

PFAS drinking water treatment trade-offs: comparing the health burden of GAC treatment to the health benefits of reduced PFAS exposure

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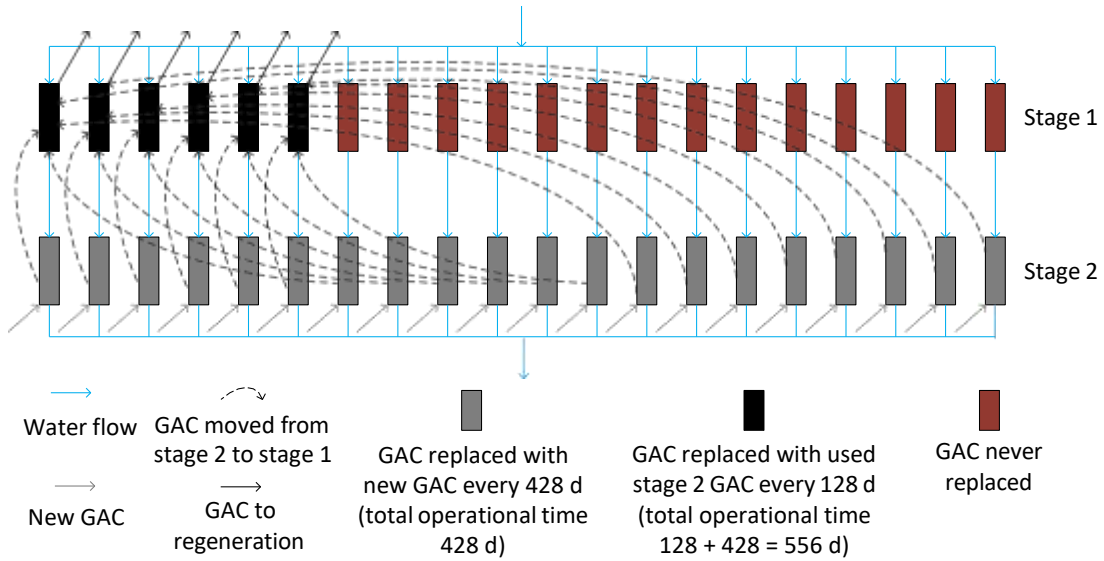
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*SI Table 1: Full names and relative potency factors (RPF) of PFAS included in the calculation of PFOA-equivalent (PEQ) concentrations.^{1,2} Compounds indicated with an * are included in the EFSA-4 tolerable weekly intake.*

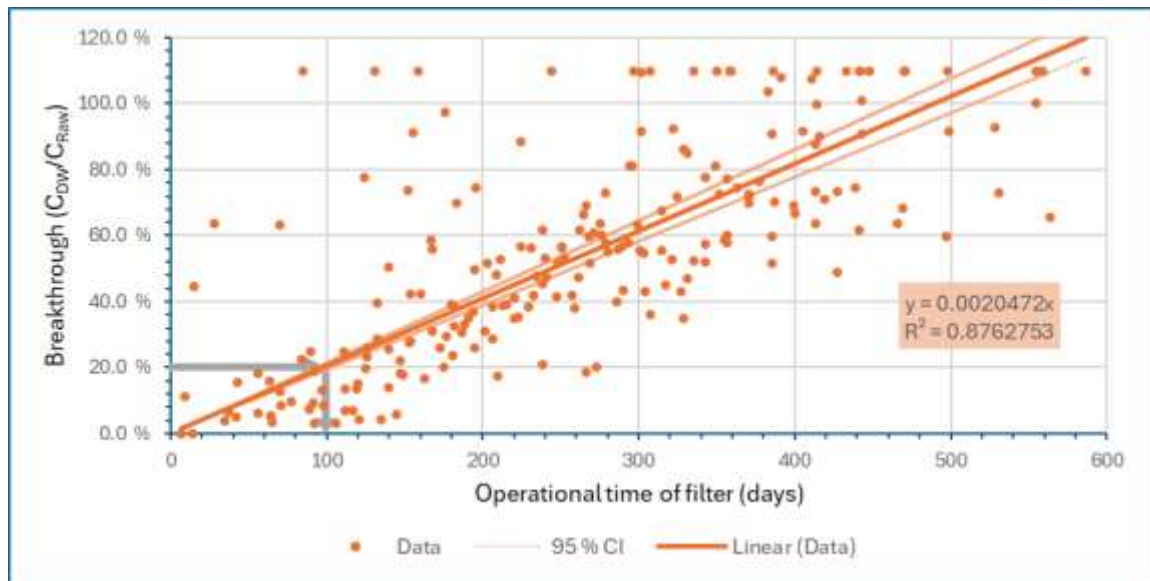
| Abbreviation | Full name | Relative potency factor (RPF) |
|--------------|--------------------------------------|-------------------------------|
| PFOA* | Perfluorooctanoic acid | 1 |
| PFOS* | Perfluorooctane sulfonic acid | 2 |
| PFNA* | Perfluorononanoic acid | 10 |
| PFHxS* | Perfluorohexane sulfonic acid | 0.6 |
| PFBA | Perfluorobutanoic acid | 0.05 |
| PFPeA | Perfluoropentanoic acid | 0.05 |
| PFHxA | Perfluorohexanoic acid | 0.01 |
| PFHpA | Perfluoroheptanoic acid | 1 |
| PFDA | Perfluorodecanoic acid | 10 |
| PFUnDA | Perfluoroundecanoic acid | 4 |
| PFDoDA | Perfluorododecanoic acid | 4 |
| PFTTrDA | Perfluorotridecanoic acid | 3 |
| PFTeDA | Perfluorotetradecanoic acid | 0.3 |
| PFHxDA | Perfluorohexadecanoic acid | 0.02 |
| PFODA | Perfluorooctadecanoic acid | 0.02 |
| PFBS | Perfluorobutane sulfonic acid | 0.001 |
| PFPeS | Perfluoropentane sulfonic acid | 0.6 |
| PFHpS | Perfluoroheptane sulfonic acid | 2 |
| PFDS | Perfluorodecane sulfonic acid | 2 |
| HFPO-DA | Hexafluoropropylene oxide dimer acid | 0.06 |
| DONA | 4,8-Dioxa-3H-perfluorononanoic acid | 0.03 |
| 6:2 FTOH | 6:2 fluorotelomer alcohol | 0.02 |
| 8:2 FTOH | 8:2 fluorotelomer alcohol | 0.04 |

