Long-term trends in cadmium and lead concentrations in *Mytilus edulis* from coastal European monitoring sites

https://github.com/slm119/ENV872_FinalProject

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1 Rationale and Research Questions

Heavy metal pollution is a global environmental and human health concern. Cadmium and lead heavy metals which continue to be a major public health concern (World Health Organization). Increasingly, research indicates that there may in fact be no "safe" level of cadmium or lead exposure, and so reducing levels of these neurotoxic, carcinogenic metals is a continuing battle for global health. Cadmium and lead are both naturally occurring in the environment, but their concentrations have increased due to human activity including fossil fuel combustion, industrial manufacturing, mining activities, among others. Monitoring the spatial and temporal distribution of these heavy metal pollutants is essential to tracking trends and identifying pollution hot spots.

Concentrations of heavy metals in the soft tissues of bivalves like *Mytilus edulis* (blue mussel) have been used for decades as a bioindicator for heavy metal pollution in marine environments because these bivalves are capable of bioaccumulating heavy metals from their environment (Phillips, 1977). *Mytilus edulis* are also harvested and farmed commercially for human consumption, and so monitoring heavy metals in their tissues is also important for managing potential human health risks from consumption. The International Council for the Exploration of the Sea (ICES) maintains an extensive database of contaminants in marine biota which includes sampling data for *Mytilus edulis* from several long-term sampling programs throughout Europe dating back to 1979. Median concentrations of cadmium and lead have decreased noticeable between 1979 and 1991 likely due to environmental regulations put in place during that time period such as the banned use of tetraethyl leaded gasoline; however, this decreased has slowed noticeably in recent decades (Fig 3).

For this analysis, I explored two questions concerning cadmium and lead concentrations in *Mytilus edulis* using the ICES monitoring data from 1990 to 2018.

1. How have concentrations of cadmium and lead in *Mytilus edulis* changed overall in the study region since the 1990s?
2. Do cadmium and lead concentrations in *Mytilus edulis* differ by country?

2 Dataset Information

The global coastline data used in the study area map is publicly available through the National Oceanographic and Atmospheric Administration's (NOAA) National Center for Environmental Information (NCEI) online data portal located here: https://www.ngdc.noaa.gov/mgg/shorelines/. The L1 resolution files were used from the Global Self-consistent Hierarchical High-resolution Geography (GSHHG) dataset. Geographic reference system is WGS84 (decimal degrees).

Data for cadmium and lead concentrations in *Mytilus edulis* were downloaded from the International Council for the Exploration of the Sea (ICES) DOME database on Feb 26, 2020 (available here: http://dome.ices.dk/views/ContaminantsBiota.aspx). This data portal holds a collection of marine related monitoring data sourced from several regional European monitoring groups including ICES, OSPAR, HELCOM, AMAP, and Expert Groups. Data for all metal and metalloid concentrations in biota were downloaded and then filtered to include only *Mytilus edulis* species and the specific heavy metals of interest for this study. The sampling data for *Mytilus edulis* was restricted to include only concentrations reported for the "whole soft body" of the bivalve and expressed in mass of metal per mass of the organism wet weight. There was no information provided to allow a conversion from dry weight to wet weight concentrations, and so dry weight records (a small minority of the data) were excluded from the analysis. Additionally, records flagged as having "suspect" data quality were excluded from the data set. Dataset variables are described in Table 1 below.

