Supporting Information for

The Amazon River – a major source of organic plastic additives to the tropical North Atlantic?

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SUPPLEMENTARY TEXTS

Text S1: Method details concerning GC-MS and LC-HRMS analysis.

GC-MS analysis (Organophosphate ester analysis)

GC analyses were conducted with an Agilent 7820A Series GC coupled with an Agilent 5977E MS, operating in SIM and electron impact (EI, 70 eV) modes. The separation was achieved in a 30 m x 0.25 mm i.d. x 0.25 μ m HP-5MS capillary column (Agilent J&W). All target contaminants were quantified by the internal standard (IS) procedure. The injection volume was 2 μ L and the helium carrier gas flow was 1 mL min⁻¹. The temperatures of the MS transfer line, the ion source and the quadrupole was set to 300, 230 and 150 °C, respectively. The following conditions were applied: injector temperature: 270 °C (splitless) and the oven was programmed from 90 °C to 132 °C at 3 °C min⁻¹, to 166 °C at 10 °C min⁻¹, to 175 at 1 °C min⁻¹ (holding time 2 min), to 232 °C at 2 °C min⁻¹, and then to 300 °C at 25 °C min⁻¹ (holding time 5 min).

LC-QTOF analysis (bisphenol and perfluorinated compound analysis)

Ten microliters of the extract were injected for simultaneous quantification of BPs and PFCs using LC-electrospray ionization quadrupole time of flight mass spectrometry (LC-ESI-QTOF, Agilent 1290 Infinity LC system coupled with Agilent 6530 Accurate-Mass Q-TOF, Agilent Technologies, Les Ulis, France). The syringe was washed externally with MeOH for 20 s before injection to avoid contamination of the injection port or following samples. Separation was achieved on an Agilent Zorbax Eclipse XDB reversed phase column (50 mm x 2.1 mm, 1.8 µm), with the temperature set at 30 °C. Elution was performed with MQ (A) and MeOH (B) under gradient conditions: 0 min 95:5 A/B, 1 min 95:5 A/B, 10 min 0:100 A/B, 15 min 0:100 A/B, 15.1 min 95:5 A/B, 20 min 95:5 A/B. ESI interface was operated in negative mode (3.5 kV capillary voltage), and MS TOF mass acquisition was performed in the range of 50-600 m/z at a rate of 1 spectrum s⁻¹. ESI parameters were set as follows: 300 °C gas temperature, 11 L min⁻¹ drying gas, 40 psig nebulizer, 350 °C sheath gas temperature, 11 L min⁻¹ sheath gas flow. TOF parameters were: 1500 V nozzle voltage, 175 V fragmentor voltage, 65 V skimmer voltage, 750 V octopole voltage. Ion chromatogram extraction was performed with 10 ppm mass tolerance, based on exact monoisotopic mass and retention time.

SUPPLEMENTARY FIGURES







<u>Figure S1:</u> Comparisons between salinity maps of stations Y01-Y14 and different sizes of seeded particle patches (5 km, 10 km, 16 km, 25 km and 50 km). For each patch size, the mean salinity inside the patch and the standard deviation are reported. Black solid lines represent salinity contours for salinity values of 34, 35, 36 and 37. It can be observed that for a patch radius of 50 km, the particles are seeded over a high range of salinities, likely representing different water masses.



Figure S2: Detection frequencies (in %) of the 20 organic compounds analyzed. <u>Abbreviations</u>: TPP: Tripropylphosphate, TiBP: Tri-iso-buthyl phosphate, TnBP: Tri-n-butyl phosphate, TCEP: Tris-(2-chloroethyl) phosphate, TCPP: Tris-(2-chloro, 1-methylethyl) phosphate, TDCP: Tris-(2-chloro-, 1-chloromethylethyl) phosphate, TPhP: Triphenyl phosphate, EHDPP: 2-ethylhexyl-diphenyl phosphate, TEHP: Tris(2-ethylhexyl) phosphate, BPA: Bisphenol A, BPAF: Bisphenol AF, BPAP: Bisphenol F, BPS: Bisphenol S, BPZ: Bisphenol Z, PFHA: Perfluoro hexanoic acid, PFHS: Perfluorohexane sulfonic acid, PFOA: Perfluorooctane sulfonic acid, PFOSF: Perfluoro octane sulfonyl fluoride.







