

Table S1. Gas chromatography (GC) and mass spectrometry (MS) conditions.

| GC conditions | |
|----------------------|---|
| Column | DB-5ms (0.25 mm i.d. × 30 m length, film thickness 0.25 μm) |
| Injection volume | 1 μL |
| Injection system | Splitless |
| Injection temp. | 270 °C |
| Carrier gas | He |
| Gas flow rate | 1.2 mL/min |
| Column temp. | 60 °C (1 min hold) - 190 °C up (15°C/min, 3 min hold) - 220 °C up (2 °C/min, 1 min hold) - 300°C up (5 °C/min, 20 min hold) |
| Interface temp. | 270 °C |
| Ion source temp. | 280 °C |
| Quadrupole temp. | 150 °C |

Table S2. Mass transition and approximate retention time of target analytes and internal standards.

| Target analyte | Quantification ion (m/z) | Confirmation ion (m/z) | Retention time (min) |
|---------------------------------|--------------------------|------------------------|----------------------|
| Acenaphthylene | 152 | 151 | 9.46 |
| Acenaphthene | 154 | 153 | 9.75 |
| Fluorene | 166 | 165 | 10.71 |
| Anthracene | 178 | 176 | 13.57 |
| Phenanthrene | 178 | 176 | 13.38 |
| Fluoranthene | 202 | 201 | 19.35 |
| Pyrene | 202 | 201 | 20.76 |
| Chrysene | 228 | 226 | 31.18 |
| Benz[<i>a</i>]anthracene | 228 | 226 | 30.94 |
| Benzo[<i>b</i>]fluoranthene | 252 | 253 | 37.90 |
| Benzo[<i>k</i>]fluoranthene | 252 | 253 | 38.05 |
| Benzo[<i>a</i>]pyrene | 252 | 253 | 39.44 |
| Dibenz[<i>a,h</i>]anthracene | 278 | 279 | 44.44 |
| Indeno[1,2,3- <i>cd</i>]pyrene | 276 | 277 | 45.13 |
| Benzo[<i>ghi</i>]perylene | 276 | 277 | 44.21 |
| D10-Acenaphthene | 164 | 162 | 9.69 |
| D10-Phenanthrene | 188 | 184 | 13.30 |
| D10-Pyrene | 212 | 211 | 20.65 |
| D12-Benzo[<i>a</i>]pyrene | 264 | 263 | 39.32 |

Table S3. Total concentration of PAHs grouped by ring number in analyzed samples among twelve sampling sites (ng/g-dw or ng/L).

| Sample type | Prefecture | 3 ring | 4 ring | 5 ring | 6 ring |
|--------------------|-------------------|---------------|---------------|---------------|---------------|
| Wharf roach | Aomori | 18.8 | 29.9 | 0.07 | 0.41 |
| | Akita | 27.4 | 66.7 | 0.16 | 1.64 |
| | Yamagata | 12.1 | 40.7 | 0.24 | n.d. |
| | Niigata | 20.1 | 64.8 | 0.13 | 0.22 |
| | Ishikawa | 22.4 | 24.4 | 0.79 | 0.21 |
| | Kyoto | 12.9 | 28.0 | 0.40 | 0.74 |
| | Hyogo | 7.79 | 30.6 | 1.83 | 3.90 |
| | Shimane | 9.87 | 33.5 | 0.33 | 0.41 |
| | Yamaguchi | 5.29 | 19.9 | 0.39 | 1.36 |
| | Fukuoka | 17.1 | 35.2 | 0.27 | 0.17 |
| | Saga | 22.8 | 44.0 | 6.79 | 11.3 |
| | Nagasaki | 10.1 | 27.3 | 0.23 | 0.08 |
| Drifting seaweed | Aomori | 5.89 | 14.0 | 5.63 | 8.90 |
| | Akita | 1.03 | 10.1 | 0.75 | 0.44 |
| | Yamagata | 3.85 | 25.7 | 0.97 | 0.52 |
| | Niigata | 0.66 | 10.3 | 0.38 | 0.45 |
| | Ishikawa | 16.6 | 59.8 | 3.20 | 2.84 |
| | Kyoto | 6.88 | 71.9 | 4.98 | 5.72 |
| | Hyogo | 5.82 | 43.3 | 0.65 | 0.77 |
| | Shimane | n.a. | n.a. | n.a. | n.a. |
| | Yamaguchi | 5.77 | 9.73 | 2.59 | 3.24 |
| | Fukuoka | 11.4 | 35.9 | 8.73 | 6.87 |
| | Saga | 5.53 | 8.59 | 1.57 | 2.96 |
| | Nagasaki | 12.9 | 64.8 | 17.8 | 13.9 |
| Sea water | Aomori | 10.7 | 1.43 | 0.17 | 0.16 |
| | Akita | 15.3 | 2.73 | 0.69 | 0.39 |
| | Yamagata | 6.79 | 0.90 | 0.11 | n.d. |
| | Niigata | 8.42 | 1.35 | 0.13 | n.d. |
| | Ishikawa | 11.3 | 1.36 | 0.15 | n.d. |
| | Kyoto | 12.8 | 2.56 | 0.21 | 0.07 |