Table 1: Description and statistics for variables in ICES DOME monitoring data for cadmium and lead in *Mytilus edulis*

Variable name	Description	Statistics for Cd	Statistics for Pb
PARAM	Parameter	"Cd" – cadmium (6762 records)	"Pb" – lead (6709 records)
MYEAR	Monitoring year (may differ from the sampling year, NOT used in temporal trend analysis)	1990 - 2018	1990 - 2018
DATE	Sample date	February 6, 1990 to Feb 27, 2019	February 6, 1990 to Feb 27, 2019
Latitude and Longitude	units: decimal degrees		
Country	Country where measurement was reported		
Value.mgperkg	Concentration of contaminant in subsample. Units: mg metal/kg organism mass (wet weight of whole soft body).	range DL – 38.9; median 0.18; mean 0.32	range DL - 98.01; median 0.28; mean 0.01
NOINP	Number of individuals included in the subsample	range 1-703	range 1-703
DETLI.mgperkg	Reported detection limit of measurement equipment, units in mg/kg	range 0.000007 to 1; 1792 unreported	range 0.0001 - 0.6; 1736 unreported
QFLAG	Quality flag (see DOME metadata for full description of codes)	24 <; 1 D; 2 Q; 6735 NA	69 <; 0 D; 5 Q; 6635 NA

3 Exploratory Analysis

3.1 Geographic distribution of monitoring data

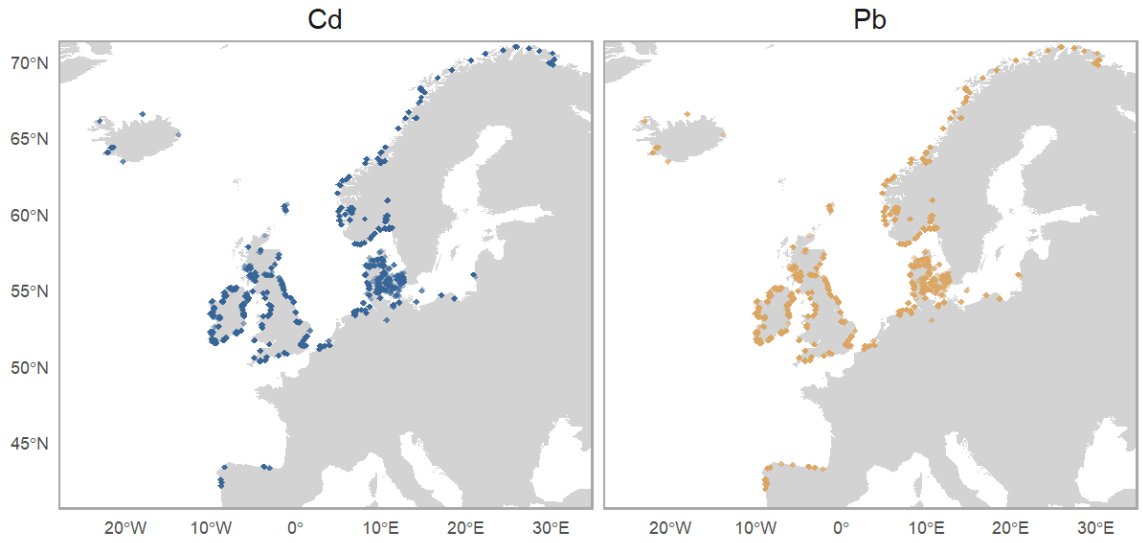

Figure 1: Monitoring locations of ICES DOME samples for cadmium and lead concentrations in *Mytilus edulis* from 1990 to 2018

Sample locations were highly clustered near European Atlantic and North Sea shorelines and were similar for cadmium and lead (Fig 1). The total number of samples during the study period varied significantly by country. For both metals, Norway alone accounted for about 43 percent of all the records (Fig 2). The top four countries with the most records (Norway, UK, Ireland, and Denmark) accounted for 90.9 percent of all records for cadmium and 89.3 percent of all records for lead. In general, sample numbers were similar between cadmium and lead because in many cases each *Mytilus edulis* sample was tested for both types of metals except Spain did not include many records for cadmium. Sampling records were sparse for coastlines in the Baltic Sea. Countries which did not include any sampling records in this database include France, Sweden, Finland, Estonia, Lithuania, and Poland.