Concentrations in PEQ are calculated by summing the concentrations of individual PFAS multiplied with their RPF. For example, a water sample that contains 1 ng/L of each of the EFSA₄ compounds will have a PEQ concentration of 1·1 (PFOA) + 1·2 (PFOS) + 1·10 (PFNA) + 1·0.6 (PFHxS) = 13.6 ng PEQ/L.

Supplementary Information



SI Figure 1: Overview of GAC treatment process at the Leiduín drinking water treatment site from Waternet. Operational times shown are calculated based on the average reactivation frequency in 2024.



SI Figure 2: Breakthrough of PEQ, from raw water (C_{RAW}) to drinking water (C_{DW}) versus operational time of GAC filters, fit to a linear regression model. Note that the operational time used to determine the required reactivation frequency was twice the operational time found from the fitted equation: since multiple GAC filters are operated in parallel, this ensures that the average operational time over all filters is equal to the determined maximum.

Supplementary Information

SI Table 2: Inventory for Water Treatment Process (GAC focus) - Single Use Coal and Wood GAC

| Inputs | Amounts | Unit | Comments |
|------------------------|--|----------------|---|
| GAC: | (a) 931; (b) 2002; (c) 1774; (d) 2298 | | <u>Coal-based GAC</u> : Ecoinvent dataset 'Activated carbon, granular {RER} activated carbon production, granular from hard coal Cut-off, U' <u>Wood-based GAC</u> : Project data, as in SI table 6. |
| Tap Water | 70726271 | m ³ | Tap water {Europe without Switzerland} market for Cut-off, U |
| Electricity | 3823756 | kWh | Electricity, medium voltage {NL} market for Cut-off, U. Estimation of WTP use data for GAC treatment. |
| Transport of Wood-GAC | (a) 32585; (b) 70070; (c) 62074; (d) 80430 | tkm | Transport, freight, lorry 16-32 metric ton, euro6 {RER} market for transport, freight, lorry 16-32 metric ton, EURO6 Cut-off, U. Distance of transport from GAC Activation to WTP – 35km. |
| Transport of Spent GAC | (a) 23250; (b) 50050; (c) 44350; (d) 57450 | tkm | Transport, freight, lorry 16-32 metric ton, euro6 {RER} market for transport, freight, lorry 16-32 metric ton, EURO6 Cut-off, U. Spent GAC to Incineration plant - 20 km. |
| Output | | | |
| Drinking water | 70653937 | m ³ | Project defined, WTP data |
| Waste | | | |
| Spent WoodGAC, wet | (a) 1163.75; (b) 2502.5; (c) 2217; (d) 2873 | ton | Hazardous waste, for incineration {Europe without Switzerland} market for hazardous waste, for incineration Cut-off, U, as in Ellis et al. 2023. Spent GAC is assumed to contain 25% moisture as in Vilen, 2022. |

