

# **Management of per- and polyfluoroalkyl substances (PFAS) - laden wastewater sludge in Maine:**

## **Perspectives on a wicked problem**

Running title: PFAS in Maine

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## 1 Abstract

2 This article discusses the challenges and potential solutions for managing wastewater sludge that contains  
3 per- and polyfluoroalkyl substances (PFAS), using the experience in Maine as a guide toward addressing the  
4 issue nationally. Traditional wastewater treatment, designed to remove excess organic waste and nutrients, does  
5 not eliminate persistent toxic pollutants like PFAS, instead partitioning the chemicals between discharged  
6 effluent and the remaining solids in sludge. PFAS chemistry, the molecular size, alkyl chain length, fluorine  
7 saturation, charge of the head group, and the composition of the surrounding matrix influence PFAS  
8 partitioning between soil and water. Land application of sludge, incineration, and storage in a landfill are the  
9 traditional management options. Land application of Class B sludge on agricultural fields in Maine peaked in  
10 the 1990s, totaling over 2 million cubic yards over a 40-year period, and has contaminated certain food crops  
11 and animal forage, posing a threat to the food supply and the environment. Additional Class A EQ (Exceptional  
12 Quality) composted sludge was also applied to Maine farmland. The State of Maine banned land application of  
13 wastewater sludge in August 2022. Most sludge was sent to the state-owned Juniper Ridge Landfill which  
14 accepted 94,270 tons of dewatered sludge in 2022, a 14% increase over 2019. Between 2019 and 2022 the sum  
15 of PFOS and PFOA concentrations in sludge sent to the landfill ranged from 1.2 to 104.9 ng/g dw. In 2022 the  
16 landfill generated 71.6 million liters of leachate. The concentration of sum of 6 PFAS in the leachate increased  
17 sixfold between 2021 and 2022 reaching 2,441 ng/L. The retention of PFAS within solid-waste landfills and the  
18 potential for long-term release of PFAS through liners into groundwater requires ongoing monitoring. Thermal  
19 treatment, incineration, or pyrolysis can theoretically mineralize PFAS at high temperatures, yet the strong C-F  
20 bond and reactivity of fluorine requires extreme temperatures for complete mineralization. Future alternatives  
21 may include interim options such as pre-conditioning PFAS with non-polar solvents prior to immobilization in  
22 landfills, removing PFAS from leachate, interrupting the cycle of PFAS moving from landfill, via leachate, to  
23 wastewater treatment, then back to the landfill via sludge. Long-term solutions may involve destructive  
24 technologies such as electron beam irradiation, electrochemical advanced oxidation, or hydrothermal

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1 liquefaction. The article highlights the need for innovative and sustainable solutions for managing PFAS-  
2 contaminated wastewater sludge.

### 3 I. INTRODUCTION

4 PFAS (per- and polyfluoroalkyl substances) are anthropogenic organic chemicals that contain extremely  
5 strong carbon-fluorine bonds, which persist in the environment decades beyond their intended commercial use,  
6 and circulate repeatedly through our natural and engineered environments. Today, the land application of  
7 PFAS-laden wastewater sludge has impacted agricultural fields across the U.S. and caused contamination of  
8 food crops and animal forage, threatening the health of our food supply and our population. Maine is at the  
9 forefront of a precedent-setting response to address PFAS contamination. This paper reviews the traditional  
10 approaches that are currently used for managing PFAS-contaminated wastewater sludge, from land application  
11 on agricultural fields to disposal in landfills and considers the emerging options for a sustainable solution to  
12 remove PFAS from its ongoing circulation through the environment.

13 Wastewater treatment, designed to remove nutrient pollutants and pathogens that are harmful to human  
14 health and aquatic ecosystems, and excess organic matter that depletes the oxygen in receiving waters, was  
15 never designed to eliminate persistent toxic pollutants such as PFAS. Instead, wastewater treatment  
16 redistributed PFAS to our agricultural fields and surface waters. In 1972, amendments to previous legislation  
17 created the Clean Water Act, with “...*the national goal that the discharge of pollutants into the navigable*  
18 *waters be eliminated by 1985*”<sup>1</sup>. Prior to the Clean Water Act, municipal and industrial waste streams were  
19 screened for solids, then discharged directly into surface waters, where excess nutrients and oxygen demand  
20 fueled ecologically disruptive plant growth and decomposition, depleting oxygen from aquatic systems. Solids  
21 floated in mats on the surface of receiving waters and lined the shores with fetid deposits<sup>2</sup>. After the Clean  
22 Water Act, wastewater treatment removed a large portion of the organic waste, solids and pathogens from the  
23 effluent subsequently discharged into surface waters. The solids, termed wastewater sludge, began to

1 accumulate near treatment plants. Land application of the sludge to agricultural fields as a source of nutrients  
2 and carbon-rich soil amendments, became a preferred solution and was marketed as ‘beneficial reuse’.

3 The health threats from persistent metals or bacterial pathogens that could be present in sludge were  
4 recognized and addressed by concentration limits set by the U.S. Environmental Protection Agency (EPA), Part  
5 503 Rule <sup>3,4</sup>, and sludge meeting those limits were given the euphemism biosolids. (The EPA now considers the  
6 terms sludge and biosolids to be interchangeable<sup>5</sup>, the authors have chosen to use sludge, the universally  
7 recognized term). Sludge that met concentration limits for nine trace metals, had no detectable biological  
8 pathogens, and reduced vector attraction were deemed of exceptional quality and termed Class A EQ. Class B  
9 sludge met the metals requirements, yet had detectable pathogens which required delays of food crop harvest of  
10 30 days to 30 months following field application<sup>3</sup>. Class B sludge was generally used on fields of hay and feed  
11 corn at dairy farms.

12 However, the original federal guidance on land application of sludge did not consider the presence of  
13 emerging contaminants (Part 503 regulations were later revised to address dioxins found present in the sludge).  
14 PFAS, along with other toxic persistent organics and pharmaceuticals, are not destroyed by current wastewater  
15 treatment technologies. Instead, wastewater treatment partitions intact or reformulated PFAS to either  
16 discharged effluent or to sludge<sup>6</sup>. PFAS were not included in the Part 503 land-application regulations for  
17 sludge, and the spreading of PFAS-contaminated sludge on farm fields, beginning in the late 1970s, continues  
18 to impact crops and groundwater today.

