Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.


**Supplementary information**

# Selenium concentration in herring from the Baltic Sea tracks decadal and spatial trends in external sources

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**Text S 1.** Differences between sexes

We find that, except for the gonads, the difference between Se concentrations in female and male tissues are less than 23% (average 10% difference) across tissues and only 6% for the whole fish. While the Se concentration is higher in female gonads, the size of the gonads is larger in males (female: 0.89 g (range: 0.29-1.80); male: 2.05 g (range: 0.67-3.8); SI Table S4) resulting in the total Se mass found in gonads to be similar (0.66 μg; 0.70 μg). The gonads change size depending on the fish reproductive phase. Based on the size of the gonads all specimen are likely in the early- to mid-maturation phase where gonad weight is increasing [*Bucholtz et al.*, 2008]. The result indicates that the increase in size of the reproductive organs might happens without an additional transfer of Se resulting in a dilution of Se in the tissue. We conclude that there is no large difference in herring concentration between the sexes for the tissue of interest to this study but that reproductive state can play a role for the gonad concentration.

***Table S 1.*** *Names and coordinates for stations used in the study. Data source: Danielsson et al. [2018]; Enhus et al. [2011]; Soerensen and Faxneld [2020]*

| **Station** | **Latitude** | **Longitude** | **Basin** |
|---|---|---|---|
| Ranefjarden | 65.76 | 22.42 | Bothnian Bay |
| Harufjarden | 65.58 | 22.88 | Bothnian Bay |
| Kinnbacksfjarden | 64.85 | 21.27 | Bothnian Bay |
| Holmoarna | 63.68 | 20.88 | Bothnian Sea |
| Gaviksfjarden | 63.13 | 18.64 | Bothnian Sea |
| Langvindsfjarden | 61.77 | 17.46 | Bothnian Sea |
| Bothnian Sea offshore | 61.25 | 19.5 | Bothnian Sea |
| Angskarsklubb | 60.53 | 18.16 | Bothnian Sea |
| Sea of Åland offshore | 60.25 | 19.35 | Bothnian Sea |
| Lagno | 59.43 | 18.63 | Northern Baltic Proper |
| Northern Baltic Proper offshore | 58.75 | 20.5 | Northern Baltic Proper |
| Landsort | 58.69 | 18 | Northern Baltic Proper |
| Byxelkrok | 57.41 | 16.99 | Southern Baltic Proper |
| Utlangan | 56.01 | 15.83 | Southern Baltic Proper |
| Hanobukten | 55.76 | 14.29 | Southern Baltic Proper |
| Eastern Bornholm basin offshore | 55.75 | 17.5 | Southern Baltic Proper |
| Abbekas | 55.32 | 13.61 | Southern Baltic Proper |
| Vaderoarna | 58.55 | 10.96 | Skagerak |
| Fladen | 57.2 | 11.81 | Kattegat |
| Kullen | 56.33 | 12.38 | Kattegat |
| Lilla Vartan | 59.35 | 18.13 | Stockholm Archipelago |