- (a) 2024 GAC requirements
- (b) average estimated GAC requirements
- (c) minimum estimated GAC requirements
- (d) maximum estimated GAC requirements

Supplementary Information

SI Table 3: Inventory for Water Treatment Process (GAC focus) - Reactivated Coal and Wood GAC .

| Inputs | Amounts | Unit | Comments |
|-----------------|--|----------------|---|
| Virgin GAC | (a) 116.4 (b) 250.3 (c) 221.7 (d) 287.3 | ton | <u>Coal-based GAC</u> : Ecoinvent dataset 'Activated carbon, granular {RER} activated carbon production, granular from hard coal Cut-off, U' <u>Wood-based GAC</u> : Project defined, as in table SI 6. |
| Reactivated GAC | (a) 814.6; (b) 1751.8; (c) 1551.9; (d) 2010.8 | ton | <u>Coal-based GAC</u> : Project defined, based on ecoinvent dataset 'Activated carbon, granular {RER} treatment of spent activated carbon, granular from hard coal, reactivation Cut-off, U' <u>Wood-based GAC</u> : Project defined, as in SI 7. |
| Tap Water | 70726271 | m ³ | Tap water {Europe without Switzerland} market for Cut-off, U |
| Electricity | 3823756 | kWh | Electricity, medium voltage {NL} market for Cut-off, U |
| Transport | (a) 32585; (b) 70074; (c) 62076; (d) 80434 | tkm | Sum of transport need for virgin and reactivated GAC. Transport, freight, lorry 16-32 metric ton, euro6 {RER} market for transport, freight, lorry 16-32 metric ton, EURO6 Cut-off, U. Distance of transport from GAC (re)activation to WTP – 35km. |
| Outputs | Amounts | Unit | Database process |
| Drinking water | 70653937 | m ³ | Project defined |
| Spent GAC | (a) 1164; (b) 2503 (c) 2217; (d) 2873; | ton | Accounted as wet GAC, 25% moisture. <u>Coal-based GAC</u> : 'Activated carbon, granular {RER} treatment of spent activated carbon, granular from hard coal, reactivation Cut-off, U' <u>Wood-based GAC</u> : Project defined, as input for reactivation, as in SI 7, |

(a) 2024 GAC requirements

(b) average estimated GAC requirements

(c) maximum estimated GAC requirements

(d) minimum estimated GAC requirements

Supplementary Information

SI Table 4: Inventory for Wood GAC Production Process

| Inputs | Amounts | Unit | Comments |
|----------------------|-------------|------|--|
| Wood chips | 8,44 | kg | Bark chips, wet, measured as dry mass {GLO} market for Cut-off, U. |
| Electricity | 1,7 | kWh | Medium voltage, NL. Amount based on Vilen, 2022. |
| Nitrogen | 0,15 | kg | Nitrogen, liquid {RER} market for Cut-off, U. Amount based on Vilen, 2022 ³¹ |
| Tap water | 2,11 | kg | Tap water {Europe without Switzerland} market for Cut-off, S. Amount based on Vilen, 2022. ³¹ |
| Transport | | | |
| Truck | 0,422 | tkm | Transport, freight, lorry 16-32 metric ton, euro6 {RER} market for transport, freight, lorry 16-32 metric ton, EURO6 Cut-off, U. Transport of wood chips to GAC production only, with an assumed distance of 50km, as in Vilen, 2022. ³¹ |
| Outputs | | | |
| Wood-based GAC | 1 | kg | Project defined stream. |
| Emissions | | | |
| Activation | | | |
| CO2 | 1,81E+00 | kg | Based on Gu et al, 2018 ³² (originally per kg biochar feedstock, converted based on 2,11 kg wood biochar / kg wood-based GAC). |
| H2O | 9,07E-02 | kg | |
| N2 | 1,83E+00 | kg | |
| O2 | 1,59E+00 | kg | |
| H2 | 4,22E-03 | kg | |
| CO | 8,86E-03 | kg | |
| CH4 | 8,44E-04 | kg | |
| SO2 | 1,27E-03 | kg | |
| HCl | 1,39E-06 | kg | |
| Nox | 8,84E-05 | kg | |
| N2O | 1,39E-06 | kg | |
| Acetaldehyde | 1,16E-05 | kg | |
| Benzene | 8,36E-05 | kg | |
| Formaldehyde | 4,64E-08 | kg | |
| Methanol | 4,64E-06 | kg | |
| Naphthalene | 9,31E-06 | kg | |
| Phenol | 2,04E-06 | kg | |
| Propanal | 9,31E-08 | kg | |
| Particulates | 4,62E-02 | kg | |
| Carbonization | | | |
| Acetaldehyde | 9,65029E-06 | kg | Taking emissions to air from ecoinvent dataset "wood pellets, burned in stirling heat and power co-generation unit, 3kW electric", with methane, carbon dioxide and carbon monoxide reduced by 43% as in Vilen, 2022 11 as described in main text. |
| Ammonia | 0,000273688 | kg | |
| Arsenic | 1,58201E-07 | kg | |
| Benzene | 0,000143963 | kg | |
| Benzene, ethyl- | 4,74604E-06 | kg | |
| Benzene, hexachloro- | 1,13905E-12 | kg | |
| Benzo(a)pyrene | 7,91007E-08 | kg | |