19 PFAS were created and initially marketed before chemical regulation was enacted in the United States.  
20 Commercial use of PFAS began in the 1950s in fire-fighting foams, nonstick cookware, water-resistant  
21 clothing, carpet protectors, food packaging, and painting materials<sup>7</sup>. Their environmental persistence, water  
22 solubility, and adsorption to sediments and minerals enhanced worldwide dispersion<sup>8</sup>. The superior  
23 effectiveness of eight-carbon PFOA and PFOS in creating PFAS that repelled both oil and water led to their  
24 commercial dominance until the early 2000s when their environmental persistence and the emerging public  
25 awareness of their toxicity led to replacement with PFAS believed to be less biologically persistent.

1 PFAS chemicals are associated with immune suppression, metabolic effects, and limited findings on  
2 neurodevelopmental effects. The results from early toxicity studies which began in the 1960s and associated  
3 immune suppression with PFOA and PFOS exposure were not publicly released<sup>9</sup>, and subsequently omitted  
4 from initial exposure limits<sup>10,11,12</sup>. Studies since 2000 have corroborated and expanded those earlier findings. A  
5 study in Faroe Islands children reported that a 2-fold increase in PFAS serum concentrations in 5-year-old  
6 children correlated with a 49% decline in humoral antibody response to tetanus and diphtheria vaccines<sup>10</sup>.  
7 Looker et al. (2014)<sup>13</sup> reported a negative correlation in adults between PFOA concentrations in blood serum  
8 and antibody production in response to A/H3N2 influenza vaccination, preventing sufficient antibody  
9 production for long-term protection against the virus. Elevated serum PFAS concentrations were associated  
10 with modest increases in total cholesterol and low-density lipoprotein cholesterol<sup>14</sup>. A study of pregnant women  
11 reported a positive association between PFOS and related analytes and impaired glucose tolerance and between  
12 PFOA and total cholesterol<sup>15</sup>. Min et al. (2012)<sup>16</sup> reported elevated blood pressure in adults with background  
13 PFOA blood levels, while no association was found in children<sup>17</sup>. The West Virginia C-8 study of PFOA  
14 contamination of groundwater near a DuPont Teflon manufacturing plant found a probable link between PFOA  
15 exposure and testicular and kidney cancers<sup>18</sup>. Further, a subsequent population-based study reported a two-fold  
16 increased risk of kidney cancer for those with the greatest PFOA serum concentrations<sup>19</sup>. Hepatotoxicity and  
17 developmental toxicity associated with PFAS exposure found in rodent studies may not be directly applicable to  
18 humans given differences in nuclear receptors involved in the toxicological response (PPAR-alpha) in the liver,  
19 and the extended half-times of PFAS in humans<sup>20</sup> (half-times in human serum exceed three years).

20 PFAS contamination of farm fields from wastewater sludge is a national problem, first identified in Maine  
21 in 2016 when water from a public monitoring well near the Stoneridge Dairy Farm in southern Maine was reported  
22 to have 140 ng/L combined PFOS and PFOA. The subsequent investigation on the dairy farm by the Maine  
23 Department of Environmental Protection (DEP) found combined PFOA (perfluorooctanoic acid) and PFOS  
24 (perfluorooctane sulfonate) concentrations up to 1,420 ng/L in milk, over six times greater than Maine's limit for  
25 unadulterated milk<sup>21</sup> (210 ng/L). In the farm's surface soil PFOS and PFOA combined reached approximately

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1 900 ng/g dw (dry weight)<sup>22</sup>, over 25 times greater than Maine's screening level for contaminated soils<sup>23</sup>. In the  
2 summer of 2020, another collection of PFAS-contaminated fields were found in central Maine. PFOS  
3 concentrations in milk from a farm in the town of Fairfield reached 32,200 ppt, 153 times greater than the PFAS  
4 action threshold for adulterated milk<sup>21</sup>. Subsequent testing of nearby wells in Fairfield with possible exposure to  
5 PFAS found 39% of the 456 private drinking water wells tested contaminated with the sum of 6 PFAS (PFOA,  
6 PFOS, PFHxS perfluorohexane sulfonic acid, PFHpA or perfluoroheptanoic acid, PFNA perfluorononanoic acid,  
7 PFDA perfluorodecanoic acid) exceeded the Maine Interim Drinking Water Standard of 20 ng/L<sup>24</sup>. In 24 wells  
8 PFAS concentrations were between 1,000 and 10,000 ng/L and exceeded 10,000 ng/L in 15 private drinking water  
9 wells in the Fairfield region<sup>25</sup>. Across Maine, 23% of the 2,068 private drinking water wells suspected of PFAS  
10 contamination due to proximity to an identified PFAS source were reported to have sum of 6 PFAS greater than  
11 the Maine Interim Drinking Water Standard<sup>26</sup>.

Maine state agencies responded to the PFAS crisis on multiple fronts, combining the efforts of Maine's  
12 Department of Environmental Protection, Center for Disease Control, Department of Agriculture, Conservation  
13 and Forestry, Department of Inland Fisheries and Wildlife, the University of Maine, and the University of Maine  
14 Cooperative Extension. Then between 2021 and 2022 the Maine Legislature passed 10 PFAS bills on topics  
15 including lowering the Interim Maine PFAS Drinking Water Standard to 20 ng/L (sum of 6 PFAS), providing  
16 funding for monitoring and clean-up of contaminated sites, supporting farmers with contaminated fields, ending  
17 all land application of wastewater sludge or compost made from sludge, and requiring all manufacturers to report  
18 PFAS presence in products and to ban unnecessary sales by 2030. Despite the widespread media coverage of  
19 PFAS in Maine, residents may still lack awareness of the issue and this knowledge gap may limit public decision  
20 makers opportunities to fund long-term efforts to address PFAS. Further, distrust of scientific information about  
21 PFAS contamination may compound the issue; in two surveys of Maine residents (spring and fall 2022) conducted  
22 by the authors, 13% of respondents indicate that the PFAS problem in Maine has been overstated<sup>27</sup>. We also  
23 documented the knowledge gap present in Maine where respondents indicated on a scale of 0 (know nothing) to  
24 100 (know everything) that they currently know on average 35 but that they should know 80 about PFAS.  
25



## II EXAMINATIONS

### A. *PFAS partitioning in wastewater and soil*

Understanding the fate and transport of PFAS in the environment gives insight into the chemicals' movement in soil, groundwater, and wastewater. The chemistry of PFAS influences their tendency to partition to either the soil solid phase or porewater, water contained in the gaps between grains of soil, following the land application of sludge or to partition to the solids in wastewater sludge or clear effluent during the wastewater treatment process. This chemical behavior then defines PFAS movement through soil into ground or surface waters and entry into the food web via plant uptake or direct absorption by biota. PFAS contain a non-polar, hydrophobic alkyl chain of carbon atoms with all or some of the hydrogen atoms replaced with fluorine<sup>7</sup>. The unique strength of the carbon-fluorine bonds in this alkyl "tail" makes PFAS indestructible under normal environmental conditions. PFAS also contain a hydrophilic, polar head group, commonly sulfonate or carboxylate.