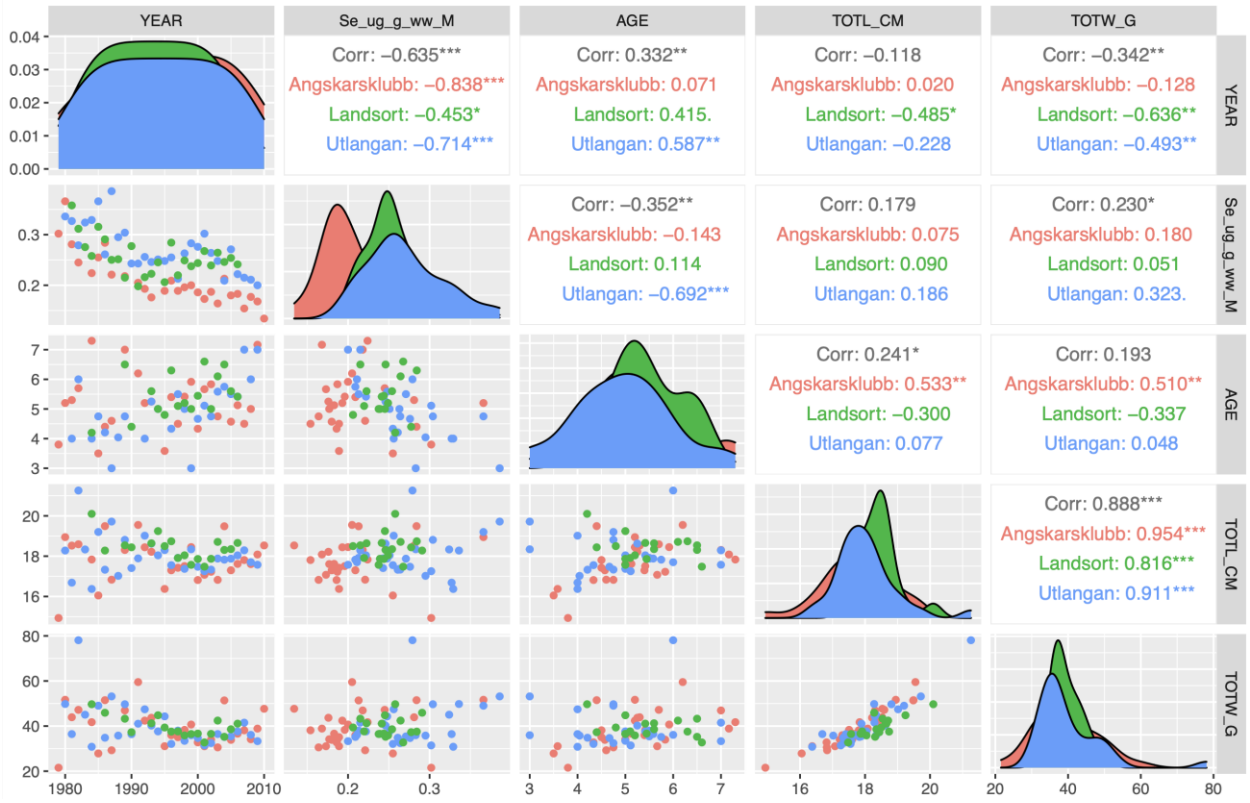



***Figure S 1***. *Correlation between Se muscle concentration and biological variables from the retrospective time series indicating the effect of year but no systematic effect of age, length, or weight of herring.*

***Table S 2**. Compilation of overlapping data on muscle and liver Se concentration and individual conversion factors (k).*

| Site | Year | n in sample pool | Se muscle (μg $g^{-1}$ ww) | Muscle (% dw $ww^{-1}$) | Se muscle (μg $g^{-1}$ dw) | Se liver (μg $g^{-1}$ dw) | $k_{liver/muscle}$ |
|---|---|---|---|---|---|---|---|
| Angskarsklubb | 2009 | 12 | 0.17 | 21.9[A] | 0.77 | 5.25 | 6.82 |
| Angskarsklubb | 2010 | 12 | 0.13 | 21.3 [A] | 0.63 | 5.60 | 8.89 |
| Utlangan | 2009 | 12 | 0.20 | 20.9 [A] | 0.96 | 8.20 | 8.54 |
| Landsort | 2006/2009[B] | 12 | 0.24 | 21.9 | 1.10 | 6.36 | 5.78 |
| Lilla Vartan females | 2016 | 25 | | | 0.52 | 3.81 | 7.33 |
| Lilla Vartan males | 2016 | 25 | | | 0.45 | 3.80 | 8.44 |
| Lilla Vartan mixed sexes[C] | 2016 | 12 | | | 0.48 | 3.34 | 6.96 |
| All samples | | | | | | | 7.49 |

A) Muscle dw/ww ratio is extracted from the average of 12 herring muscle samples from the Swedish monitoring program of the given year and site. The ratio is thus from the same site and year but from different fish than those from which the muscle sample for Se analysis was taken from.

B) In Landsort, there was no overlap between muscle data in the retrospective dataset and liver samples in the Swedish monitoring program, we therefore combined the latest year in the retrospective time series (2006) with the earliest year from the monitoring program (2009). For Landsort's herring, the muscle dw/ww ratio was measured on the same fish analyzed for Se.

C) In addition to the 2×25 fish collected for the tissue study, 12 herring (6 female and 6 male) were collected and muscle, liver and stomach were analyzed for Se in each fish individually using the same method used for the tissue study. In order to avoid combining pooled samples with individual samples when calculating the average *k*, we create a "pooled sample" by using the average concentration for the 12 individual herring.