Supplementary Information

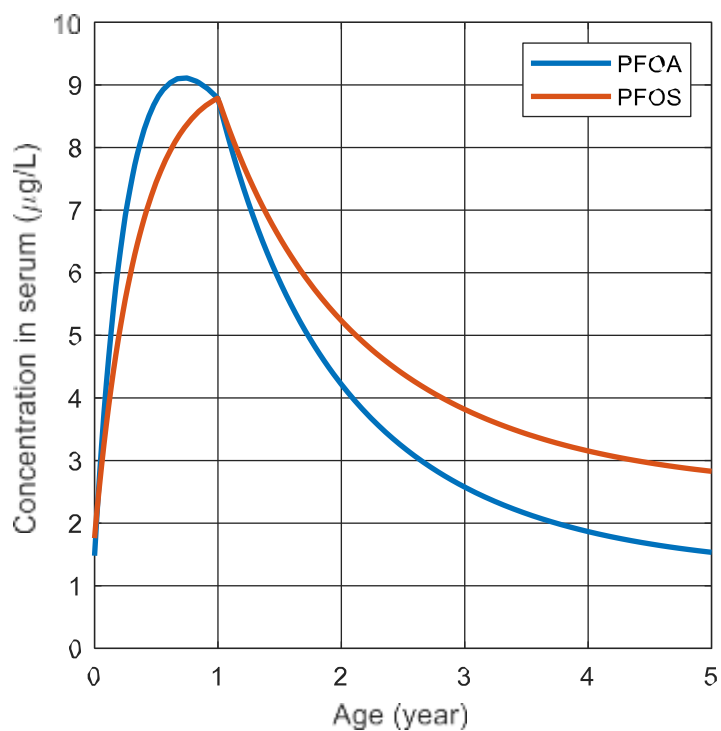
| | | |
|---|-------------|----|
| Bromine | 9,49209E-06 | kg |
| Cadmium | 1,10741E-07 | kg |
| Calcium | 0,000925478 | kg |
| Carbon dioxide, biogenic | 8,711158032 | kg |
| Carbon monoxide, biogenic | 0,002885594 | kg |
| Chlorine | 2,84763E-05 | kg |
| Chromium | 6,26478E-07 | kg |
| Chromium VI | 6,32806E-09 | kg |
| Copper | 3,48043E-06 | kg |
| Dinitrogen monoxide | 0,000474604 | kg |
| Dioxin, 2,3,7,8 Tetrachlorodibenzo-p- | 4,90424E-12 | kg |
| Fluorine | 7,91007E-06 | kg |
| Formaldehyde | 2,05662E-05 | kg |
| Hydrocarbons, aliphatic, alkanes, unspecified | 0,000143963 | kg |
| Hydrocarbons, aliphatic, unsaturated | 0,000490424 | kg |
| Lead | 3,95504E-06 | kg |
| Magnesium | 5,69525E-05 | kg |
| Manganese | 2,68942E-05 | kg |
| Mercury | 4,74604E-08 | kg |
| Methane, biogenic | 3,60699E-05 | kg |
| m-Xylene | 1,89842E-05 | kg |
| Nickel | 9,49209E-07 | kg |
| Nitrogen oxides | 0,011074101 | kg |
| NMVOC, non-methane volatile organic compounds, unspecified origin | 0,000363863 | kg |
| PAH, polycyclic aromatic hydrocarbons | 1,75604E-06 | kg |
| Particulates, < 2.5 um | 0,001582014 | kg |
| Phenol, pentachloro- | 1,28143E-09 | kg |
| Phosphorus | 4,74604E-05 | kg |
| Potassium | 0,003701914 | kg |
| Sodium | 0,000205662 | kg |
| Sulfur dioxide | 0,000395504 | kg |
| Toluene | 4,74604E-05 | kg |
| Zinc | 4,74604E-05 | kg |

