFOA
 148 and PFOS were determined by HPLC-MS/MS. A zero adsorption property of HA was
 149 confirmed on the experimental condition without GAC before the GAC adsorption experiment.
 150 Matrix effect of HA was calculated from the following equation (Eq. 1):

$$151 \text{ Matrix effect (\%)} = \left[1 - \left(\frac{\text{Slope of matrix-included adsorption curve}}{\text{Slope of non-matrix included adsorption curve}} \right) \right] \times 100 \quad (\text{Eq. 1})$$

152

153 The slope of adsorption curve was calculated with C₀ and C_e in Figure 1. The adsorption
 154 isotherm for PFOA and PFOS of GAC was calculated using the Freundlich equation and
 155 Langmiur equation. The Freundlich equation can be expressed as follows equation (Eq. 2):

$$156 Q_e = K_F C_e^{n^{-1}} \quad (\text{Eq. 2})$$

157

158 Linearizing equation (Eq. 2), It can be expressed as follows equation (Eq. 3):

$$159 \log Q_e = \frac{1}{n} \log C_e + \log K_F \quad (\text{Eq. 3})$$

160

161 Q_e (μg g⁻¹) is amount of PFOA and PFOS adsorbed per unit weight of adsorbent at equilibrium,
 162 C_e (μg L⁻¹) is equilibrium concentration, K_F [(ng g⁻¹) (μg L⁻¹)⁻ⁿ] is the Freundlich isotherm
 163 constant related to adsorption capacity, n⁻¹ is adsorption intensity.

164 The Langmiur equation can be expressed as follows equation (Eq. 4):

$$165 Q_e = \frac{q_{max} K_L C_e}{1 + K_L C_e} \quad (\text{Eq. 4})$$

166

167 Linearizing equation (Eq. 4), It can be expressed as follows equation (Eq. 5):

168
$$\frac{1}{Q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}K_L C_e} \quad (\text{Eq. 5})$$

169

170 q_{max} is maximum sorption capacity ($\mu\text{g g}^{-1}$), K_L is the constant related to the energy of
171 adsorption ($\text{L } \mu\text{g}^{-1}$).

172

173 ***Soil preparation for plant uptake experiment***

174 Soils with SOM contents of 2.6 wt% and 4.0 wt% were collected from the top soil of
175 agricultural field in Gyeongsang National University and dried for 14 days under fume hood.

176 This soil was analysed and identified to have a background PFOA and PFOS concentration of
177 0.601 and 0.343 $\mu\text{g kg}^{-1}$ in 2.6 wt% SOM of the soil, and 4.802 and 3.701 $\mu\text{g kg}^{-1}$ in 4.0 wt%
178 SOM of the soil, respectively. The soil was divided into two subsamples with and without
179 PFOA and PFOS standard solutions spiked to give a resultant concentration of 1000 $\mu\text{g kg}^{-1}$.

180 All the soil samples were homogenized for 10 days and aged for six months in a green house.

181 Then each of the PFAAs non-spiked and spiked soils were prepared with and without 1 wt%

182 GAC treatment. The lettuce was cultivated with 10-replication in Wagner pots (0.02 m²) that

183 allowed no leaching of irrigated water, each pot containing 2.5 kg of soil. The analysis of SOM,

184 dissolved organic carbon (DOC), pH, EC, and available P₂O₅ was performed using the methods

185 reported by Choi et al. (2019). Briefly, SOM was measured by the Walkley-Black chromic acid

186 wet oxidation method and DOC determined by a TOC analyzer (TOC-L, Shimadzu). The pH

187 and EC for the soil were measured a 50 mL of the five-fold diluted suspended aqueous solution.

188 Available P₂O₅, and exchangeable cations for K, Ca, Mg were analyzed by Lancaster methods

189 and ICP-OES (OPTIMA 5300, PerkinElmer), respectively. The soil properties are described in
190 Table 2.

191

192 **Table 2** Physico-chemical properties of the plantation soil for pot experiment

SOM (wt%)	Texture	DOC (g kg ⁻¹)	pH (1:5)	EC (dS m ⁻¹)	Available P ₂ O ₅ (mg kg ⁻¹)	Exchangeable cations (cmol _c kg ⁻¹)		
						K	Ca	Mg
2.6	Sandy clay loam	0.192	6.57	2.26	324	0.45	14.9	4.34
4.0	Sandy clay loam	0.442	5.97	2.95	300	0.09	0.1	1.68

193

194

195 *Cultivation of lettuce*

196 Lettuce seeds were purchased from Asia Seed Co. Ltd. (Seoul, Korea) and seeds sown in an
197 individual pot that had been treated with 2.5 g of NPK complex fertilizer (21%-8%-10%). The
198 lettuce was cultivated for two months in green house, at which point the aerial part of the lettuce
199 was harvested. The sampled lettuce was chopped and homogenized with dry ice and stored in
200 a freezer at -20 °C until extraction.