3.2 Temporal distribution of monitoring data

Yearly median concentrations in *Mytilus edulis* for both cadmium and lead concentrations show a noticeable decrease between 1979 to about 1990 (Fig 3). After 1990, however, it is unclear whether there continues to be a decrease in heavy metal concentration; a monthly time series analysis is used to determine whether a there is a monotonic trend after 1990. Additionally, monitoring data was more sparse before 1990 and too much interpolation would be needed to include it in a trend analysis. For both metals, April to July were most likely to have missing sample data during the study period and September through November were the most compete months with no missing monthly data (Table 2).

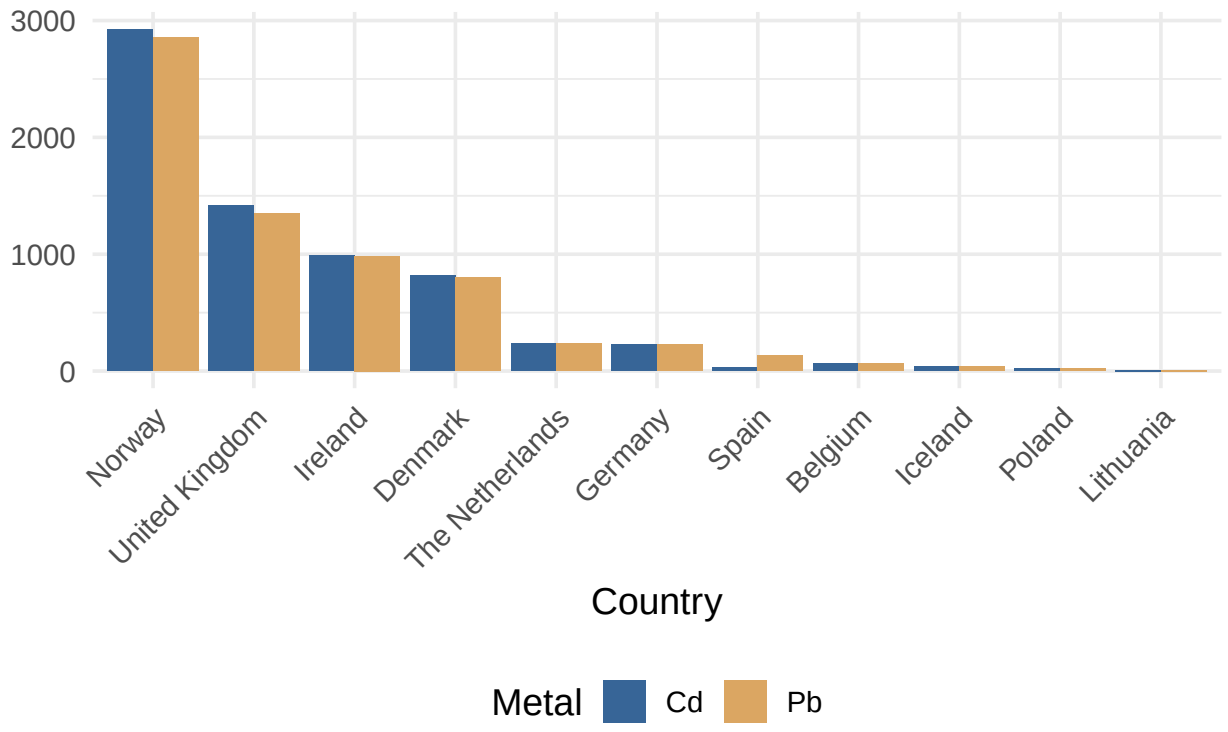

Figure 2: Number of cadmium and lead monitoring records for *Mytilus edulis* by country between 1990 and 2019.