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SI Table 5: Inventory for Reactivated Wood GAC Production Process

| Inputs | Amounts | Unit | Comments |
|----------------------|----------------|-------------|--|
| Spent GAC | 1,00 | kg | Wet, as output from drinking water process |
| Electricity | 0,41 | kWh | Medium voltage, NL |
| Nitrogen | 0,04 | kg | Liquid, RER |
| Tap water | 0,55 | kg | Europe without Switzerland |
| <i>Transport</i> | | | |
| Truck | 0,03 | tkm | Transport, freight, lorry 16—32 t, RER. Same transport mode as Wood-GAC. Distance: 30km for return of spent GAC to Norit Activated Carbon for reactivation. |
| | | | |
| Outputs | | | |
| Reactivated Wood GAG | 0,89 | kg | 1 enters, losses are 12,5% on final prod. |
| Emissions | | | |
| CO ₂ | 1,81E+00 | kg | Based on activation of biochar as in Gu et al, 2018 (originally per kg biochar feedstock, converted based on 2,11 kg wood biochar / kg wood-based GAC). Biogenic CO, CO ₂ and CH ₄ . |
| H ₂ O | 9,07E-02 | kg | |
| N ₂ | 1,83E+00 | kg | |
| O ₂ | 1,59E+00 | kg | |
| H ₂ | 4,22E-03 | kg | |
| CO | 8,86E-03 | kg | |
| CH ₄ | 8,44E-04 | kg | |
| SO ₂ | 1,27E-03 | kg | |
| HCl | 1,39E-06 | kg | |
| Nox | 8,84E-05 | kg | |
| N ₂ O | 1,39E-06 | kg | |
| Acetaldehyde | 1,16E-05 | kg | |
| Benzene | 8,36E-05 | kg | |
| Formaldehyde | 4,64E-08 | kg | |
| Methanol | 4,64E-06 | kg | |
| Naphthalene | 9,31E-06 | kg | |
| Phenol | 2,04E-06 | kg | |
| Propanal | 9,31E-08 | kg | |
| Particulates | 4,62E-02 | kg | |

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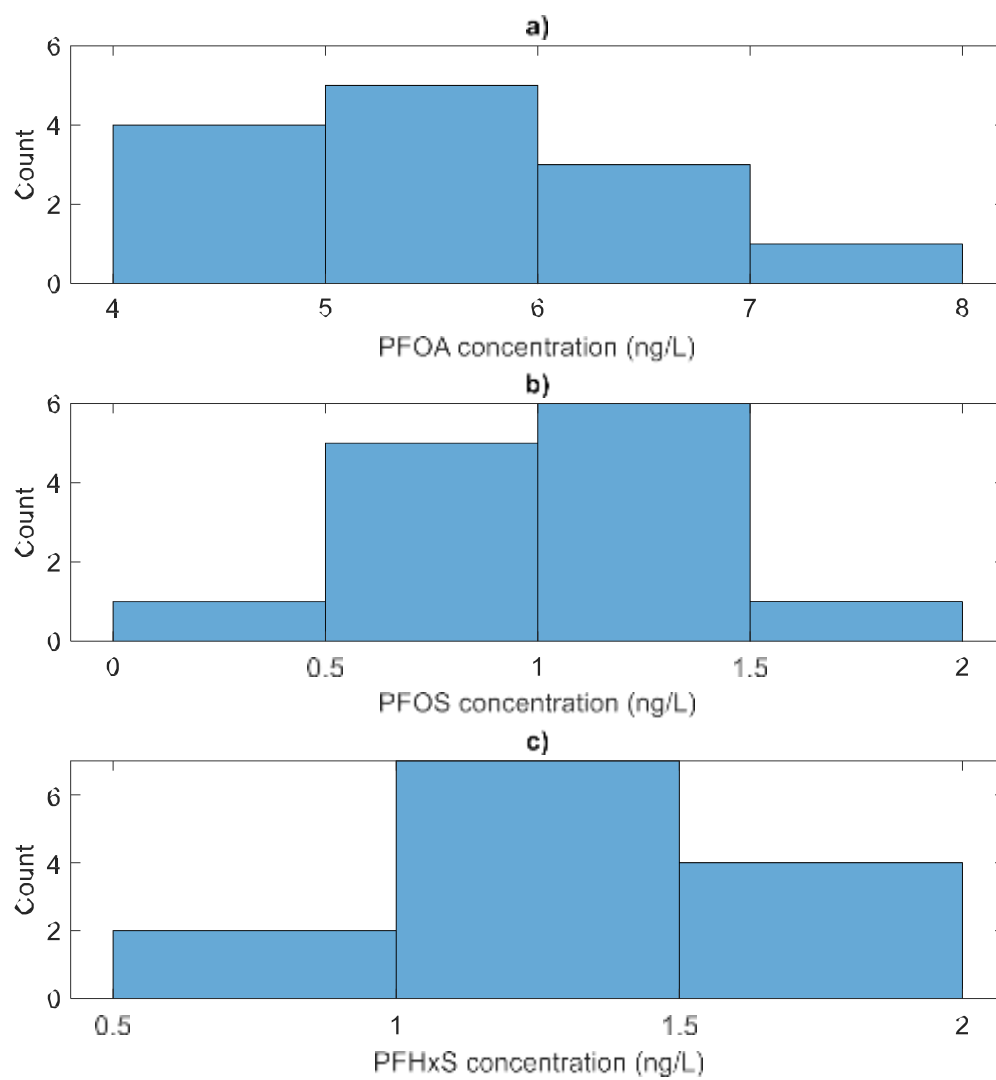