201

202 *Sample preparation for the analysis of PFOA and PFOS in soil and crops*

203 The sample preparation for the extraction of soil was followed the reported method by Choi et
204 al. (2017). Briefly, the soil samples were dried at the room temperature under fume hood and
205 passed through 2 mm sieve. 1.0 g of dried soil was extracted with 1.0% aqueous acetic acid
206 (10.0 mL) by mechanical shaking for an hour and sonication at 30 °C for 15 min. The extract
207 was centrifuged for 10 min then the supernatants were collected in a new PP tube. A mixture

208 of 90:10 (v/v) methanol and 1.0 % aqueous acetic acid was added to the extracted soil, then the
209 re-extraction was performed three times. The collected extracts were concentrated to 15 mL
210 then diluted with dH₂O. The HLB cartridges were conditioned using 10 mL of methanol and
211 10 mL of dH₂O and the diluted extract was loaded to the cartridge with a rate of 1.3-1.6 mL
212 min⁻¹, then washed with 5 mL of 30% methanol. The cartridge was eluted with 10 mL of
213 methanol, then the eluent was concentrated to incipient dryness using nitrogen evaporation,
214 before being re-constituted with 1 mL of methanol. The solution was cleaned up with 20 mg
215 of ENVI-CarbTM then filtered with nylon syringe filter. Before the instrumental analysis,
216 isotope labelled injection standards were spiked with 0.010 mg L⁻¹ of ¹³C₄-PFOA and of ¹³C₄-
217 PFOS.

218 The sample preparation method reported method by Choi et al. (2018) was followed for the
219 extraction of crops. Briefly, 10.0 g of crop sample was extracted with 10 mL of 90% aqueous
220 methanol (10 mL x 2) and with 75% tetrahydrofuran in DW (10 mL x 2) by shaking (1 h) and
221 sonication (20 min). The extracts were centrifuged at 3500 rpm and supernatants were collected
222 in a new PP tube. The combined extracts were concentrated to 10 mL under nitrogen then
223 diluted with dH₂O to a 50 mL. The extracts were concentrated under nitrogen, before being
224 reconstituted with 1 mL of methanol. The diluted sample was extracted with HLB SPE
225 cartridges as described for soil and the rest of the process was followed with the same as the
226 soil treatment.

227

228 ***Instrumental analysis and quality assurance***

229 The concentration of PFOA and PFOS were determined using HPLC-MS/MS. The HPLC
230 system was an Agilent HP1100 (Agilent Technologies, CA, USA) coupled to a triple quadruple
231 mass spectrometer API3200TM system (AB SCIEX, ON, Canada) operated using ESI in

232 negative ionization mode. Separation was performed using a Siliachrom C18 column (3 μm ,
233 100 mm x 2.1 mm; Silicycle Co. Ltd, Canada). The optimized analytical conditions are
234 described in supporting information (Table S1). The accuracy of the method was evaluated by
235 determining recoveries associated with relative standard deviation (% RSD) of PFOA and
236 PFOS fortified at 0.05 $\mu\text{g kg}^{-1}$ in lettuce and soil. The recoveries of PFOA and PFOS were
237 70.4-78.7% and the interday precision ranged from 5.6-8.3%, respectively. The linearity of
238 PFOA and PFOS was acceptable for quantitation across the range of 0.010 to 1.000 ng mL^{-1}
239 ($R^2 > 0.997$). The method limit of quantitation (mLOQ) of PFOA and PFOS were determined
240 at 0.001 and 0.010 $\mu\text{g kg}^{-1}$ for lettuce and soil, respectively. More detailed information is
241 described in Table S2.