32

10^oW

20^oW

30°W

8°S

60^oW

50^oW

40°W

















Figure S3: Spatial salinity distribution of the surface waters of the study area during each sampling day. The stations indicated in white are the ones sampled the day from which the MERCATOR salinity data originates.



Figure S4: Map showing the position of the sampling area (left) and a zoom (right) showing the sampling stations with the spatial distribution of Kd490 (unit 1/m) in October 2017. Kd490 is a proxy for colored suspended matter and can be used to assess the chlorophyll a distribution.

















Figure S5: Water mass backwards trajectories of all stations (Y01-Y14, from upper left to lower right) except Y16, which is too close to the coast for reliable simulation results. Each map has a caption indicating the station ID, the percentage of particles that passed through the black box (indicating the Amazon Estuary) and the number of days that passed between the sampling of the water masses and their journey in the black box (indicated in brackets). The minimum of salinity (S) reached by particles during their journey in the black box is indicated as well as the river plume (in grey).







Figure S6: Air mass backward trajectories modelled for each sampling station using the NOAA HYSPLIT Model (<u>https://www.arl.noaa.gov/hysplit/hysplit/</u>). Black stars indicate the location of the sampling stations. The model was set to start at 10 m altitude and to run for 315 hours

SUPPLEMENTARY TABLES

| Station ID | Latitude | Longitude | Date | Salinity | |
|------------|---------------|---------------|------------|----------|--|
| | 11000 00011 1 | | 10/02/2015 | | |
| Y 01 | 11°38.2081' N | 21°23.8295° W | 10/07/2017 | 34.4 | |
| Y 03 | 7°46.3369' N | 19°00.1133' W | 10/09/2017 | 34.8 | |
| Y 04 | 8°16.5055' N | 21°31.5612' W | 10/10/2017 | 33.8 | |
| Y 05 | 8°23.2678' N | 25°07.6840' W | 10/11/2017 | 35.1 | |
| Y 06 | 10°19.9553' N | 28°58.0255' W | 10/12/2017 | 34.6 | |
| Y 07 | 11°01.9355' N | 32°28.8749' W | 10/13/2017 | 34.3 | |
| Y 08 | 11°09.4697' N | 35°50.0678' W | 10/14/2017 | 34.8 | |
| Y 09 | 11°22.7072' N | 40°01.6629' W | 10/15/2017 | 34.7 | |
| Y 10 | 11°43.4907' N | 45°03.1796' W | 10/16/2017 | 35.0 | |
| Y 11 | 11°57.9510' N | 49°05.2901' W | 10/17/2017 | 34.5 | |
| Y 12 | 12°33.1485' N | 52°40.1368' W | 10/18/2017 | 34.6 | |
| Y 13 | 13°59.3505' N | 55°42.7139' W | 10/19/2017 | 35.1 | |
| Y 14 | 15°56.6274' N | 59°41.6454' W | 10/20/2017 | 33.7 | |
| Y 16 | 14°32.1003' N | 61°07.9776' W | 10/21/2017 | 33.2 | |

Table S1: GPS positions of the sampling stations, collection dates and surface salinity.