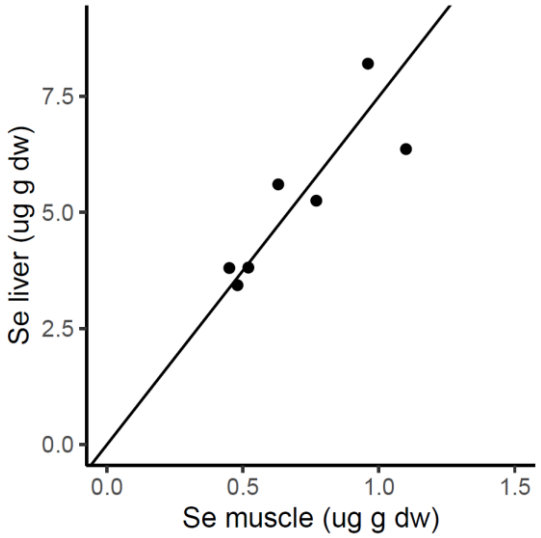



***Figure S 2**. Scatterplot of muscle and liver Se concentrations. The solid line represent the conversion factor (k) of 7.49.*

***Table S 3.*** *Data used for the calculation of Se discharge load from terrestrial sources.*

| River basin | Catchment area ($km^2$) | River discharge ($m^3 s^{-1}$) | Se ($\mu g L^{-1}$) | Se input ($Mg a^{-1}$) | Surface area ($km^2$) | Se input ($\mu g m^2 a^{-1}$) |
|---|---|---|---|---|---|---|
| **Bothnian Bay and Sea** | 481440 | 5792 | 0.16 | 29.3 | 115516 | 253 |
| Indalsälven | 25518 | 606 | 0.07 | 1.3 | | |
| Torne River | 39705 | 564 | 0.20 | 3.6 | | |
| Kemijoki | 51036 | 739 | 0.15 | 3.4 | | |
| Additional input | | 3883 | 0.17 | 20.4 | | |
| **Baltic Proper** | 574545 | 3670 | 0.55 | 63.9 | 211069 | 303 |
| Venta | 11624 | 70 | 0.36 | 0.8 | | |
| Nemunas | 92318 | 634 | 0.36 | 7.3 | | |
| Vistula | 193347 | 1344 | 0.63 | 26.7 | | |
| Pregola | 14783 | 86 | 0.44 | 1.2 | | |
| Oder | 117862 | 514 | 0.94 | 15.1 | | |
| Additional Sweden | | 571 | 0.30 | 5.3 | | |
| Additional Southern Baltic Coast | | 451 | 0.51 | 7.3 | | |
| **Kattegat (including Belt Sea)** | 86980 | 1555 | 0.37 | 18.3 | 42408 | 431 |
| Göta Älv | 48326 | 732 | 0.35 | 8.0 | | |
| Additional Kattegat | 31204 | 611 | 0.36 | 6.9 | | |
| Additional Belt Sea | 27365 | 212 | 0.50 | 3.3 | | |
| **Gulf of Finland** | 413100 | 3687 | 0.18 | 21.5 | 29600 | 727 |
| Neva River | 286553 | 2109 | 0.18 | 11.7 | | |
| Narva River | 56797 | 339 | 0.13 | 1.4 | | |
| Additional input | | 1239 | 0.22 | 8.5 | | |
| **Gulf of Riga** | 127840 | 1141 | 0.25 | 8.9 | 16330 | 543 |
| Gauja | 8652 | 67 | 0.23 | 0.5 | | |
| Daugava | 86052 | 656 | 0.28 | 5.8 | | |
| Lielupe | 17876 | 76 | 0.45 | 1.1 | | |
| Additional input | | 342 | 0.14 | 1.5 | | |
| **Baltic Sea** | 1605350 | 15845 | 0.28 | 142 | 415266 | 342 |