242

243 ***Calculation on plant uptake factor (PUF)***

244 The plant uptake factor is represented as the ratio between concentrations of a chemical analyte
245 determined in plant tissue and soil, it can be expressed as follows equation (Eq. 6) (Liu et al.
246 2019):

$$247 \quad PUF = \frac{\text{Concentration in crop}}{\text{Concentration in soil}} \quad (\text{Eq. 6})$$

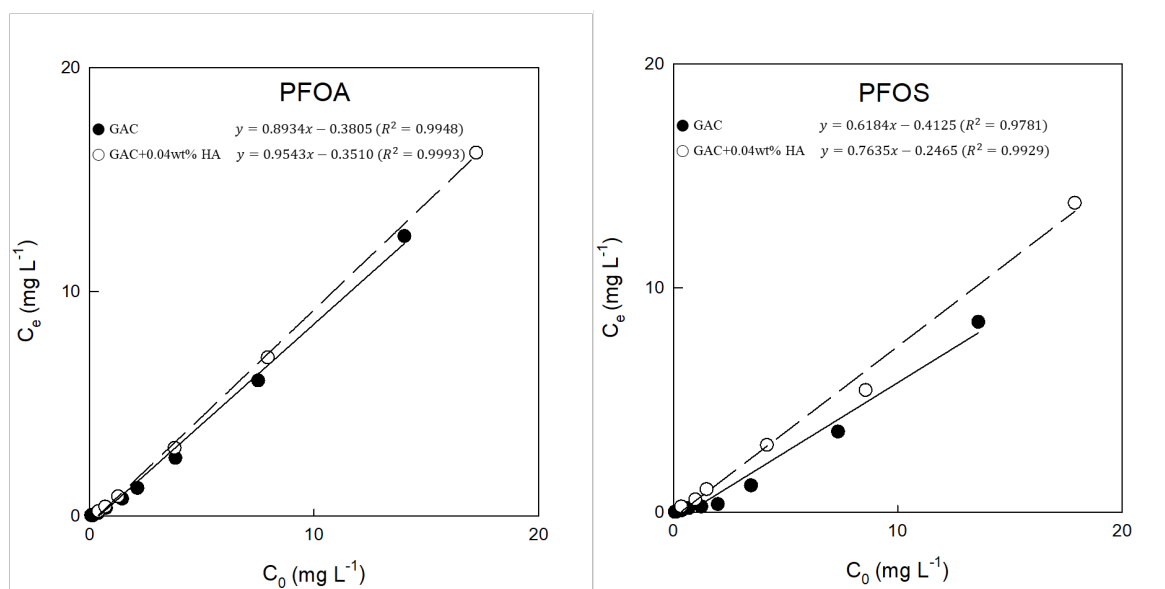
248

249 **Results and discussion**

250 ***PFOA and PFOS adsorption properties of GAC***

251 The adsorption capacity of GAC was calculated using the equilibrium concentrations obtained
252 from the various initial concentrations of aqueous solutions of PFOA and PFOS. Fig. 1 shows
253 a plot of equilibrium concentrations of PFOA and PFOS against their initial concentrations in
254 an aqueous solution containing GAC, with and without 0.04 wt.% HA. With an increase in the

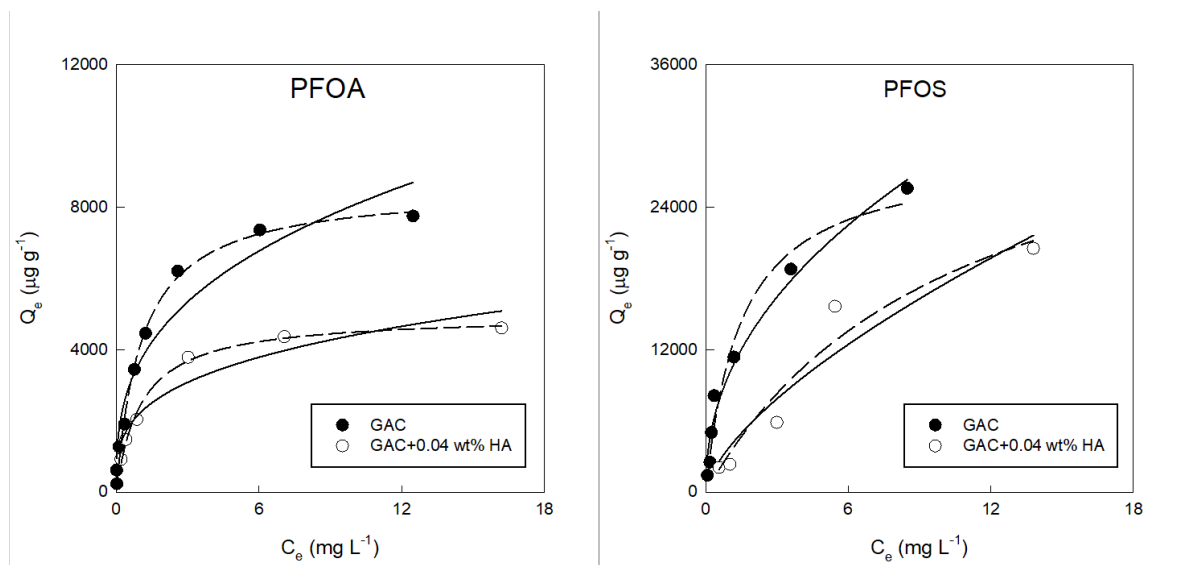
255 initial PFOA and PFOS concentration, the equilibrium concentration increased linearly, with
 256 higher values observed in the solution containing HA compared to that containing only GAC.
 257 It should be noted that 0.04 wt.% HA was initially tested separately and was found to have no
 258 impact on the adsorption of PFOA and PFOS.