| | | | | | |
|-----------|-----------|------|------|------|------|
| | Hyogo | 13.2 | 2.54 | 0.45 | 0.16 |
| | Shimane | 8.75 | 1.07 | 0.18 | 0.03 |
| | Yamaguchi | 9.23 | 0.86 | 0.12 | n.d. |
| | Fukuoka | 14.7 | 2.82 | 0.25 | 0.06 |
| | Saga | 10.6 | 1.11 | 0.20 | 0.04 |
| | Nagasaki | 13.0 | 2.45 | 0.65 | 0.32 |
| Soil/sand | Aomori | 3.47 | 16.1 | 3.59 | 0.59 |
| | Akita | 2.27 | 7.09 | 0.50 | 0.09 |
| | Yamagata | 30.5 | 6.24 | 0.87 | 0.02 |
| | Niigata | 4.69 | 13.2 | 2.10 | 0.27 |
| | Ishikawa | 41.6 | 226. | 211 | 27.5 |
| | Kyoto | 20.5 | 71.3 | 39.5 | 6.29 |
| | Hyogo | 3.78 | 28.5 | 23.1 | 2.70 |
| | Shimane | 7.88 | 31.3 | 7.26 | 1.01 |
| | Yamaguchi | 1.05 | 0.51 | 0.49 | n.d. |
| | Fukuoka | 1.20 | 1.58 | 0.58 | 0.09 |
| | Saga | 0.38 | 1.17 | 1.96 | 0.34 |
| | Nagasaki | 1.69 | 6.02 | 2.98 | 0.39 |



Figure S1. Pollution in the intertidal and supralittoral zones in the coastal Japan area.

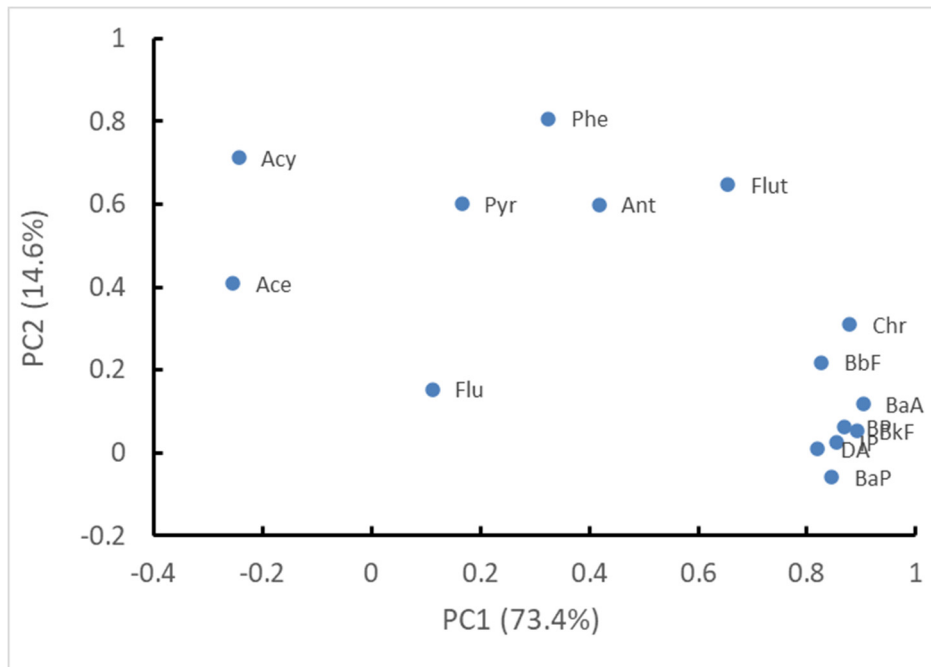


Figure S2. The loadings plot corresponding to the PCA scores plot for the first two PCs (Figure 7), accounting for 88% of the variance.