**Supplementary information**

**4DEMON: integrating 40 years of data on PCB and metal contamination in marine sediments of the Belgian Part of the North Sea.**

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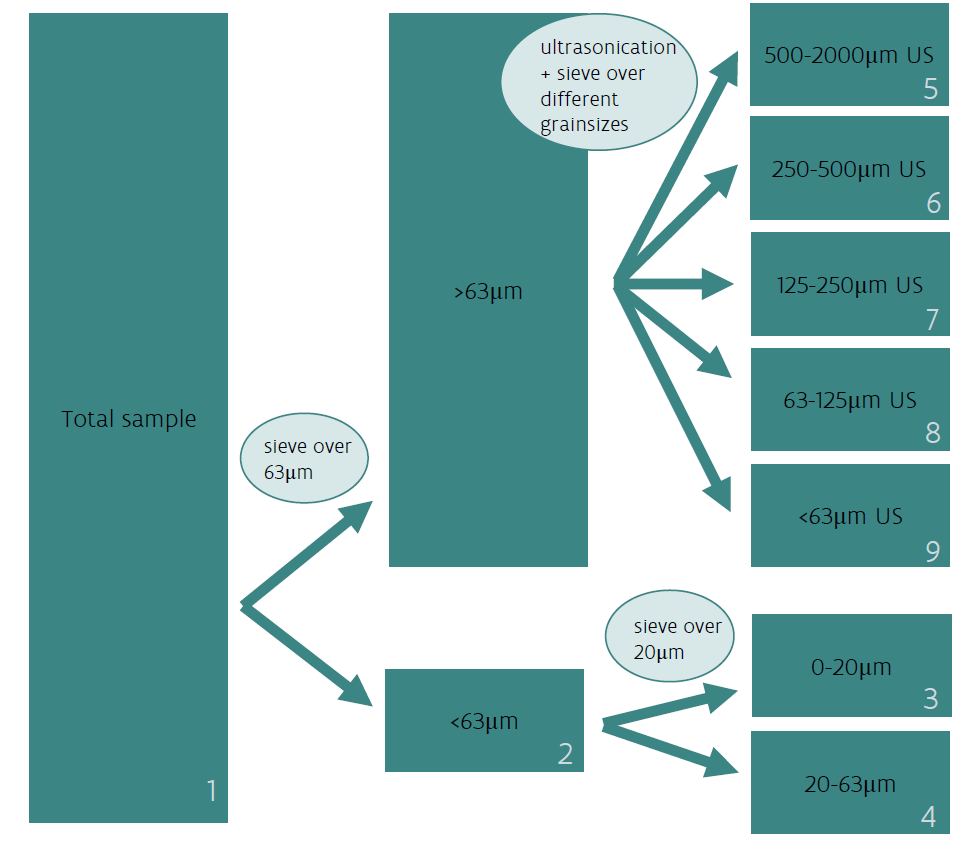
† These authors share last authorship

1. Data collection

Table S1. Overview of projects, period and number of records used for PCB and metal trend analyses in this study (DS1)

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  |  | Number of records per grain size class (µm) | | | | |
| Project | **Period** | 0-37 | 0-63 | 0-500 | 0-2000 | 0-10000 |
| PMPZ | 1971-1975 | 737 |  |  |  |  |
| MMP\_NS | 1978-1983 |  |  |  | 88 |  |
| MONIT\_SED | 1979-2015 |  | 7154 | 84 | 3118 |  |
| PhD Van Alsenoy | 1987-1988 |  | 157 |  |  | 257 |
| MONIT\_Sludge | 2004-2015 |  | 11947 |  |  |  |

1. Normalization

Figure S1. Sample fractionation scheme, based on Smedes & Nummerdor (2003)

1. Time trend models

All time trend plots for metals and PCBs are provided below. Data points represent the derived concentrations, after normalization. Coloured regression lines represent the fitted time trend within each cluster, scaled to the first quadrimester and to the most recent analytical method. A single regression is visible when there is no significant difference between cluster zones.

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S2. Trends in As at the BPNS, applying (A) granulometric normalization (<63 µm, 1997-2015)), (B) Ni normalization (1997-2015), (C) Hybrid method 1 (1997-2015) and (D) Hybrid method 2 (1997-2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S3. Trends in Cr at the BPNS, applying (A) granulometric normalization (<63 µm, 1987-2015)), (B) Ni normalization (1971-2015), (C) Hybrid method 1 (1979-2015) and (D) Hybrid method 2 (1979-2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |

Fig. S4. Trends in Cu at the BPNS, applying (A) granulometric normalization (<63 µm, 1987-2015)), (B) Ni normalization (1971-2015), (C) Hybrid method 1 (1979-2015) and (D) Hybrid method 2 (1979-2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S5. Trends in Pb at the BPNS, applying (A) granulometric normalization (<63 µm, 1987-2015)), (B) Ni normalization (1971-2015), (C) Hybrid method 1 (1979-2015) and (D) Hybrid method 2 (1979-2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S6. Trends in Zn at the BPNS, applying (A) granulometric normalization (<63 µm, 1987-2015)), (B) Ni normalization (1971-2015), (C) Hybrid method 1 (1979-2015) and (D) Hybrid method 2 (1979-2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S7. Trends in Cd at the BPNS, applying (A) granulometric normalization (<63 µm, 1990 – 2015)), (B) Ni normalization (1987-2015), (C) Hybrid method 1 (1990 – 2015) and (D) Hybrid method 2 (1990 – 2015).