259
 260 **Fig. 1** Adsorption properties of GAC for PFOA and PFOS. C_0 (mg L^{-1}) was the initial
 261 concentration of PFOA and PFOS and C_e (mg L^{-1}) was the equilibrium concentration of
 262 PFOA and PFOS.

263
 264 In Fig. 1, the slopes of the plots of PFOA and PFOS adsorption on GAC adsorption are higher
 265 for the solutions containing 0.04% HA. The slopes were used to calculate the matrix effects of
 266 0.04 wt.% HA on the adsorption of PFOA (-6.8%) and PFOS (-23.5%), on GAC. Therefore,
 267 the presence of HA appeared to reduce the adsorption of both PFAAs on GAC, with the effect
 268 being more pronounced for PFOS than PFOA.

269



270

271 **Fig. 2** Sorption isotherms of PFOA and PFOS on GAC. Q_e ($\mu\text{g g}^{-1}$) was amount of PFOA and
 272 PFOS adsorbed per unit weight of GAC equilibrium and C_e (mg L^{-1}) was equilibrium
 273 concentration in water. The solid lines are the fitted Freundlich isotherms and the dashed
 274 lines are the fitted Langmuir isotherms

275

276 The adsorption isotherms of PFOA and PFOS fit both the Langmuir and the Freundlich
 277 equations (Fig. 2). The maximum adsorption capacities (q_{max}) of GAC for PFOA and PFOS
 278 were 9.091 mg g^{-1} and 27.778 mg g^{-1} , respectively, in the solution without HA. The values
 279 decreased to 5.208 mg g^{-1} and 17.241 mg g^{-1} for PFOA and PFOS respectively, when tested in
 280 the solution containing 0.04 wt.% HA. Thus, the addition of 0.04 wt.% HA resulted in a
 281 decrease in q_{max} for both PFOA (43%) and PFOS (38%). The adsorption intensities (K_L) for
 282 PFOA and PFOS in the solution without HA were $0.76 \text{ mL } \mu\text{g}^{-1}$ and $0.74 \text{ mL } \mu\text{g}^{-1}$, respectively.
 283 The K_L for PFOS in the solution with HA decreased to $0.21 \text{ mL } \mu\text{g}^{-1}$ (Table 3). The adsorption
 284 capacity (K_F) and adsorption intensity (n^{-1}) values are presented in Table 3.

285 The respective $\log K_{FS}$ [$(\mu\text{g kg}^{-1}) (\mu\text{g kg}^{-1})^{-n}$] values of PFOA and PFOS in the HA-free solution,
 286 were 5.41 and 5.21 and were similar to previously reported values (Silvani et al., 2019). For
 287 the solution containing 0.04 wt.% HA, the respective $\log K_{FS}$ values decreased slightly to 5.36
 288 and 4.05. Wang et al. (2015) reported that HA competes for the adsorption of PFAAs in soil,
 289 which explains the decrease in K_F values in the solution containing HA.

290

291 **Table 3** Constants of the Langmiur and Freundlich isotherm equations of GAC for PFOA
 292 and PFOS

	Chemicals	Langmiur constant			Freundlich constant		
		q_{\max}^a	K_L^b	R^2	$\log K_F^c$	$n^{-1 d}$	R^2
GAC	PFOA	9.091	0.76	0.999	5.41	0.383	0.897
	PFOS	27.778	0.74	0.968	5.21	0.585	0.921
GAC+0.04 wt% HA	PFOA	5.208	0.75	0.995	5.36	0.325	0.927
	PFOS	17.241	0.21	0.893	4.05	0.802	0.949

293 ^a q_{\max} : maximum sorption capacity (mg g^{-1}); ^b K_L : energy of adsorption ($\text{mL } \mu\text{g}^{-1}$); ^c K_F :
 294 Freundlich isotherm constant, adsorption capacity [$(\text{ng g}^{-1}) (\mu\text{g L}^{-1})^{-n}$]; ^d n^{-1} : adsorption
 295 intensity.