Table S2: Quantification and confirmation ions of OPEs (GC/MS).

| Compound | Full name | Q1 | Q2 | Q3 | Q4 |
|------------------|---|-----|-----|-----|-----|
| TPP | Tripropyl phosphate | 99 | 141 | 123 | 183 |
| TiBP | Tri-iso-buthyl phosphate | 99 | 155 | 139 | |
| TnBP | Tri-n-butyl phosphate | 99 | 155 | 211 | |
| TCEP | Tris-(2-chloroethyl) phosphate | 249 | 251 | 143 | 99 |
| TCPP-1 | Tris-(2-chloro, 1-methylethyl) phosphate | 125 | 99 | 157 | 277 |
| TCPP-2 | Tris-(2-chloro, 1-methylethyl) phosphate | 125 | 99 | 157 | 277 |
| TDCP | Tris-(2-chloro-, 1-chloromethylethyl) phosphate | 191 | 99 | 381 | 209 |
| TPhP | Triphenyl phosphate | 326 | 325 | 215 | 170 |
| EHDPP | 2-ethylhexyl-diphenyl phosphate | 251 | 250 | 326 | |
| TEHP | Tris(2-ethylhexyl) phosphate | 99 | 113 | 112 | |
| TBP- <i>d</i> 27 | Tri-n-butyl-d27 phosphate | 103 | 167 | 231 | |
| TPhP-d15 | Triphenyl-d15 phosphate | 341 | 339 | 223 | 180 |
| TCPP-d18 | Tris-(2-chloro, 1-methylethyl)-d18 phosphate | 131 | 164 | 193 | |
| TDCP-d15 | Tris-(2-chloro-, 1-chloromethylethyl)-d15 phosphate | 197 | 199 | 217 | 394 |
| TPrP- <i>d21</i> | Tri-n-propyl-d21 phosphate | 103 | 131 | 151 | 199 |
| TCEP-d12 | Tris-(2-chloroethyl)-d12 phosphate | 261 | 263 | 148 | 231 |
| MAL-d7 | Malathion-d7 | 174 | 131 | 128 | 164 |

Table S3: [M-H]⁻ values for BPs and PFCs quantification by LC-HRMS Agilent LC-QTOF 6530.

| Compound | Full name | [M-H]- |
|----------|-----------------------------------|----------|
| BPA | Bisphenol A | 227.1078 |
| BPAF | Bisphenol AF | 335.0512 |
| BPAP | Bisphenol AP | 289.1234 |
| BPF | Bisphenol F | 199.0765 |
| BPS | Bisphenol S | 249.0227 |
| BPZ | Bisphenol Z | 267.1391 |
| BPA-d16 | Bisphenol A-d16 | 243.2082 |
| PFHA | Perfluoro hexanoic acid | 312.9728 |
| PFHS | Perfluorohexane sulfonic acid | 398.9366 |
| PFOA | Perfluorooctanoic acid | 412.9664 |
| PFOS | Perfluorooctane sulfonic acid | 498.9302 |
| PFOSF | Perfluorooctane sulfonyl fluoride | 500.9259 |
| PFHA-13C | Perfluoro hexanoic acid-13C | 317.9896 |
| PFHS-13C | Perfluorohexane sulfonic acid-13C | 401.9467 |
| PFOA-13C | Perfluorooctanoic acid-13C | 420.9933 |
| PFOS-13C | Perfluorooctane sulfonic acid-13C | 506.9571 |