|  |  |
| --- | --- |
| (A) | (B) |
| (C) | (D) |
|  | |

Fig. S8. Trends in Hg at the BPNS, applying (A) granulometric normalization (<63 µm, 1990 – 2015)), (B) Ni normalization (1971 – 2015), (C) Hybrid method 1 (1979 – 2015) and (D) Hybrid method 2 (1979 – 2015).

|  |  |
| --- | --- |
|  | |
| (A) | (B) |
| (C) | (D) |
| (E) | (F) |
| (G) |  |

Fig. S9. Trends in PCBs (1991-2016) at the BPNS, granulometric normalization (< 63µm). (A) CB28, (B) CB52, (C) CB101, (D) CB118, (E) CB138, (F) CB153, (G) CB180.

Table S2. Summary of metals and PCBs trend models.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | Data count | Removed outliers | | Time effect | Zone effect | Interaction time-zone | Analytical method effect | Season effect |
| **Granulometric** | | | | | | | | |
| Al | 1129 | 1 | Yes | | yes - zone 1 higher | Yes | Yes | Yes - season 3 higher |
| Fe | 1149 | 4 | Yes | | Yes | Yes | No | Yes - season 3 higher |
| Ni | 1143 | 0 | Yes | | Yes | Yes | Yes | No |
| As | 1009 | 0 | Yes | | Yes | Yes | Yes | No |
| Cd | 1124 | 0 | Yes | | Yes | No | Yes | No |
| Cr | 1143 | 2 | Yes | | Yes - zone 1 higher | Yes | Yes | No |
| Cu | 1131 | 0 | Yes | | yes - zone 1 lower | Yes | Yes | No |
| Pb | 1153 | 3 | Yes | | yes - zone 1 lower | Yes | Yes | No |
| Zn | 1155 | 0 | Yes | | Yes | No | No | Yes - season 3 higher |
| Cd | 1124 | 0 | Yes | | No | No | Yes | No |
| Hg | 1124 | 0 | Yes | | Yes | no | no | no |
| CB28 | 935 | 0 | Yes | | Yes | Yes | No | Yes - season 3 higher |
| CB521 | 935 | 0 | No | | Yes | No | No | Yes - season 3 higher |
| CB101 | 956 | 0 | Yes | | Yes | Yes | No | Yes - season 3 higher |
| CB118 | 969 | 1 | Yes | | Yes | Yes | No | No |
| CB138 | 967 | 0 | Yes | | yes - zone 1 higher | Yes | No | No |
| CB153 | 450 | 0 | Yes | | Yes | Yes | Yes | No |
| CB180 | 952 | 0 | Yes | | Yes | Yes | No | No |
| **Hybrid 1** | | | | | | | | |
| As | 994 | 1 | Yes | | Yes - zone 1 higher | Yes | Yes | Yes - season 3 higher |
| Cd | 1188 | 0 | Yes | | yes - zone 1 lower | No | Yes | No |
| Cr | 1198 | 0 | Yes | | Yes | Yes | Yes | Yes - season 2 higher |
| Cu | 1176 | 2 | Yes | | Yes | Yes | Yes | Yes - season 1 higher |
| Hg | 1199 | 0 | Yes | | Yes | Yes | Yes | Yes - season 1 higher |
| Ni | 1212 | 1 | Yes | | Yes - zone 1 and 4 higher, zone 3 and 5 lower | No | Yes | Yes - season 2 higher |
| Pb | 1228 | 1 | Yes | | Yes - zone 1 and 4 higher, zone 3 and 5 lower | No | Yes | Yes - season 2 higher |
| Zn | 1204 | 1 | Yes | | Yes - zone 5 lower | Yes | Yes | Yes |
| **Hybrid 2** | | | | | | | | |
| As | 1069 | 0 | Yes | | Yes - zone 1 higher with stronger increase | Yes | Yes | Yes |
| Cd | 1318 | 2 | Yes | | Yes - zone 1 lower | No | Yes | Yes - season 2 lower |
| Cr | 1400 | 0 | Yes | | Yes - zone 1 lower | No | Yes | Yes |
| Cu | 1344 | 3 | Yes | | No | No | Yes | Yes - season 3 lower |
| Hg | 1386 | 1 | Yes | | Yes - zone 1 lower | No | Yes | Yes |
| Ni | 1390 | 0 | Yes | | Yes | No | Yes | Yes |
| Pb | 1373 | 1 | Yes | | Yes | Yes | Yes | Yes |
| Zn | 1420 | 2 | Yes | | Yes | No | Yes | Yes |
| **Ni normalized** | | | | | | | | |
| As | 1036 | 2 | Yes | | Yes - zone 4 lower | Yes | Yes | No |
| Cd | 1270 | 0 | Yes | | yes - zone 1 lower | No | Yes | Yes - season 2 lower |
| Cr | 1469 | 1 | Yes | | Yes - zone 1 lower | No | Yes | No |
| Cu | 1379 | 0 | Yes | | Yes | Yes | Yes | No |
| Hg | 1439 | 0 | Yes | | Yes | Yes | Yes | No |
| Pb | 1415 | 0 | Yes | | Yes | No | Yes | No |
| Zn | 1435 | 0 | Yes | | No | No | Yes | Yes |

1 CB52 model with AR-1 auto-correlation

2 Model zone 2 fitted on 10 year