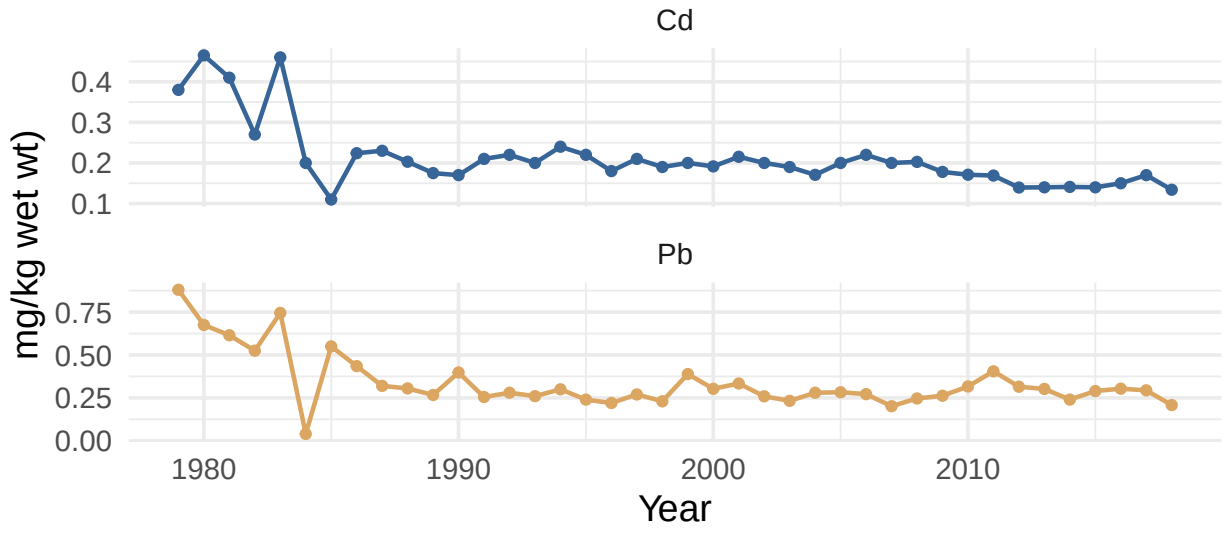

Figure 3: Yearly median concentrations of cadmium and lead in *Mytilus edulis* ICES monitoring data from 1979 to 2018. Notice the stark decrease from 1979 to the late 1980s and the leveling off after 1990.

Table 2: Summary of the sampling percentage for each month of the year from Feb 1990 to Jan 2019. September, October, and November were the only months with sampling data for all years.

Month	Cd	Pb
January	55%	59%
February	76%	72%
March	79%	79%
April	45%	45%
May	28%	28%
June	31%	31%
July	31%	28%
August	76%	76%
September	100%	100%
October	100%	100%
November	100%	100%
December	76%	76%

3.3 Distribution of cadmium and lead concentrations

Concentrations for cadmium ranged from below the instrument detection limit to 98.1 mg/kg with a median of 0.28 mg/kg and mean of 0.81 mg/kg. Lead concentrations ranged from below the detection limit to 38.9 mg/kg with a median of 0.18 mg/kg and mean of 0.32 mg/kg. Both metals exhibited a strong positive skew and so are displayed on a log scale in most figures. Detection limits for measurements instruments ranged substantially for both metal samples (Table 1), and many samples concentrations were within the range of detection limits for other samples (Fig 5).