PFAS partitioning between water and solids is influenced by molecular size and alkyl chain length, fluorine saturation of the alkyl chain, the charge of the head group and the composition of the surrounding matrix, be it soil or sludge. The PFAS molecule becomes more hydrophobic as the perfluorinated chain lengthens. Larger molecules (molecular weight > 350 g/mol) may improve PFAS adhesion by creating multiple points of contact with soil surfaces<sup>28,29</sup>. Perfluorinated sulfonates (PFSA) adsorb more readily onto activated carbon<sup>30,31</sup>, sludges<sup>32,33</sup>, and sediments<sup>34,35</sup> than do perfluorinated carboxylates (PFCA) with the same carbon numbers<sup>36,37</sup>.

Charge strongly influences PFAS adsorption to particles. Perfluoroalkyl acids like PFSA and PFCA in the environment become anionic (negatively charged) in a pH-dependent manner, and become more water soluble, and so partition more to the soil porewater, with higher pH<sup>38,39</sup>. The pH sensitivity of short-chain PFAS, with five or fewer carbons, is lower than that of long-chain PFAS, with six or more carbons possibly because of their



1 higher solubility in water<sup>40</sup>. Negatively charged organic matter and clay bond more strongly to cationic (positively  
2 charged) and zwitterionic (compounds with both positively and negatively charged functional groups) PFAS,  
3 making the sorption of cationic PFAS positively correlated with organic matter.

4 The  $K_d$  value, also known as the *soil partition coefficient*, is a partitioning index for compounds that can  
5 be helpful to estimate their mobility in systems like soil or sludge that have both solid and aqueous components.  
6 The  $K_d$  values of longer chain PFAS (C-6 or greater) increase with chain length, driven by their hydrophobicity,  
7 and enhanced by the presence of soil organic carbon<sup>8,34,40</sup>. Further, the presence of zwitterionic head groups  
8 increases  $K_d$  value and soil attraction; making them less susceptible to leaching and increasing soil retention<sup>35</sup>.  
9 The  $K_d$  values of shorter-chain PFAS do not vary with carbon length, thus their affinity to soil primarily depends  
10 on charged head groups, though the alkyl tail of PFAS can adsorb to nonpolar sites on mineral surfaces due to  
11 weak hydrophobic affinity<sup>41</sup>. Shorter chain PFAS frequently partition into soil porewater and show increased  
12 mobility<sup>40</sup>. Decreasing soil grain size and the fine texture of silt and clays increases anionic PFAS adsorption and  
13 decreases movement through porewater<sup>42</sup>.

14 The fate and transport of PFAS collected in wastewater sludge reflects partitioning during wastewater  
15 treatment, and partitioning predicts the subsequent movement of PFAS through soil following land application of  
16 the sludge or into the leachate, water that has percolated through and collected beneath landfills, after landfilling.  
17 At the wastewater treatment plant, shorter chain PFAS, especially PFCA, are more likely to be discharged with  
18 the liquid effluent while longer chain PFAS partition more strongly into the sludge solids. When sludge is applied  
19 to soil, the ratio of material that infiltrates the soil versus running off to the nearest surface water body depends  
20 on precipitation, slope, and method of application. However, the higher solubility of the shorter chain PFAS in  
21 the water phase increases their concentration in porewater and infiltration to groundwater, and increases the  
22 likelihood of plant uptake, while the longer chain compounds tend to remain bound to the solids at shallow soil  
23 depths. Similarly, in landfills, transport of the shorter chain PFAS into the leachate is enhanced relative to the  
24 longer chain PFAS. The high concentration of longer chain, especially C-8, PFAS in groundwater and biota

1 reflects the decades-long dominance of these PFAS in consumer products and production rather than increased  
2 environmental transport.

### 3 4 **B. Traditional approaches to managing wastewater sludge**

5 Each day in the U.S. roughly 15,000 wastewater treatment plants (WWTPs) process over 24,000  
6 million gallons of wastewater per day, removing harmful nutrients and organic matter from the incoming waste  
7 stream and creating relatively clean effluent that is discharged into surface waters. The residual sludge, solids  
8 containing nutrients, organic matter, metals and persistent toxic chemicals, is a byproduct of the treatment process.  
9 In 2018 an estimated 5.8 million dry metric tons of wastewater sludge was generated nationally with 24,300 dry  
10 tons produced in Maine. Current management options for sludge are limited to disposal in a solid waste landfill,  
11 incineration, or land application to agricultural fields or forest. Options exist for further biological stabilization  
12 via anaerobic/aerobic digestion or composting which can change the sludge properties, and reduction in total mass  
13 of the sludge by partial dewatering (e.g., belt press, centrifugal dewatering) but these processes do not remove  
14 PFAS, and must still be followed by one of the existing management options. Disadvantages impair all three  
15 management options.