***Table S 4.*** *Water Se concentrations in lakes and streams between 1977 and 2012.*

| **Historic Se concentrations** | **year** | **ng $L^{-1}$** | **Reference** |
|---|---|---|---|
| Sweden 25 lakes | 1977-1978 | 103±42 | [*Remberger*, 1980] |
| Finland 1 lake | 1978 | 109 | [*Remberger*, 1980] |
| Sweden streams (n=2) | 1991 | 129/86 | [*Wang et al.*, 1994] |
| Finland headwater streams (n=207) | 1991-1992 | 73±27 | [*Wang et al.*, 1994] |
| Finland 7 lakes | 1992 | 82 | [*Alfthan and Aro*, 2005] |
| Sweden 3 lakes | 1997 | 51±18 | [*Hultberg*, 2002] |
| Finland 7 lakes | 1999 | 96 | [*Alfthan and Aro*, 2005] |
| Sweden 51 streams (filtered < 0.45µm) | 1998-2001 | 218±165 | [*Salminen et al.*, 2005] |
| Finland 65 streams (filtered < 0.45µm) | 1998-2001 | 176±73 | [*Salminen et al.*, 2005] |
| Denmark 5 streams (filtered < 0.45µm) | 1998-2001 | 554±185 | [*Salminen et al.*, 2005] |
| Germany 74 streams (filtered < 0.45µm) | 1998-2001 | 577±567 | [*Salminen et al.*, 2005] |
| Poland 56 streams (filtered < 0.45µm) | 1998-2001 | 713±365 | [*Salminen et al.*, 2005] |
| Estonia 11 streams (filtered < 0.45µm) | 1998-2001 | 143±60 | [*Salminen et al.*, 2005] |
| Latvia 7 streams (filtered < 0.45µm) | 1998-2001 | 260±92 | [*Salminen et al.*, 2005] |
| Lithuania 14 streams (filtered < 0.45µm) | 1998-2001 | 369±108 | [*Salminen et al.*, 2005] |
| Sweden 1 stream (n=3) | 2010-2012 | 95 | [*Lindell*, 2015] |

***Table S 5.*** *Tissue wet weights (g) and fish lengths (cm) for the study at Lilla Vartan [Danielsson et al., 2018]. Mean values and ranges in parenthesis.*

| **Tissue** | **Female samples** | **Male samples** |
|---|---|---|
| N | 25 | 25 |
| Muscle with skin (g) | 13 (12-16) | 12 (11-14) |
| Muscle (g) | 11 (9.4-12) | 10 (8.5-12) |
| Skin (g) | 1.2 (0.94-1.6) | 1.3 (1.1-1.7) |
| Stomach (g) | 2.5 (1.9-3.7) | 2.0 (1.3-3.0) |
| Gills (g) | 1.3 (1.1-1.8) | 1.2 (0.89-1.5) |
| Gonads (g) | 0.89 (0.29-1.8) | 2.1 (0.67-3.8) |
| Eyes (g) | 0.86 (0.59-1.1) | 0.88 (0.65-1.2) |
| Spine (g) | 0.14 (0.10-0.18) | 0.20 (0.12-0.30) |
| Liver (g) | 0.72 (0.47-0.86) | 0.55 (0.41-0.74) |
| Kidney (g) | 0.28 (0.15-0.36) | 0.27 (0.16-0.34) |
| Brain (g) | 0.18 (0.13-0.21) | 0.17 (0.13-0.22) |
| Carcass(g) | 5.5 (4.2-6.5) | 5.3 (4.4-6.6) |
| Body weight (g) | 40 (36-45) | 38 (35-44) |
| Total length (cm) | 18 (17-19) | 18 (17-19) |