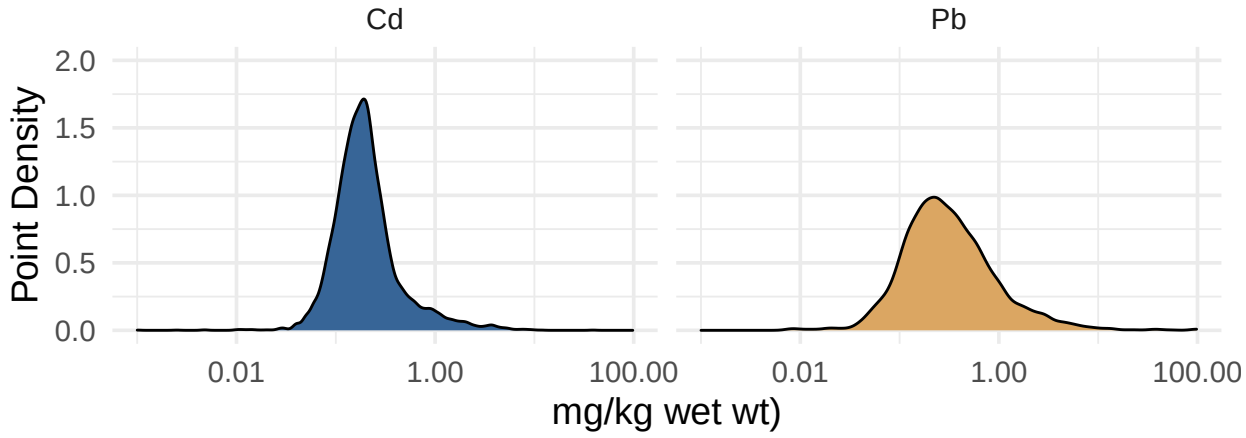

Figure 4: Distribution of sample concentrations of cadmium and lead in *Mytilus edulis* from ICES data 1990 to 2019.

4 Analysis

4.1 Question 1: How have cadmium and lead concentrations in *Mytilus edulis* changed over time?

Data for lead and cadmium concentrations were aggregated using the monthly median for each month between February 1990 to January 2019. Months with no sample data were interpolated using a linear interpolation method (Fig 6).

Cadmium concentrations in *Mytilus edulis* have decreased significantly over the study period by an estimated 0.002 mg/kg from 1990 to 2019 (Seasonal Mann-Kendall; S = -1044; p <0.0001, Seasonal Sen's Slope). Cadmium concentrations also exhibited a seasonal variation where concentrations in November, December, and January exhibited significantly different decreasing trend than other months (Seasonal Mann-Kendall, $p < 0.05$). Lead showed no significant monotonic trend over the study period (Seasonal Mann-Kendall; S = -284; p = 0.125).

4.2 Question 2: Do cadmium and lead concentrations in *Mytilus edulis* differ by country?

Comparison of concentrations between countries was restricted to only countries with more than 100 records over the study period. A non-parametric group-wise comparison was used to compare concentration distributions by country.

Analyses for cadmium revealed that concentrations distributions differed significantly between all countries except Germany and the UK which were statistically similar (Fig 7; Kruskal-Wallis Rank Sum Test, chi-squared = 897.8, df = 5, $p < 0.0001$; Dunn's Test with Benjamin-Hochberg (1995) adjustment, $p < 0.05$ for all comparisons except Germany and UK). The Netherlands had the highest distribution of cadmium concentrations with a median of 0.41 mg/kg (see Table 3).

Lead concentration distributions between countries also differed significantly by country (Fig 7; Kruskal-Wallis Rank Sum Test, chi-squared = 1302.8, df = 6, $p < 0.0001$; Dunn's Test with Benjamin-Hochberg (1995) adjustment, alpha = 0.05). The Netherlands, Spain, and UK had the highest distribution of lead concentrations and were statistically to each other. Next highest lead concentrations were in Norway, then Germany and Ireland (not significantly different from each other), and then the lowest lead concentrations were in Denmark (Dunn's Test, $p < 0.05$).

Table 3: Summary of median concentrations of cadmium and lead in *Mytilus edulis* by country during the study period from 1990 – 2019.