| | 100 | | |
|-----------------------------|------|-----------------------|--|
| Compound – | (pg) | (ng L ⁻¹) | |
| TPP (CAS 513-08-6) | 5 | 0.13 | |
| TiBP (CAS 126-71-6) | 1 | 0.03 | |
| TnBP (CAS 126-73-8) | 2 | 0.05 | |
| TCEP (CAS 115-96-8) | 10 | 0.19 | |
| TCPP (CAS 13674-84-5) | 10 | 0.25 | |
| TDCP (CAS 13674-87-8) | 10 | 0.25 | |
| TPhP (CAS 115-86-6) | 5 | 0.10 | |
| EHDPP (CAS 1241-94-7) | 5 | 0.13 | |
| TEHP (CAS 78-42-2) | 10 | 0.20 | |
| TBP-d27 (CAS 61196-26-7) | 2 | 0.05 | |
| TPhP-d15 (CAS 1173020-30-8) | 5 | 0.13 | |
| TCPP-d18 | 30 | 0.75 | |
| TDCP-d15 (CAS 1447569-77-8) | 11 | 0.28 | |
| BPA (CAS 80-05-7) | 10 | 1.0 | |
| BPAF (CAS 14878-61-1) | 1 | 0.1 | |
| BPAP (CAS 1571-75-1) | 20 | 2.0 | |
| BPF (CAS 620-92-8) | 10 | 1.0 | |
| BPS (CAS 80-09-1) | 1 | 0.1 | |
| BPZ (CAS 843-55-0) | 50 | 5.0 | |
| PFHA (CAS 307-24-4) | 80 | 8.0 | |
| PFHS (CAS 3871-99-6) | 1 | 0.1 | |
| PFOA (CAS 335-67-1) | 20 | 2.0 | |
| PFOS (CAS 1763-23-1) | 1 | 0.1 | |
| PFOSF (CAS 307-35-7) | 10 | 1.0 | |

<u>Table S4:</u> Limits of quantification (LOQ) expressed as absolute amounts (pg) and ng L^{-1} in seawater.

| Table S5: Concentrations (in ng L ⁻¹ |) of all 20 molecules analyzed for each s | ampling station as well as extractior | ı, transport and laboratory blank v | alues. Sampling station data a | realready blank-corrected. For each |
|---|---|---------------------------------------|-------------------------------------|--------------------------------|-------------------------------------|
| sampling station, the average of the | concentrations measured in the two rep! | icates is given. | | | |