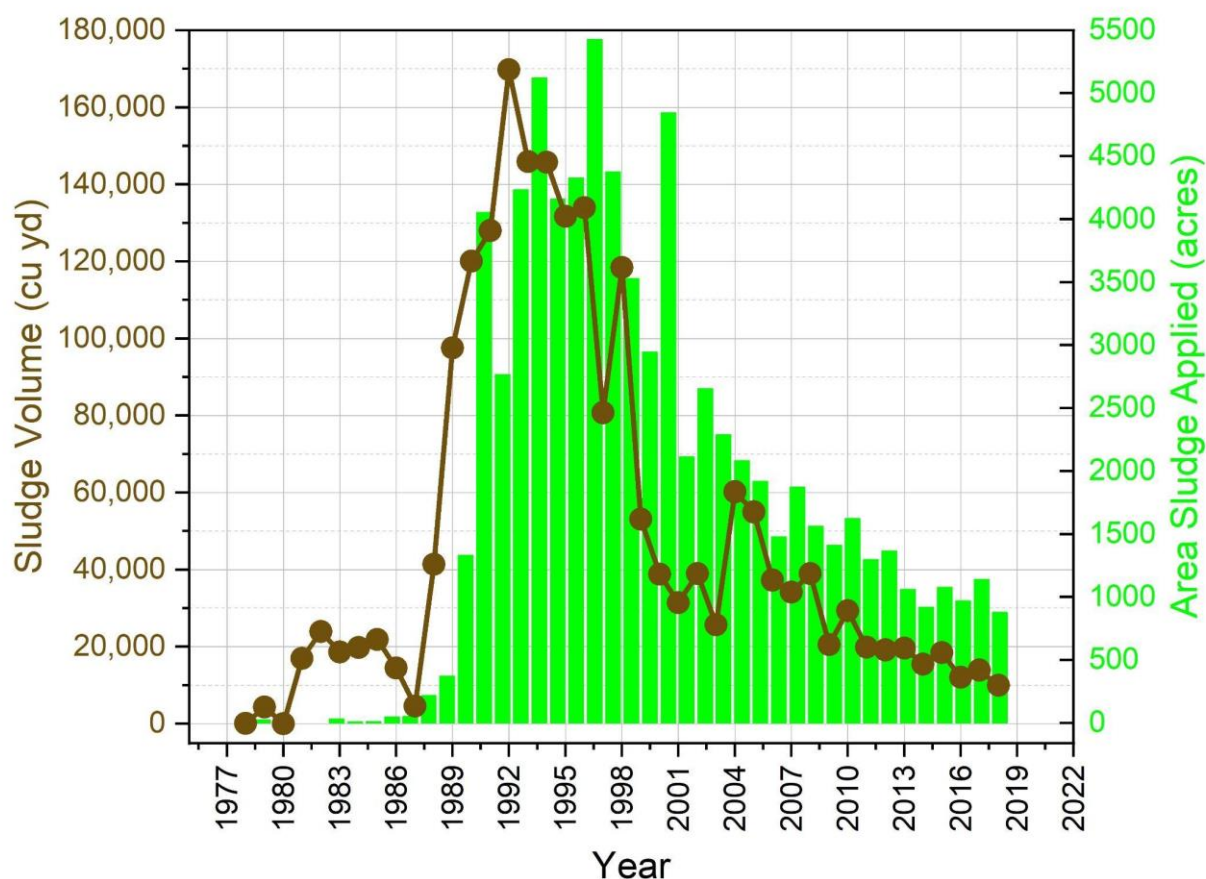
#### 16 a. *Land Application of Wastewater Sludge in Maine*

17 Following the Clean Water Act requirement to expand wastewater treatment, Maine and other states  
18 sought options for the sludge accumulating from the new wastewater treatment plants. Land application of  
19 stabilized or composted sludge on pasture and cropland appeared a functional alternative that supplied essential  
20 plant nutrients to the soil while adding organic carbon as a soil amendment. Farmers were encouraged to  
21 participate in the free sludge distribution program<sup>43</sup>.

22 Between 1978 and 2018 over two million cubic yards of wastewater sludge were applied as fertilizer and  
23 soil amendment to over 1,300 licensed agricultural fields in Maine<sup>44</sup>. As seen in Fig. 1, land application of sludge  
24 peaked between 1989 and 1998, averaging 127,000 cu yd per year over that 10-year period. One dairy farm in the

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1 heavily impacted Fairfield area of central Maine received over 56,000 cu yd of mixed municipal and industrial  
2 wastewater sludge<sup>44</sup>.



5  
6 **Figure 1.** Land application of Class B wastewater sludge in Maine peaked between 1989 and 1998; sludge  
7 volume (cu yd) given in line and closed circles, the acres of agricultural land where sludge was applied each  
8 year is given in green bars. Data from Maine Department of Environmental Protection<sup>44</sup>.

9  
10 Class B sludge was land applied to a small percentage of Maine's total agricultural land, never exceeding  
11 0.8%, despite the decline in total cropland in Maine from approximately 611K acres in 1982 to 473K acres in  
12 2017<sup>45,46,47,48</sup>. Class B sludge was generally used on dairy farms for hay and feed corn crops.

1 By 2018 Class B sludge comprised only 9% of the total sludge generated and processed in Maine. In the  
2 same year, Class A EQ sludge, primarily sludge originating both in and outside of Maine that was composted at  
3 Casella Organics Compost Facility in Unity, Maine, and other smaller composting facilities, totaled 7,112 dry US  
4 tons, 29% of the state's sludge total<sup>49</sup>. Use of Class A EQ sludge did not require a permit under EPA Part 503,  
5 and it was used throughout the state on agricultural lands, gardens, lawns, and sport fields. Records of the final  
6 application sites for Class A EQ sludge are not available.

7 As a first step in defining the extent of PFAS contamination in Maine's agricultural fields, the legacy of  
8 the 40-year period of land application of sludge (1978 – 2018), Maine began sampling soil to define the extent of  
9 PFAS contamination in all agricultural fields that had received Class B wastewater sludge and began analyzing  
10 the current PFAS concentrations in sludge generated by WWTPs. Together those two findings would determine  
11 whether ongoing sludge applications to specific agricultural fields would be prohibited because the combined  
12 PFAS concentration in the soil and in the proposed sludge application would exceed allowable beneficial use  
13 screening levels for PFAS, defined by the state as PFOA, 2.5, PFOS, 5.2, and PFBS 1,900 ng/g, dry weight<sup>23</sup>.