296

297 The adsorption intensity/heterogeneity is represented by n^{-1} . If $n^{-1} > 1$, it indicates that chemical
 298 adsorption is the dominant process; if $n^{-1} < 1$, physical adsorption is the dominant process,
 299 which is most effective when the value is 0.1–0.5 (Bang and Kim, 2017). Here, all the values
 300 of n^{-1} for PFOA and PFOS ranged from 0.325–0.582, except for PFOS in the solution with
 301 0.04% HA where the value was 0.802. These results indicate that in the absence of HA, GAC
 302 effectively adsorbed PFOA and PFOS through physical adsorption. However, the physical
 303 adsorption efficiency of GAC for PFOS decreased in the presence of 0.04 wt.% HA, suggesting
 304 that HA could interfere with the adsorption efficiency of GAC for PFOS, and with GAC

305 treatment in soil with high SOM content. Thus, the immobilization ability of GAC for PFOS
306 in high SOM containing environment would be lower than that for PFOA.

307

308 *Effect of SOM on the uptake of PFOA and PFOS in lettuce*

309 The uptake of PFOA and PFOS in lettuce was investigated under two conditions (2.6 wt.% and
310 4.0 wt.% of SOM) and two concentrations ('high' and 'low') of PFOA and PFOS (Table 4).

311 No visible evidence of phytotoxicity due to PFOA/PFOS was observed in the lettuce during

312 cultivation. In the soil with 2.6 wt.% SOM, the average PUFs for low and high concentrations

313 of PFAAs were 0.629 and 0.252 for PFOA and PFOS respectively. In the soil with 4.0 wt.%

314 SOM, the PUFs were approximately two times lower: 0.353 (PFOA) and 0.108 (PFOS). Plant

315 uptake factors have been shown to vary by crop type, part of the crop, and crop growth rates

316 (Ghisi et al. 2019; Xiang et al. 2018). They are also influenced by the leaching and adsorption

317 potential of the soil which is influenced by soil properties such as pH, mineral content, anion

318 exchange capacity and SOM (Li et al. 2019; Wei et al. 2017; Xiang et al. 2018). The log K_{ds}

319 values of soil were reported in the range of -0.97-1.69, and -1.0-35.3, for PFOA and PFOS

320 respectively, with higher K_{ds} values observed in soil with a greater SOM content

321 (Kupryianchyk et al. 2015; Martz et al. 2019; Milinovic et al. 2015; Silvani et al. 2019;

322 Zareitalabad et al. 2013). This explains why lower PUFs for PFOA and PFOS were observed

323 in the soil containing 4.0% SOM compared to the soil with 2.6% SOM. In addition, the PUF

324 for PFOA was consistently higher than that for PFOS. The low PFAA-contaminated soil

325 showed similar PUFs for both PFOA and PFOS in the soil with 2.6 wt.% SOM (0.349 and

326 0.297, respectively) and with 4.0 wt.% SOM (0.127 and 0.115, respectively). However, the

327 high PFAA-contaminated soil showed a much larger increase in the PUF for PFOA compared

328 to that for PFOS in both the 2.6 wt.% SOM soil (0.908 and 0.207 respectively) and 4.0 wt.%

329 SOM soil (0.578 and 0.100 respectively). It could be because of the difference in leaching of
 330 PFOA and PFOS in the soil environment (Milinovic et al., 2015; McSachlan et al., 2019).

331

332 **Table 4** The PUFs of PFOA and PFOS in lettuce cultivated on 2.6 wt% and 4.0 wt% SOM of
 333 soils with or without GAC