| | Extraction Blank | Extraction Blank | Extraction Blank | Transport Blank | Laboratory Blank | Y 01 | Y 03 | Y 04 | ¥ 05 | Y 06 | Y 07 | Y 08 | Y 09 | Y 10 | Y 11 | Y 12 | ¥ 13 | Y 14 | Y 16 |
|--------------------|---|---|---|---|---|--|---|--|--|---|---|---|-------|---|---|---|---|---|---------------------|
| TPP | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| TiBP | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>3.9</td><td>6.5</td><td>1.8</td><td>2.7</td><td>2.0</td><td>5.2</td><td>6.2</td><td>11</td><td>33</td><td>38</td><td>30</td><td>17</td><td>1.7</td><td><loq< td=""></loq<></td></loq<> | n.d. | n.d. | n.d. | n.d. | 3.9 | 6.5 | 1.8 | 2.7 | 2.0 | 5.2 | 6.2 | 11 | 33 | 38 | 30 | 17 | 1.7 | <loq< td=""></loq<> |
| TnBP | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| TCEP | n.d. | n.d. | n.d. | n.d. | n.d. | <loq< td=""><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | 1.0 | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>1.3</td><td>n.d.</td><td>1.6</td><td>1.2</td><td>1.6</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<> | 1.3 | n.d. | 1.6 | 1.2 | 1.6 | <loq< td=""><td><loq< td=""></loq<></td></loq<> | <loq< td=""></loq<> |
| TCPP | 29 | 7.9 | 31 | 5.4 | 6.7 | 203 | 248 | 141 | 142 | 205 | 290 | 577 | 466 | 921 | 1300 | 966 | 596 | 104 | 74 |
| TDCP | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| TPhP | <loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td><loq< td=""><td>2.2</td><td>6.9</td><td>1.5</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>n.d.</td><td><loq< td=""><td>2.2</td><td>6.9</td><td>1.5</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td><loq< td=""><td>2.2</td><td>6.9</td><td>1.5</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | n.d. | <loq< td=""><td>2.2</td><td>6.9</td><td>1.5</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | 2.2 | 6.9 | 1.5 | 1.0 | <loq< td=""><td><loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>1.3</td><td><loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<> | 1.3 | <loq< td=""><td>1.5</td><td>1.6</td><td>1.3</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<> | 1.5 | 1.6 | 1.3 | <loq< td=""><td><loq< td=""></loq<></td></loq<> | <loq< td=""></loq<> |
| EHDPP | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>2.4</td><td>5.4</td><td>2.4</td><td>1.2</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>2.4</td><td>5.4</td><td>2.4</td><td>1.2</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td>2.4</td><td>5.4</td><td>2.4</td><td>1.2</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>2.4</td><td>5.4</td><td>2.4</td><td>1.2</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>2.4</td><td>5.4</td><td>2.4</td><td>1.2</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | 2.4 | 5.4 | 2.4 | 1.2 | <loq< td=""><td><loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>1.2</td><td>1.0</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | 1.2 | 1.0 | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""></loq<></td></loq<> | <loq< td=""></loq<> |
| TEHP | n.d. | n.d. | n.d. | 15 | 16 | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| Σ_{9} | 29 | 79 | 31 | 20.4 | 22.7 | 211.5 | 267.8 | 146 7 | 146.9 | 207.0 | 295.2 | 583 2 | 4808 | 955.0 | 13411 | 998.8 | 615.9 | 105 7 | 74 0 |
| OPEs | 2) | 1.9 | 51 | 20.4 | 22.1 | 211.5 | 207.0 | 140.7 | 140.7 | 207.0 | 275.2 | 505.2 | 400.0 | 755.0 | 1341.1 | <i>))</i> 0.0 | 015.7 | 103.7 | 74.0 |
| BPA | 1.0 | 1.0. | <loq< td=""><td>n.d.</td><td>n.d.</td><td>1.2</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td><loq< td=""><td>1.8</td><td>1.8</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | n.d. | n.d. | 1.2 | n.d. | n.d. | n.d. | n.d. | n.d. | <loq< td=""><td>1.8</td><td>1.8</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | 1.8 | 1.8 | <loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. |
| BPAF | <loq< td=""><td><loq< td=""><td>1.0</td><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td>1.0</td><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | 1.0 | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| BPAP | n.d. | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| BPF | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| BPS | <loq< td=""><td>n.d.</td><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td><loq< td=""><td>n.d.</td><td>1.1</td><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | n.d. | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td><loq< td=""><td>n.d.</td><td>1.1</td><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | <loq< td=""><td>n.d.</td><td>1.1</td><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | n.d. | 1.1 | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. |
| BPZ | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| Eres BPs | 1.0 | 1.0 | 1.0 | | | 1.2 | | | | | | | 1.8 | 2.9 | | | | | |
| PFHA | 2.2 | <loq< td=""><td>1.0</td><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>9.7</td><td>16</td><td>11</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | 1.0 | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>9.7</td><td>16</td><td>11</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>9.7</td><td>16</td><td>11</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | 9.7 | 16 | 11 | n.d. | n.d. | n.d. |
| PFHS | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | 4.4 | n.d. | n.d. | n.d. | n.d. |
| PFOA | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>2.9</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | 2.9 | n.d. | n.d. | n.d. | n.d. |
| PFOS | <loq< td=""><td>n.d.</td><td><loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>2.3</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<></td></loq<> | n.d. | <loq< td=""><td><loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>2.3</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<></td></loq<> | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>2.3</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>2.3</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | 2.3 | n.d. | n.d. | n.d. | n.d. |
| PFOSF | n.d. | n.d. | n.d. | <loq< td=""><td><loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<></td></loq<> | <loq< td=""><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td><td>n.d.</td></loq<> | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| Σ_5 PFCs | 2.2 | | 1.0 | | | | | | | | | | | 9.7 | 25.6 | 11.0 | | | |
| Total | | | | | | 212.7 | 267.8 | 146.7 | 146.9 | 207.0 | 295.2 | 583.2 | 482.6 | 967.6 | 1366.7 | 1009.8 | 615.9 | 105.7 | 74.0 |
| | | | | | | • | | | | | | | | | | | | | |