14 In October 2021, Maine DEP began testing PFAS concentrations in all 1,314 fields that receive Class B  
15 sludge, using a tiered system to prioritize sampling. Sites were categorized in three tiers based on the volume of  
16 sludge applied, the presence of homes within ½ mile of the application field and the likelihood that PFAS were  
17 present in the land-applied sludge based on the source of the wastewater solids; ultimately 33 communities were  
18 prioritized for testing<sup>50</sup>. In January 2023 Maine DEP released preliminary results on soil testing of most Tier I  
19 and some Tier II sites<sup>51</sup>. In the findings reported, PFAS soil concentrations in the fields that received sludge  
20 applications were generally greater than background concentrations while greater than 75% of the samples had  
21 detectable PFOS and PFOA and over 50% had ten PFAS at detectable concentrations. Further, 58% of the PFOS  
22 soil concentrations and 36% of the PFOA soil concentrations exceeded Maine's Soil Leaching to Groundwater  
23 remediation criteria (3.6 ng/g and 1.7 ng/g, respectively) prompting groundwater monitoring in the area. In the  
24 samples analyzed to date, the three PFAS found at the greatest concentrations were PFOS (mean 53.89 ng/g,  
25 maximum 1,080 ng/g), 8:2 FTS (8:2 Fluorotelomer sulfonic acid; mean 68.5 ng/g, maximum 2,180 ng/g), and N-

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1 EtFOSSA (N-ethyl perfluorooctane sulfonamido acetic acid; mean 77.3 ng/g, maximum 1,510 ng/g). Low soil  
 2 concentrations of PFBS were reported (meaning approximately 1 ng/g, maximum 10 ng/g).

3 In concert with soil testing in fields that had received Class B sludge, the state mandated that all sludge  
 4 from WWTPs with an active permit to land-apply their Class B sludge be tested for PFAS. Between 2019 and  
 5 2022, one to four grab samples of sludge from each qualifying WWTP were analyzed for a minimum of three  
 6 PFAS (PFOS, PFOA, and PFBS; Table 1). Statewide, sludge from over 90% of the WWTPs exceeded acceptable  
 7 PFAS concentrations and were prohibited from land application. Table 1 summarizes the 2019 - 2022 PFAS  
 8 concentrations from WWTPs that delivered sludge to Juniper Ridge Landfill in 2022. In that subset, PFOS was  
 9 detected in 99% of the 115 sludge samples analyzed, PFOA was detected in 78% of the samples, and 17% had  
 10 detectable PFBS. No data are available on the historical PFAS concentrations for sludge during the preceding 40  
 11 years of land application. While industrial WWTPs had been permitted to spread sludge when Maine began land  
 12 application in the late 1970s, by 2018 no industrial permits remained active and information on PFAS  
 13 concentrations in industrial WWTP sludge was not mandated by the state when sludge testing began in 2019.

**Table 1.** Concentrations of PFAS in wastewater sludge (2019-2022) for 41 of the 45 POTWs that delivered wastewater sludge to Juniper Ridge Landfill in 2022. The majority of sludge tested exceeded acceptable concentrations for the land application of sludge.

Year	2019	2020	2021	2022
Landfilled sludge (US wet tons)	82,888	82,407	90,070	94,270
Sludge samples analyzed	32	41	27	12
<b>PFOS + PFOA</b>				
n detected	32	41	27	11
mean ± SD (ng/g dw)	25 ± 20	23 ± 20	26 ± 21	34 ± 33
min-max (ng/g dw)	5 - 83	3 - 74	2 - 73	1 - 105
<b>PFOS</b>				
n detected	32	41	27	11
mean ± SD	21 ± 17	16 ± 14	20 ± 16	29 ± 26
min-max	5 - 77	3 - 52	2 - 55	1 - 66
<b>PFOA</b>				
n detected	23	34	25	8

mean $\pm$ SD	6 $\pm$ 5	8 $\pm$ 12	6 $\pm$ 7	8 $\pm$ 13
min-max	1 - 21	1 - 63	1 - 25	1 - 40
<b>PFBS</b>				
n detected	6	6	6	2
mean $\pm$ SD	6 $\pm$ 11	2 $\pm$ 2	3 $\pm$ 1	3 $\pm$ 1
min-max	1 - 29	0.5 - 5	2 - 5	2 - 4

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In Maine, the gradual reduction in land application of sludge after 2000 declined further after PFAS testing of WWTP sludge began in 2019. Given the persistence of PFAS in the soil, repeated applications of PFAS-contaminated sludge would lead to additive accumulations of PFAS in the receiving fields. In response to agency insight and strong public pressure, the Maine Legislature banned all land application of wastewater sludge effective August 2022<sup>52</sup>.

#### b. *Landfill Disposal of Wastewater Sludge*

The August 2022 ban on land application of wastewater sludge diverted all sludge generated in the state to landfills, with most sent to the state-owned Juniper Ridge Landfill located north of Old Town in central Maine, operated by Casella Waste Management. Modern landfills are designed to contain materials that can't be reused or recycled and avoid emissions to the air and water. They are constructed with a nearly impermeable liner at the bottom and a cap at the top to prevent water from passing into the waste and from the bottom of the landfill. And there is a gas collection system with flaring to avoid uncontrolled air emissions. Unfortunately, with time and wear, the liners may become more permeable, possibly releasing contaminants, such as PFAS, to the groundwater and air<sup>53</sup>.

In 2022 Juniper Ridge received 94,272 tons of wastewater sludge, a 14% increase in sludge received in 2019 (82,888 tons; Table 1). PFAS concentrations were reported by the state for 41 of the 45 municipal WWTPs that delivered sludge to Juniper Ridge in 2022. Between 2019 and 2022, the average range of sum PFOS + PFOA in sludge was between 23 and 34 ng/g dw, with maximum concentrations between 73 and 105 ng/g dw<sup>22</sup>. Roughly half of the WWTPs analyzed for PFAS three or more times over that four-year period. Sludge concentrations of



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1 sum PFOS + PFOA from individual WWTPs varied by up to 82 ng/g dw between annual grab samplings; no  
 2 seasonal or annual trend in sum PFOS + PFOA was evident.

3 Landfill disposal requires ongoing monitoring and maintenance to prevent release of waste products.  
 4 Juniper Ridge Landfill was chosen as the primary disposal site in Maine as it has a state-of-the-art double  
 5 geomembrane liner and collection system for leachate, water that has filtered through the landfill cells from either  
 6 precipitation or decomposition. In 2022 Juniper Ridge Landfill generated 71.6 M liters of leachate, a 24% increase  
 7 over the 57.7 M liters of leachate produced in 2021 (the landfill had both increased capacity and accepted  
 8 additional sludge deliveries). This leachate was sent to an industrial WWTP (ND Paper LTD, Old Town, Maine)  
 9 for treatment. The concentration of the sum of 6 PFAS in the leachate, analyzed three times between December  
 10 2021 and October 2022, increased from 410 ng/L to 2,441 ng/L (Table 2). The relative composition of the six  
 11 PFAS reported were fairly constant in the leachate despite the six-fold increase in the sum of 6 PFAS  
 12 concentration. PFOA was the dominant PFAS in the leachate (44 – 48%), followed by PFHpA (27 - 30%), PFHxS  
 13 (13 – 18%) and PFOS (4-7%).