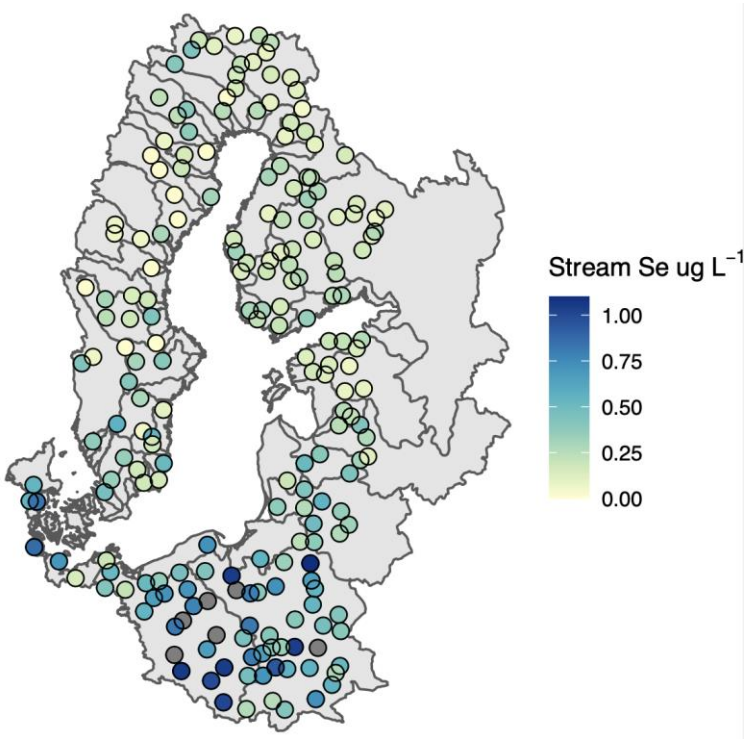
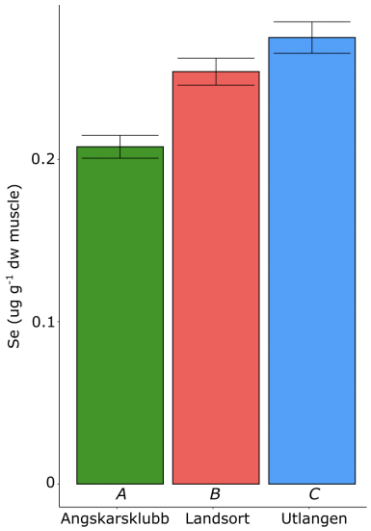



***Figure S 3.*** *Stream Se concentrations from 1998-2001 in countries around the Baltic Sea from Salminen et al. [2005]. Observations shown in grey are >1.1 µg L$^{-1}$. Also indicated in grey are the watershed border.*

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            Df  Sum Sq Mean Sq F value   Pr(>F)
SITE         2 0.05213 0.02606   25.50 8.30e-09 ***
YEAR         1 0.03484 0.03484   34.09 2.08e-07 ***
Residuals   62 0.06337 0.00102
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***Figure S 4.*** *A) Mean and standard deviation of the historic time series for Se in muscle, B) results from ANCOVA analysis indicating that there is a significant difference between the concentrations from the three sites in the retrospective study (Levene's test was not significant, which implies that the variance is homogenous).*