n.d. = not detected; LOQ = limit of quantification

Abbreviations: TPP: Tripropyl phosphate, TiBP: Tri-iso-buthyl phosphate, TnBP: Tri-n-butyl phosphate, TCEP: Tris-(2-chloroethyl) phosphate, TCPP: Tris-(2-chloro, 1-methylethyl) phosphate, TDCP: Tris-(2-chloro, 1-methylethyl) phosphate, TDCP: Tris-(2-chloro, 1-chloromethylethyl) phosphate, TPP: Triphenyl phosphate, EHDPP: 2-ethylhexyl-diphenyl phosphate, TEHP: Tris(2-ethylhexyl) phosphate, BPA: Bisphenol A, BPAF: Bisphenol AF, BPAP: Bisphenol AP, BPF: Bisphenol S, BPZ: Bisphenol Z, PFHA: Perfluoro hexanoic acid, PFHS: Perfluorohexane sulfonic acid, PFOA: Perfluorooctane sulfonic acid, PFOS: Perflu

Table S6: Surrogate recovery rates (in %) for each sampling station and the extraction blanks. Transport and laboratory blanks were not spiked with surrogate standards, which is why no data are available in this case.

| | Extraction Blank | Extraction Blank | Extraction Blank | Transport Blank | Laboratory Blank | Y 01 | Y 03 | Y 04 | ¥ 05 | Y 06 | Y 07 | Y 08 | Y 09 | Y 10 | Y 11 | Y 12 | ¥ 13 | Y 14 | ¥ 16 |
|----------|---------------------|---------------------|---------------------|--------------------|---------------------|------|------|------|------|------|------|------|------|------|------|-------|------|------|------|
| D27-TBP | 83.0 | 84.4 | 77.6 | n.a. | n.a. | 87.7 | 74.9 | 81.6 | 75.8 | 64.4 | 74.4 | 72.1 | 79.9 | 82.7 | 82.0 | 87.7 | 86.2 | 63.2 | 82.2 |
| D18-TCPP | 63.8 | 62.4 | 62.2 | n.a. | n.a. | 69.5 | 67.3 | 66.7 | 68.5 | 67.8 | 74.5 | 69.4 | 71.6 | 73.4 | 75.5 | 79.1 | 67.0 | 59.8 | 72.1 |
| D15-TDCP | 106.9 | 89.1 | 54.4 | n.a. | n.a. | 76.0 | 74.9 | 81.5 | 69.2 | 63.3 | 74.4 | 71.3 | 80.4 | 64.8 | 71.1 | 81.9 | 82.8 | 67.0 | 85.1 |
| D15-TPhP | 94.4 | 82.6 | 63.2 | n.a. | n.a. | 64.5 | 62.0 | 69.9 | 56.5 | 48.7 | 67.4 | 61.7 | 73.3 | 62.0 | 71.9 | 101.9 | 82.8 | 77.8 | 83.5 |

n.a. = not available

Abbreviations: D27-TBP: D27-Tri-n-butyl phosphate, D18-TCPP: D18-Tris (2-chloroisopropyl) phosphate, D15-TDCP: D15-Tris (1,3-dichloro-2-propyl) phosphate, D15-TPhP: D15-Triphenyl phosphate.