14 The ratio of PFOA:PFOS was notably greater in the landfill leachate (6.2 - 12.7; Table 2) relative to the  
 15 ratio of PFOA:PFOS in the incoming sludge (0.3 - 0.5; Table 1), reflecting the greater water solubility of  
 16 PFOA, having a carboxylate head group and one fewer fluorinated carbon in the tail, in the leachate. The PFAS  
 17 concentrations in the wastewater sludge are based on data from the 41 municipal WWTPs for which PFAS  
 18 concentrations are available, which together comprise 83% of the sludge received at Juniper Ridge in 2022.  
 19 Further, 98% of the industrial sludge landfilled at Juniper Ridge comes from ND PP LTD, a pulp mill with an  
 20 industrial WWTP, which also treats the leachate from the Juniper Ridge landfill.

21  
 22 **Table 2.** Sum of 6 PFAS concentrations (ng/L) in landfill leachate from Juniper Ridge Landfill in Old Town,  
 23 Maine increased sixfold between 2021 and 2022.

Leachate Sample Dates	7-Dec-21	26-May-22	4-Oct-22
PFAS sum of 6 (ng/L)	410	1,944	2,441
PFOA	182	914	1,170



PFOS	29	72	162
PFHpA	110	578	691
PFHxS	73	343	321
PFNA	11	37	72
PFDA	5	ND	25

1 Current liner systems have been robust enough if designed properly, but they are not proven to hold  
2 PFAS-laden leachate in the system for decades. Since PFAS are surface active molecules, the presence of PFAS  
3 in the leachate may increase the permeability through faults or pinholes in geomembranes. Long-term release of  
4 PFAS through liners into the groundwater requires monitoring long after the solid waste facility is closed.  
5 Lastly, after the lifetime of the “PFAS-waste landfill”, the site may still need preventative (e.g., monitoring)  
6 action or remediation<sup>54</sup>.

### c. PFAS thermal treatment

7 Thermal treatments, such as incineration (with air) and pyrolysis (lacking oxygen), are commonly used  
8 means of mineralizing organic materials, especially if they contain recalcitrant components, to H<sub>2</sub>O, CO<sub>2</sub>,  
9 oxidized sulfur and nitrogen compounds, and mineral acids. Incineration is commonly applied to municipal solid  
10 waste and biosolids, and can theoretically mineralize PFAS when they are held at over 1000°C for at least two  
11 seconds<sup>55</sup>. While other halogenated organic chemicals, such as chlorinated organics, can be destroyed by  
12 incineration at 900°C the strong C–F bond and reactivity of fluorine<sup>55,56</sup> mean that fluorinated organic compounds  
13 require temperatures up to 1400°C to achieve complete thermal mineralization<sup>57,58</sup>. Furthermore, in dewatered  
14 wastewater sludge or granular activated carbon (GAC) the solids may inhibit volatilization of the PFAS at  
15 predicted boiling temperatures, and sorption to solids may influence pathways of degradation<sup>59</sup>.

16 While complete mineralization of PFAS requires temperatures over 1000°C, their decomposition begins  
17 at much lower temperatures. Depending on the compound type, reaction conditions (temperature, time, mixing  
18 and oxygen) and background material (pH, surfaces, presence of catalysts), product formation varies widely  
19 <sup>55,58,59</sup>. Decomposition of the parent compound may begin at temperatures as low as 200-300°C (for PFCAs) or  
20 450-500°C (for PFSAs and fluoropolymers), and generate a variety of fluorinated byproducts<sup>55,56,58,60</sup>. The  
21 functional group of PFAS is usually the least stable part of the molecule and can be detached or eliminated at  
22

1 low/initiation temperatures. The fluorinated backbone may then enter a variety of reaction pathways generating  
2 smaller, more stable byproducts of unknown composition, making it difficult to determine mineralization  
3 efficiencies and identify byproducts<sup>61,62</sup>. In fact, most experiments on PFAS thermal decomposition fail to obtain  
4 a complete mass balance indicating the generation of unknown fluorinated byproducts<sup>58</sup>. Some of these products  
5 released into the gas phase may be harmful to the environment, such as fluorinated greenhouse gases like  
6 tetrafluoromethane, hexafluoroethane, and ozone-depleting chlorofluorocarbons<sup>56,63,64</sup>.

7 Some recalcitrant PFAS or their combustion by-products have been detected in the solid residuals of  
8 pyrolysis and incineration. While PFAS concentrations in the ash were lower than in the input sludge, sometimes  
9 by an order of magnitude<sup>37,65</sup>, their presence in the residual indicates that these treatments resulted in incomplete  
10 combustion. Further study is needed to identify end products and decomposition pathways of thermal treatment  
11 under realistic conditions<sup>58,63</sup>. These and other uncertainties have led to bans on incineration of concentrated  
12 PFAS waste streams in hazardous waste incinerators by the Department of Defense<sup>66</sup> and the State of Illinois,  
13 although the Illinois ban does not apply to wastewater sludge<sup>67</sup>.

### 14 **C. Future alternatives for managing wastewater sludge**

15 The ubiquitous presence of PFAS in wastewater sludge, and the likely occurrence of other toxic chemicals  
16 yet to be identified, precludes the continued land application of sludge. Incineration requires further investigation  
17 of unidentified thermolysis byproducts, short-chain PFAS, or air-borne PFAS-laden particulate matter that can be  
18 released and may be subject to long-range transport. Landfill disposal creates a potential point source of PFAS if  
19 leachate escapes to groundwater or uncontrolled air emissions release volatile PFAS. Even with a functioning  
20 leachate collection system, the PFAS in the landfill leachate will cycle through a WWTP, to again be partitioned  
21 between the effluent and the sludge, with the sludge returning to the landfill. To solve this “wicked problem”, the  
22 future alternatives must be truly innovative and should realize these PFAS-specific challenges. Importantly,  
23 innovative solutions come at a cost, that will be primarily funded by taxpayers. Maine survey respondents were  
24

1 given the opportunity to ‘vote’ in favor or opposed to incurring either a hypothetical property tax increase or sales  
2 tax increase on themselves with the resulting funds being allocated to PFAS management in the state. Although  
3 66% voted in favor of public spending on PFAS, priorities for spending were not unanimous nor do they  
4 necessarily align with the needs identified in this manuscript. Interestingly, only 7% of respondents indicated that  
5 the top priority for PFAS funds should be landfills. Funding allocation choices may reflect citizens’ limited  
6 understanding of the current options and future alternatives available to address PFAS, some of which are outlined  
7 below.