SI Figure 3: Verification that MATLAB PBPK model is an accurate reproduction of EFSA's model: PFOA and PFOS serum concentrations until age 5 using the same parameters as in EFSA's original model, to compare with Figure M.1 (page 379; PFOA) and Figure M.3 (page 382, PFOS) from Schrenk et al., 2020.³

SI Table 6: Overview of data used in DALY calculation

| Endpoint | Population | P (per 10 ⁶ people) ⁴ | W (DALYs/case) | I (cases/person/year) |
|-------------------|---------------|---|---------------------|-----------------------|
| Kidney cancer | Adults >20 | 792,000 | 9.1 ⁵ | 0.000194 ⁶ |
| Testicular cancer | Males >18 | 403,000 | 1.8 ⁷ | 0.000064 ⁶ |
| Hypothyroidism | Females 18-49 | 200,000 | 0.19 ^{8,9} | 0.002 ¹⁰ |
| Hypertension | Adults > 20 | 792,000 | 0.12 ¹¹ | 0.008 ¹⁰ |

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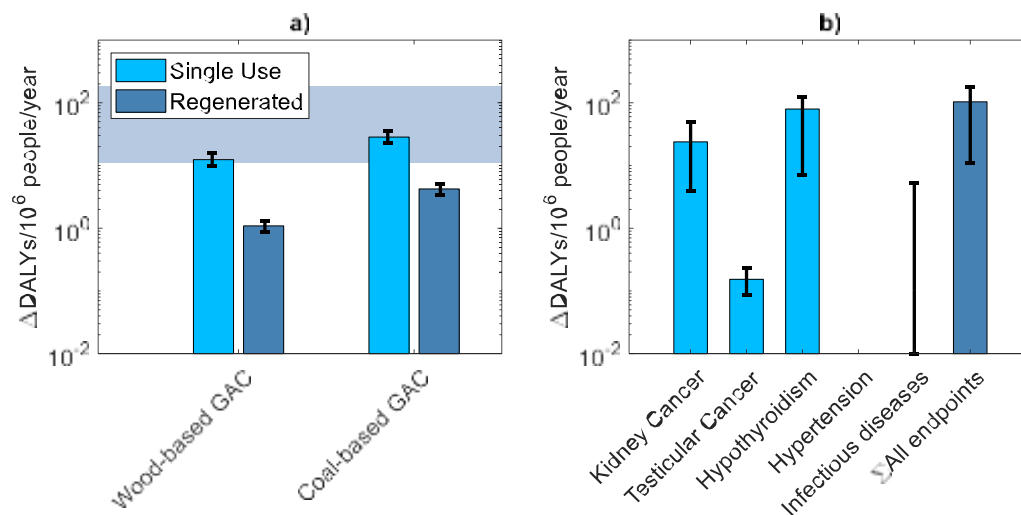


SI Figure 4: Distribution of 2024 drinking water concentrations of a) PFOA, b) PFOS and c) PFHxS. Total $n = 13$ for all.

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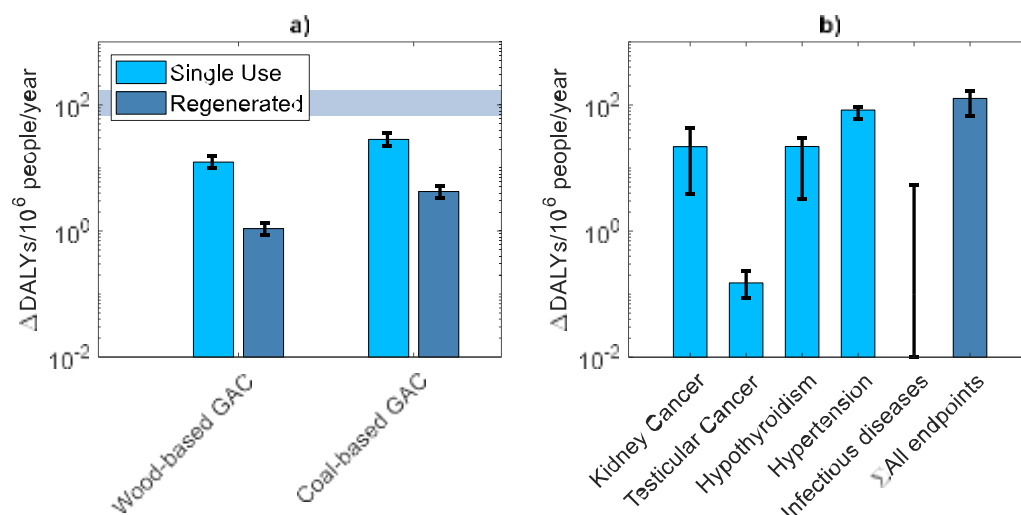
SI Table 7: Health impacts (DALYs/10⁶ p/year) from GAC treatment for each scenario, excluding DALYs related to water consumption. 2024 = reactivation frequency as in 2024 (current); Prosp. = prospective reactivation frequency