| | 0.5 par | ticles km ⁻² | 1.3 part | icles km ⁻² | 2.5 particles km ⁻² | | Sensitivity | |
|---------------------------------|------------------------------|--|------------------------------|--|--------------------------------|--|-------------|--|
| Y 01 | % of particles | travel time (days) | % of particles | travel time (days) | % of particles | travel time (days) | | |
| 5 km 10 km 16 km 25 km | 0% 0% 0% 0% | | 0% 0% 0% | | 0% 0% 0% | | 0 % | |
| Y 03 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 0% 0% 0% 0% | | 0% 0% 0% 0% | | 0% 0% 0% 0% | | 0% | |
| Y 04 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 74% 67% 60% | [-124,-125] [-124,-125] [-124,-126] [-124,-180] | 100% 82% 70% 60% | [-124,-125] [-124,-125] [-124,-125] [-124,-126] | 100% 80% 68% 61% | [-124,-125] [-124,-125] [-124,-128] [-124,-127] | 40% | |
| Y 05 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 0% 0% 0% 7% | [-92,-126] | 0% 0% 0% 8% | [-92,-126] | 0% 0% 0% 7% | [-92,-126] | 8% | |
| Y 06 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 100% | [-90,-146] [-90,-146] [-88,-151] [-88,-147] | 100% 100% 100% 100% | [-90,-113] [-89,-147] [-88,-147] [-82,-148] | 100% 100% 100% 100% | [-90,-146] [-83,-151] [-84,-147] [-81,-162] | 0% | |
| Y 07 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 100% | [-88,-92] [-88,-92] [-88,-92] [-88,-114] | 100% 100% 100% | [-88,-92] [-88,-92] [-88,-92] [-88,-117] | 100% 100% 100% | [-88,-92] [-88,-92] [-88,-92] [-88,-127] | 0% | |
| Y 08 | | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 100% | [-77,-78] [-76,-78] [-76,-78] [-76,-85] | 100% 100% 100% | [-76,-78] [-76,-78] [-76,-78] [-76,-85] | 100% 100% 100% | [-76,-78] [-76,-78] [-76,-78] [-76,-85] | 0% | |
| Y 09 | | | | | | | | |

<u>Table S7:</u> Sensitivity tests results for radii from 5 to 25 km and densities from 0.5 to 2.5 particles km⁻².

| | 1 | 1 | | 1 | | 1 | |
|---------------------------------|------------------------------|--|------------------------------|--|------------------------------|--|-----|
| 5 km 10 km 16 km 25 km | 0% 0% 0% 0% | | 0% 0% 0% 0% | | 0% 0% 0% 0% | | 0% |
| Y 10 | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 96% | [-76,-115] [-76,-115] [-76,-115] [-76,-119] | 100% 100% 100% 96% | [-76,-115] [-76,-115] [-76,-115] [-76,-119] | 100% 100% 100% 96% | [-76,-115] [-76,-120] [-76,-115] [-76,-119] | 4% |
| Y 11 | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 97% | [-61,-65] [-60,-91] [-60,-115] [-60,-116] | 100% 100% 100% 97% | [-61,-65] [-60,-116] [-60,-96] [-60,-122] | 100% 100% 100% 97% | [-61,-65] [-60,-116] [-60,-116] [-60,-120] | 3% |
| Y 12 | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 99% 93% 94% | [-66,-105] [-65,-140] [-52,-140] [-37,-159] | 100% 98% 92% 95% | [-65,-111] [-65,-154] [-52,-153] [-37,-140] | 100% 97% 92% 95% | [-65,-139] [-65,-153] [-52,-159] [-37,-159] | 8% |
| Y 13 | | | | | | | |
| 5 km 10 km 16 km 25 km | 100% 100% 100% 100% | [-59,-81] [-58,-106] [-57,-107] [-54,-107] | 100% 100% 100% 100% | [-59,-106] [-58,-105] [-57,-107] [-54,-107] | 100% 100% 100% 100% | [-59,-107] [-58,-106] [-57,-107] [-54,-107] | 0% |
| Y 14 | | | | | | | |
| 5 km 10 km 16 km 25 km | 33% 23% 23% 46% | [-138,-141] [-133,-156] [-78,-157] [-78,-175] | 30% 20% 26% 46% | [-138,-156] [-131,-179] [-78,-168] [-78,-180] | 29% 21% 25% 46% | [-132,-159] [-131,-174] [-78,-179] [-78,-179] | 26% |