### 8 **1. PFAS sequestration**

9 PFAS are especially difficult to eliminate when they are present in extremely low concentrations (ng/L,  
10 parts per trillion, to  $\mu\text{g/L}$ , parts per billion) in complex environmental matrices such as sludge. Removing or  
11 destroying PFAS from a medium as complex as sludge is both technically challenging and expensive. Co-locating  
12 wastes with elevated PFAS concentrations could improve PFAS removal efforts. Landfills that accept a mixture  
13 of solid wastes have detectable levels of PFAS in leachate. Since wastewater sludge is identified as an important  
14 source of PFAS in the landfill leachate<sup>33,63</sup>, it may be useful to isolate the PFAS-laden sludge within certain  
15 landfill cells, and so concentrate the PFAS in the leachate from those cells to enhance PFAS removal from the  
16 leachate before further treatment (and dilution) at a WWTP. The underlying logistical preparations (e.g., legal  
17 framework, techno-economic assessment, research), for such complicated arrangements require time, resources,  
18 and collaboration between policy makers, researchers, and practitioners. Regardless, as persistent, toxic  
19 chemicals, PFAS are best removed from the leachate using one of the existing separation techniques, such as  
20 sorption to granular activated carbon<sup>68,69</sup> or ion exchange resin<sup>70</sup>, or foam fractionation<sup>71,72,73</sup> before dilution at  
21 the WWTP, which does not have the capacity to remove and destroy this toxic chemical.

#### 22 **a. Immobilization**

23 One incipient concept developed by our team is to manage highly concentrated PFAS-laden waste by  
24 immobilizing it until PFAS destruction technologies mature enough for field operation. The immobilization is at  
25 its conceptualization stage with no preliminary data but in principle involves a two-step process, first extraction

1 of PFAS in the sludge into a non-polar solvent (e.g., hexadecane) to minimize partitioning into water, followed  
2 by immobilization of the slurry in a robust solid matrix (e.g., cement).

3 While the concept is promising, fundamental research is needed to prove the safety and sustainability of  
4 this immobilization approach. The pre-conditioning and immobilization of PFAS-laden sludge could be an  
5 interim solution to break the cycle of PFAS moving from a WWTP via sludge to a landfill, then carried back in  
6 leachate to the WWTP and provide time to develop reliable PFAS destruction technology options.

7 In the long run, if PFAS-only solid waste facilities or PFAS-immobilization for long term storage are  
8 realized and operated, maintaining the conservation of PFAS within the facility could become a potential problem  
9 because of the large quantities of waste that needs to be processed.

## 10 **2. PFAS destruction technologies**

11 Direct destructive technologies for PFAS-laden sludge are at early-stages of development, and to date  
12 only a small number of studies have been conducted. Yet these destructive technologies offer a range of technical  
13 and economic options. These technologies include activated persulfate oxidation<sup>74</sup>, UV-sulfite reduction<sup>75</sup>, low  
14 temperature mineralization in polar, non-protic solvents<sup>76</sup>, ultrasonication<sup>77</sup>, plasma<sup>78</sup>, ball milling<sup>79</sup>, electron  
15 beams, advanced electrochemical oxidation, or hydrothermal liquification (HTL). In general, the technologies  
16 that are demonstrated in water will need significant adaptations to be applicable in slurry reactors. However, some  
17 of these destructive technologies must still be adapted to treat sludge given that initial testing of these methods  
18 was primarily in relatively simple aquatic matrices. We are going to provide three examples of early-stage  
19 technologies that are shown to degrade PFAS in laboratory conditions. These technologies rely on different  
20 physicochemical principles and disparate engineering considerations in order to present a variety of discussion  
21 points. Clearly, for almost all destructive technologies proposed in the literature more research is needed before  
22 real-life applications will be possible.

### 23 a. *Electron beam irradiation*

1            Electron beam irradiation, a promising solid-phase destructive technology, utilizes high-energy electrons  
2 to decompose PFAS-laden sludge. In this process, high-energy electrons are generated by a heated filament and  
3 accelerated through a vacuum accelerator tube by a high-voltage electric field between the cathode and anode<sup>80</sup>.  
4 These electrons are then focused by magnetic and electric lenses to attack water molecules, resulting in the  
5 production of reactive agents that decompose pollutants such as PFAS, pharmaceuticals, and chlorinated  
6 chemicals<sup>81</sup>. The technology could be costly for sludge based on the accelerator capacity, applied dose, and sludge  
7 quantity, water content and PFAS concentration. However, further research is required to answer these questions.  
8 Electron beams are demonstrated in dilute aqueous solution so far. For example, Ma et al. (2017)<sup>81</sup> demonstrated  
9 the effective mineralization of PFOA and PFOS using a 500 kGy electron beam in an anoxic alkaline solution,  
10 with degradation rates of 95.7% and 85.9%, respectively. The size, associated costs, and complexity of the system  
11 are among the obstacles to the widespread use of this technology<sup>82</sup>. Additionally, the availability of infrastructure,  
12 such as an adequate power supply and skilled personnel, would be more available in a central location rather than  
13 in rural regions such as those found in many parts of Maine. Information on the energy and time required to  
14 process PFAS-contaminated sludge using electron beam irradiation is not available, and further research is  
15 necessary to optimize its performance and overcome the limitations to fully realize its potential.