| GAC type | Single use wood | | Single use coal | | Reactivated coal | | Reactivated wood | |
|-----------------------------------|-----------------|------------|-----------------|------------|------------------|-----------|------------------|-----------|
| Scenario | 2024 | Prosp. | 2024 | Prosp. | 2024 | Prosp. | 2024 | Prosp. |
| Global warming | 29 | 33 | 34 | 45 | 26 | 28 | 25 | 26 |
| Fine particulate matter formation | 31 | 38 | 37 | 50 | 29 | 31 | 28 | 29 |
| Human carcinogenic toxicity | 29 | 30 | 30 | 32 | 28 | 28 | 28 | 28 |
| Human non-carcinogenic toxicity | 8.7 | 9.1 | 10 | 13 | 8.3 | 8.7 | 8.0 | 8.1 |
| Total | 98 | 110 | 110 | 140 | 92 | 96 | 89 | 90 |

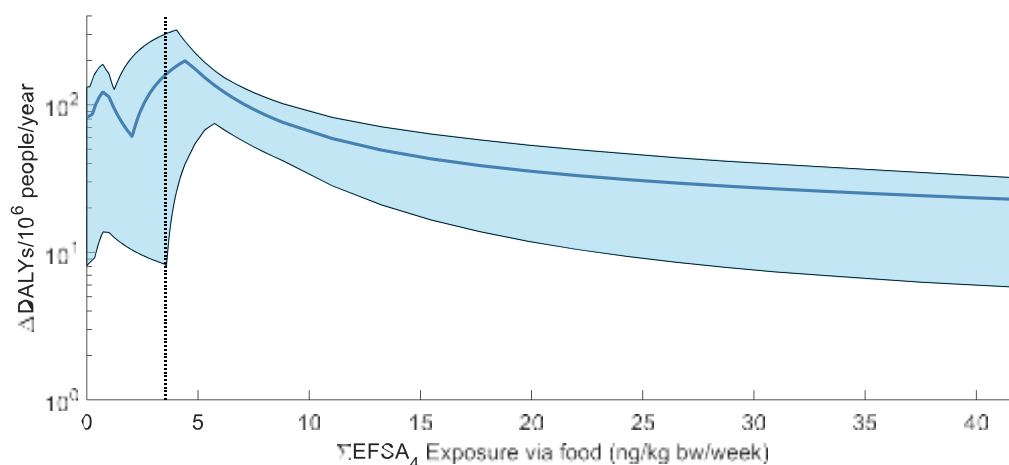


SI Figure 5: Repeated version of main text Figure 3, but with the dietary exposure set to the lower limit of the most recently estimated exposure of the Dutch population.

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SI Figure 6: Repeated version of main text Figure 3, but with the dietary exposure set to the upper limit of the most recently estimated exposure of the Dutch population.



SI Figure 7: Sensitivity analysis over the effect of food exposure on the Δ DALYs between the scenario's with current and targeted drinking water concentrations. The blue line represents the best estimate, and the shaded region the uncertainty range (min-max, derived from 95 % CI over current drinking water concentrations and ERRs). The dotted vertical line is the food exposure at 80 % of the EFSA recommendation. Note that, in the dietary exposure, the ratio between PFOA/PFNA:PFOS/PFHxS is kept constant at the ratio assumed by EFSA (1:2.4).