		PUF		Lettuce		Soil	
		(Average±SE)		(Average±SE, µg kg ⁻¹)		(Average±SE, µg kg ⁻¹)	
		PFOA	PFOS	PFOA	PFOS	PFOA	PFOS
2.6 wt% SOM	Low	0.349 ±0.062	0.297 ±0.059	0.210 ±0.040	0.102 ±0.027	0.601 ±0.071	0.343 ±0.070
	High	0.908 ±0.135	0.207 ±0.048	873.3 ±130.0	226.6 ±41.1	961.4 ±104.5	1,093 ±195.0
	High+GAC ^a	0.006 ±0.002	0.005 ±0.002	5.617 ±2.155	5.936 ±2.185	-	-
PUF suppression by GAC		99.4%	97.4%				
4.0 wt% SOM	Low	0.127 ±0.035	0.115 ±0.039	0.609 ±0.128	0.426 ±0.278	4.802 ±0.970	3.701 ±0.649
	High	0.578 ±0.116	0.100 ±0.027	618.3 ±124.9	85.70 ±12.96	1,070 ±160.8	859.2 ±92.14
	High+GAC ^a	0.079 ±0.021	0.023 ±0.006	84.45 ±11.22	19.61 ±6.388	-	-
PUF suppression by GAC		86.3%	77.1%				

334 ^a GAC was treated to 1 wt% of dried soil

335

336 ***Effect of GAC treatment on the PUF for PFOA and PFOS in lettuce***

337 When comparing the PUFs for PFOA and PFOS in soils treated with GAC, the results showed
 338 a significant reduction in PFAA uptake (Table 4). The PUFs in the soil with 2.6 wt.% SOM
 339 reduced to 0.006 (PFOA) and 0.005 (PFOS). These results were mirrored in the soil containing
 340 4.0 wt.% SOM with PUFs of 0.079 (PFOA) and 0.023 (PFOS). These results highlight the

341 effectiveness of GAC for active immobilisation of PFOA and PFOS in contaminated soils.
342 When GAC was applied to the high PFAA-contaminated soils, the PUFs in the soil with 2.6%
343 SOM were calculated to be below 0.01, and were reduced by 99.4% (PFOS) and 97.4% (PFOA);
344 in the soil with 4.0% SOM, relatively higher PUFs were observed, reduced by 86.3% (PFOS)
345 and 77.1% (PFOA). As concluded by Hale et al. (2017), GAC treatment in soil can significantly
346 reduce leaching of PFAAs. This is considered to have contributed significantly to the reduction
347 in PUF observed in the lettuce samples. In addition, the results show that the PUFs for PFOA
348 and PFOS were more effectively suppressed in the soil with low SOM than in the soil with
349 high SOM. This could be explained by the adsorption kinetics experiment of GAC with HA,
350 as an increase in SOM resulted in decreased adsorption of PFAAs on GAC. Therefore, the
351 adsorption of PFAAs by GAC in soil with high SOM was reduced, outweighing the benefits
352 of having additional SOM. Although the PUFs were relatively higher in GAC-treated soil with
353 high SOM than with low SOM, GAC treatment could effectively reduce approximately 4.3-
354 155-fold of the PFAA residues in crops. Thus, GAC application could be considered to reduce
355 the dietary exposure of PFAAs from crop consumption. Further studies are necessary on the
356 appropriate GAC application procedures in farmland soil with varying SOM content.

357

358 **Conclusion**

359 Soil organic matter was shown to adsorb PFOA and PFOS, and its presence helped reduce the
360 uptake of PFAAs from soil into crops. Similarly, the addition of GAC to soil helped reduce the
361 uptake of PFAAs. It was demonstrated that GAC can effectively adsorb both PFOA and PFOS
362 and therefore suppress the crop-uptake of PFAAs (up to 99.4% for PFOA in 2.6% SOM soil.
363 However, a higher SOM content as well as the presence of HA reduced the effectiveness of
364 GAC and ultimately its capacity to adsorb PFOA and PFOS. The PUF of lettuce grown in soil

365 with higher SOM indicated a suppression in the ability of GAC to adsorb PFAAs. Hence, the
366 existing SOM content of the soil should be considered when a carbonaceous adsorbent is
367 applied to ensure effective immobilisation and successful reduction of crop-uptake of PFAAs.
368 Further research is required to better understand the effectiveness of GAC to immobilise
369 PFAAs in soils with various amounts of SOM. This could help establish a correlation factor
370 for GAC immobilization of PFAAs based on the SOM content.

371

372

373 **Appendix. Supplementary data**

374 Supplementary data related to this article can be found at Supporting Information

375

376 **Credit authorship statement**

377 Lee DY performed the experiments and data collection; Choi GH analyzed the PFAAs in crop;
378 Choi IW, Oh KY and Seo DC analyzed the data for the adsorbent; Megson D discussed the
379 results; Kim JH wrote and revised the manuscript. All authors read and approved the final
380 manuscript.

381

382 **Conflicts of Interest**

383 The authors declare no conflicts of interest

384

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