#### 16            b. *Electrochemical advanced oxidation*

17            Second, electrochemical advanced oxidation is a method that employs electricity to generate oxidative  
18 species at the electrode to interact with contaminants such as PFAS, ultimately leading to their breakdown<sup>81</sup>. This  
19 approach surpasses conventional advanced oxidation techniques, as it combines the oxidative power of hydroxyl  
20 radicals generated through anodic water oxidation with the capability of direct electron transfer onto the anode<sup>84</sup>.  
21 The main advantages of electrochemical advanced oxidation include low energy consumption and the absence of  
22 chemical usage. Additionally, this method can be applied at ambient temperature and atmospheric pressure  
23 without requiring external reagents<sup>85</sup>. In a study by Hou et al. (2022)<sup>84</sup>, low-intensity electrochemical treatment,  
24 24 V, using iridium pentoxide plating on titanium anode/cathodes was shown to be effective in remediating PFOA  
25 and PFOS in soil over a period of 10 days. The researchers observed that PFOA was more easily remediated than

1 PFOS<sup>86</sup>, which may be attributed to the sulfonate group being more electrophilic than the carboxyl group of  
2 PFOA, and therefore having less affinity for losing electrons at the anode surface<sup>87</sup>. Despite these advantages,  
3 some challenges to the commercialization of electrochemical processes remain. For example, low current  
4 efficiencies in terms of quantities of removed contaminant per unit volume and unit time can be a limitation<sup>88</sup>. To  
5 improve the energy efficiency, current modulation i.e., lowering the currents applied to the limiting current for  
6 PFAS oxidation could be employed. Additionally, the inefficiency of the method in completely degrading short  
7 chain PFAS is a concern which requires additional research. Wang et al. (2020)<sup>89</sup> have found that short-chain  
8 PFAS are generally more resistant to electrochemical treatment compared to long-chain PFAS. They investigated  
9 a solution containing a mixture of 8 PFAS, namely PFBA, PFPeA, PFHpA, PFOA, PFBS, PFHxS, PFOS, with  
10 an initial concentration of 2  $\mu\text{M}$ , and found that after 8 hours of treatment, only 50% of PFBA was degraded,  
11 while the remaining PFAS were degraded over 90%. Finally, electrochemical oxidation can generate by-products  
12 as a function of electrode material such as  $\text{Cl}_2$ , HF,  $\text{ClO}_3$ ,  $\text{ClO}_4$ , and  $\text{BrO}_3$ <sup>90</sup>. The energy consumption of  
13 electrochemical oxidation depends on the type of electrode and supporting electrolyte used, the amount of current  
14 applied, and the size of the system. Therefore, consideration of these factors is required when evaluating the  
15 energy use and associated costs of electrochemical oxidation for PFAS-contaminated sludge treatment.

### 16 c. *Hydrothermal liquefaction*

17 Hydrothermal liquefaction, which principally is a waste-to-energy process that offers a possible  
18 destructive approach to the transformation of aqueous PFAS-laden slurries (e.g., 15 wt.% solids sludge slurry)  
19 into valuable end products<sup>91</sup>. The process could destroy PFAS because it involves subjecting sludge to elevated  
20 pressures ranging from 10 to 25 MPa and temperatures ranging from 250 to 350°C to produce a liquid biocrude  
21 oil. Hot compressed water, which is highly reactive near its critical point at 374°C and 22.0 MPa, is a key  
22 component of the process<sup>92</sup>. The advantages of HTL include production of valuable products such as biochar,  
23 biocrude oil, and aqueous products containing fertilizer precursors while potentially destroying PFAS if the  
24 process is tuned appropriately<sup>91</sup>. Demonstrations and pilot-scale experiments of HTL are currently being tested  
25 at municipal wastewater treatment facilities<sup>92,93</sup>. To make the technology operational, the effect of PFAS must be



1 evaluated because they are surface-active compounds and may partition to the oil-water interphase and change  
2 the yield of the HTL process. Also, the generation of PFAS-transformation products and their fate must be taken  
3 into consideration<sup>94</sup>. Furthermore, the high pressures and temperatures required for HTL may pose challenges  
4 regarding equipment durability, maintenance, and cost-effectiveness. The energy or time required to process  
5 PFAS-contaminated sludge using HTL is dependent on the characteristics of the sludge (e.g., water content), and  
6 HTL conditions. Further research and development are necessary to optimize the HTL process and overcome its  
7 limitations, making it a viable PFAS destruction technology for sustainable sludge management.

### 8 9 10 11 12 **III. SUMMARY AND CONCLUSIONS**

13 PFAS toxicity was not publicly recognized until the 1990s, fifty years after commercial production began  
14 and despite internal toxicity findings at 3M and DuPont that were not publicly released, when EPA received court  
15 documents from the C-8 study of drinking water contamination from the DuPont manufacturing plant in West  
16 Virginia<sup>9</sup>. Today the accumulation of PFAS seems self-perpetuating given the circular movement of these  
17 persistent chemicals from industry and manufacturing, through consumer products, into our waste streams, and  
18 the broader environment. It is likely that contamination of the food system through sludge application to  
19 agricultural soils has been replicated elsewhere; other states and nations will need to make investments to ensure  
20 that safe, sustainable, and effective PFAS recovery and destruction options are available. Maine, and a few other  
21 states, are proactively addressing the PFAS crisis through monitoring to quantify the extent of the contamination,  
22 research into methods to remove the chemicals from our wastestreams, fields and waters, and legislation to  
23 eliminate ongoing sources of PFAS contamination. In the future, conditions for effective thermal destruction or  
24 one of the early stage technologies for destroying PFAS in wastewater sludge, electron beam irradiation,  
25 electrochemical advanced oxidation, or hydrothermal liquefaction, may mature and become applicable at field-  
scale, but it is hard to predict which will fit into the environmental engineering ecosystem. The proposed PFAS  
management solutions must be in tune with our current practices in addition to being safe, economically feasible,



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1 politically acceptable, and sustainable. Indeed we recognize that acceptance of management solutions may help  
2 or hinder progress, where funding allocation may reflect citizen's perceptions of who is at fault for PFAS  
3 contamination, as well as indicating who is a trusted source of both information and mitigation of actions. A  
4 further compounding factor is limited information on the economic costs associated with various mitigation  
5 actions<sup>95</sup>, as well as the costs of inaction<sup>96</sup>.

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## 16 **AUTHOR DECLARATIONS**

- 17 II. *Conflict of Interest* – The authors have no conflicts to disclose.
- 18 III. *Ethics Approval* – Ethics approval was obtained through the University of Maine Institutional Review  
19 Board for the Protection of Human Subjects, IRB ID # 2021\_10\_16. Human participants gave informed  
20 consent before participating in the survey.

## 22 **DATA AVAILABILITY**



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1           The data that support these findings are available from State of Maine websites cited in the reference  
2 section, or from the corresponding author upon reasonable request, recognizing that some data are not publicly  
3 available due to privacy restrictions.  
4

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