SI Table 8: Serum/plasma concentrations at age 3 months in exclusively breastfed infants, comparison between measured and modelled results. The EFSA PBPK model assumes exclusive breastfeeding, as a worst-case scenario, so comparisons against plasma levels in breastfed infants are most realistic. Ranges represent the uncertainty from variation in current drinking water concentrations.

| | Measured concentrations | Modelled serum concentrations; current scenario | | |
|----------------------------|---|---|-------------------------------------|-------------------------------------|
| | Plasma conc. in exclusively breastfed infants ¹² | EFSA food exposure ³ | LB food exposure 2022 ¹³ | UB food exposure 2022 ¹³ |
| PFOA/PFNA | 4.04 | 11.7 (11.1-12.3) | 6.1 (5.5-7.1) | 16.4 (15.8-17.1) |
| PFOS/PFHxS | 3.14 | 5.5 (5.3-5.8) | 1.7 (1.5-1.8) | 2.2 (2.1-2.4) |
| Σ EFSA ₄ | 7.18 | 17.2 (16.4-18.1) | 7.8 (7.0-8.9) | 18.6 (17.9-19.5) |

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SI Table 9: Plasma concentrations (ng/mL) at different ages for the different exposure scenario's. EFSA TDI: the tolerably daily intake of 0.63 ng/kg bw/d as calculated by EFSA; 2022 LB: the lower bound dietary exposure calculated by the RIVM for the Dutch population in 2022; 2022 UB: idem, but the upper bound.¹³

| Exposure via food as: | 80 % of EFSA TDI | | 2022 LB | 2022 UB | 80 % of EFSA TDI | 2022 LB | 2022 UB |
|---|------------------|----------------------------------|----------------|------------------|-------------------|---------|---------|
| Drink. water scenario | EFSA TDI | Current DW conc.; mean (95 % CI) | | | Targeted DW conc. | | |
| Blood plasma concentrations at birth (ng/mL) | | | | | | | |
| PFOA/ PFNA | 1.5 | 2.4 (2.3-2.6) | 1.3 (1.1-1.5) | 3.4 (3.3-3.5) | 1.6 | 0.5 | 2.6 |
| PFOS/ PFHxS | 1.8 | 1.7 (1.6-1.8) | 0.5 (0.5-0.6) | 0.7 (0.6-0.7) | 1.5 | 0.3 | 0.5 |
| ΣEFSA ₄ | 3.3 | 4.1 (3.9-4.4) | 1.8 (1.6-2.1) | 4.1 (3.9-4.2) | 3.1 | 0.8 | 3.1 |
| Blood plasma concentrations at age 1 (ng/mL) | | | | | | | |
| PFOA/ PFNA | 8.7 | 14.2 (13.5-14.9) | 7.4 (6.7-8.7) | 20.0 (19.2-20.7) | 9.4 | 2.7 | 15.2 |
| PFOS/ PFHxS | 8.7 | 8.3 (8.1-8.7) | 2.5 (2.3-2.7) | 3.4 (3.1-3.6) | 7.3 | 1.6 | 2.4 |
| ΣEFSA ₄ | 17.4 | 22.5 (21.6-23.6) | 9.9 (9.0-10.4) | 23.4 (22.3-24.3) | 16.7 | 4.3 | 17.6 |
| Blood plasma concentrations at age 35 (ng/mL) | | | | | | | |
| PFOA/ PFNA | 2.0 | 3.3 (3.1-3.4) | 1.7 (1.5-1.9) | 4.6 (4.4-4.8) | 2.2 | 0.6 | 3.5 |
| PFOS/ PFHxS | 4.9 | 4.7 (4.5-4.8) | 1.4 (1.3-1.5) | 1.9 (1.8-2.0) | 4.1 | 0.9 | 1.3 |
| ΣEFSA ₄ | 6.9 | 8.0 (7.6-8.2) | 3.1 (2.9-3.4) | 6.5 (6.2-6.8) | 6.3 | 1.5 | 4.8 |

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SI Table 10: Identified uncertainties per uncertainty type. Green highlight indicates the uncertainty was discussed in the main text.

| Indeterminate | Epistemic | Ambiguity |
|---|---|---|
| Future PFAS concentrations in raw water | PFAS breakthrough with different types of GAC and raw water | Endpoints to consider in estimation of health benefits of lower PFAS exposure |
| Future PFAS regulations | Dose response relationships and DALY weights to relate PFAS serum concentration to health impacts | PFAS molecules to consider and how to account for the impact of the different PFAS molecules (e.g. equipotency, relative potency factors) |
| Type of treatment used (e.g. GAC, ion exchange, etc) and specific life cycle implications (e.g. transportation, waste management) | Concentrations of PFAS below the limit of quantification | LCA assessment method assumptions (e.g. time horizon of climate change impacts, climate change impacts on human health) |
| Future dietary exposure to PFAS and distribution across population | Inventory data for GAC production and reactivation, especially for wood GAC at commercial scale. | How to account for localization of health impact (e.g. local benefit from decreased PFAS exposure, global loss from global warming) |
| | Current dietary exposure to PFAS and distribution across population | |

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