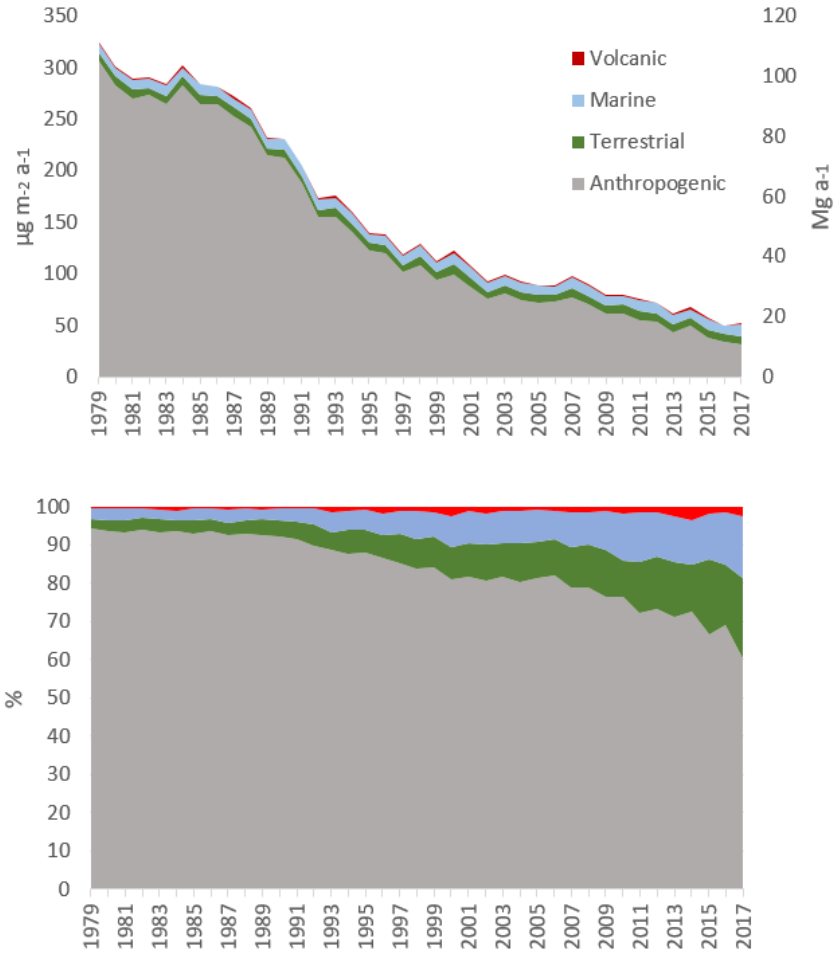


***Figure S 5.*** *A) Total modeled deposition to the Baltic Sea indicating the source of the Se, B) the contribution of each source as a function of the total deposition.*

***Table S 6.*** *Calculation of Se source load changes between 1980 and 2010 and herring muscle Se decrease. For the riverine slow decrease scenario a 40-50% decrease between 1980 and 2010 is used. To calculate the decrease in herring muscle Se the average of the three first and last year in the time series are used. Since the time series for Landsort is only covering 26 years it is not included here.*

| **Source** | **1980** | **2010** | **decrease** |
|---|---|---|---|
| | Mg $a^{-1}$ | Mg $a^{-1}$ | **%** |
| Deposition | 105 | 28 | 73 |
| Riverine | 142 | 142 | 0 |
| Total | 247 | 170 | 31 |
| | | | |
| Riverine slow decrease | 142 | 71-85 | 40-50 |
| Total slow decrease | 247 | 99-113 | 54-60 |
| | | | |
| | µg $g^{-1}$ ww | µg $g^{-1}$ ww | % |
| Bothnian Sea (Angskarsklubb) | 316 | 160 | 50 |
| Southern Baltic Proper (Utlangen) | 314 | 209 | 34 |

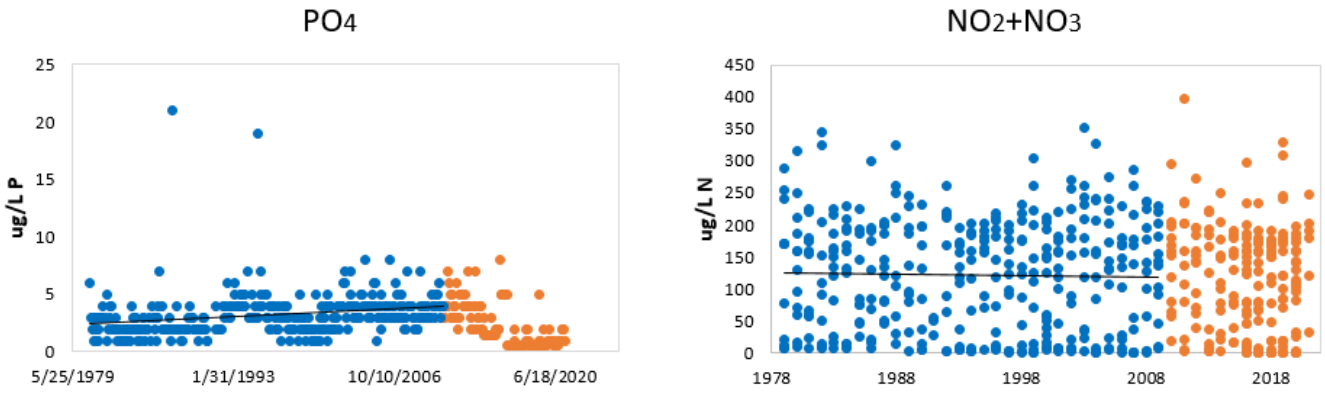


***Figure S 6.*** *Temporal change in phosphate ($PO_4$: phosphate) and nitrogen (NO2+NO3: >90% nitrate) concentrations in Dalarälven, the nearest river to the Angskarsklubb station. Similar to the overall estimate for the Bothnian Sea this river shows constant or increasing nutrient inputs during the 1979 to 2010 period (blue color), with possible decreases after 2010 (orange color). Data downloaded from https://miljodata.slu.se/MVM/.*

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