Country	Median Cd conc (mg/kg)	Median Pb conc (mg/kg)
The Netherlands	0.41	0.55
Spain	NA	0.49
Norway	0.21	0.25

Country	Median Cd conc (mg/kg)	Median Pb conc (mg/kg)
Germany	0.2	0.24
United Kingdom	0.191	0.54
Denmark	0.164	0.15
Ireland	0.11	0.22

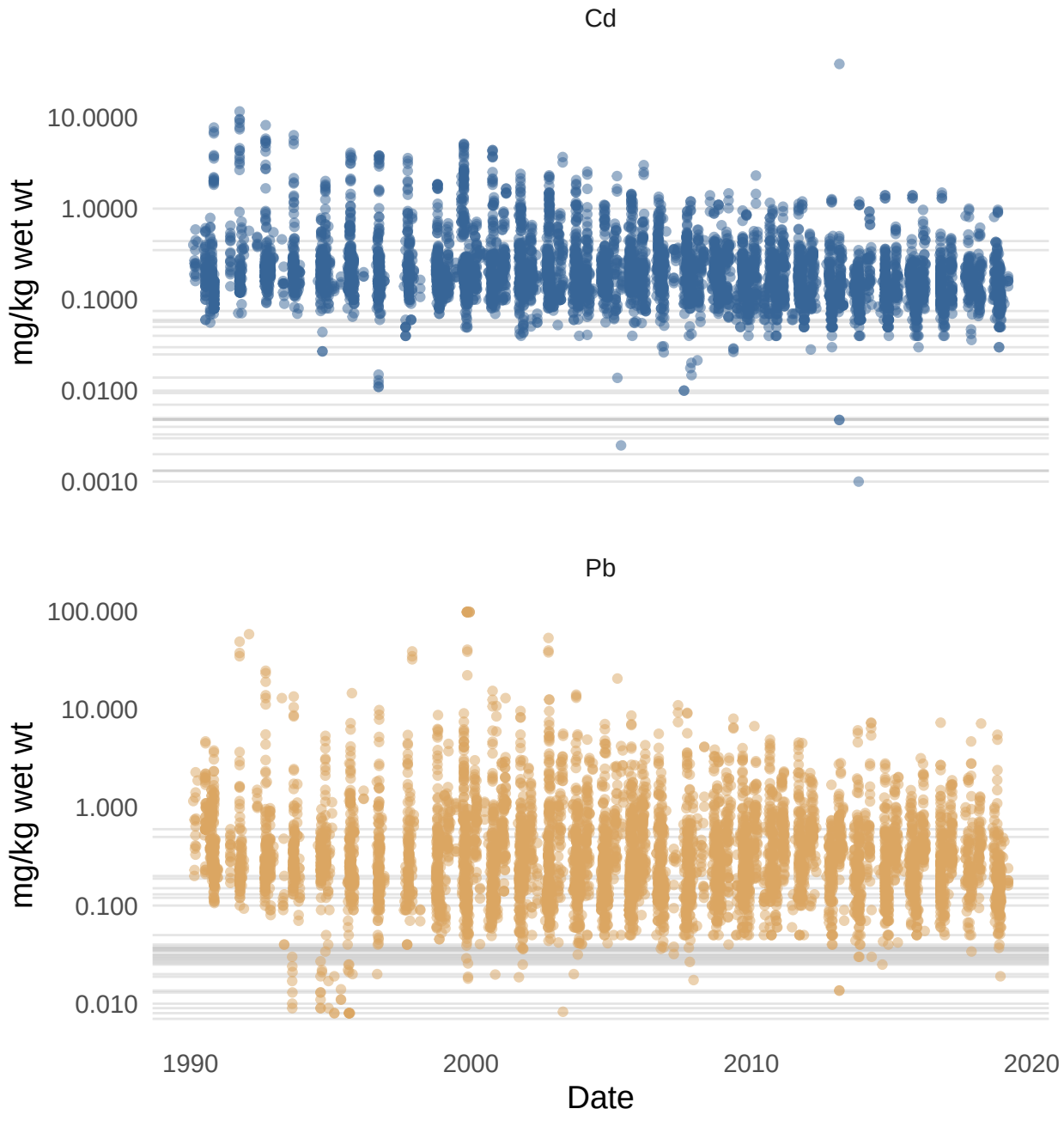

Figure 5: Cadmium and lead sample concentrations over time for ICES *Mytilus edulis* monitoring. Grey lines show the various instrument detection limits reported in each the data set.

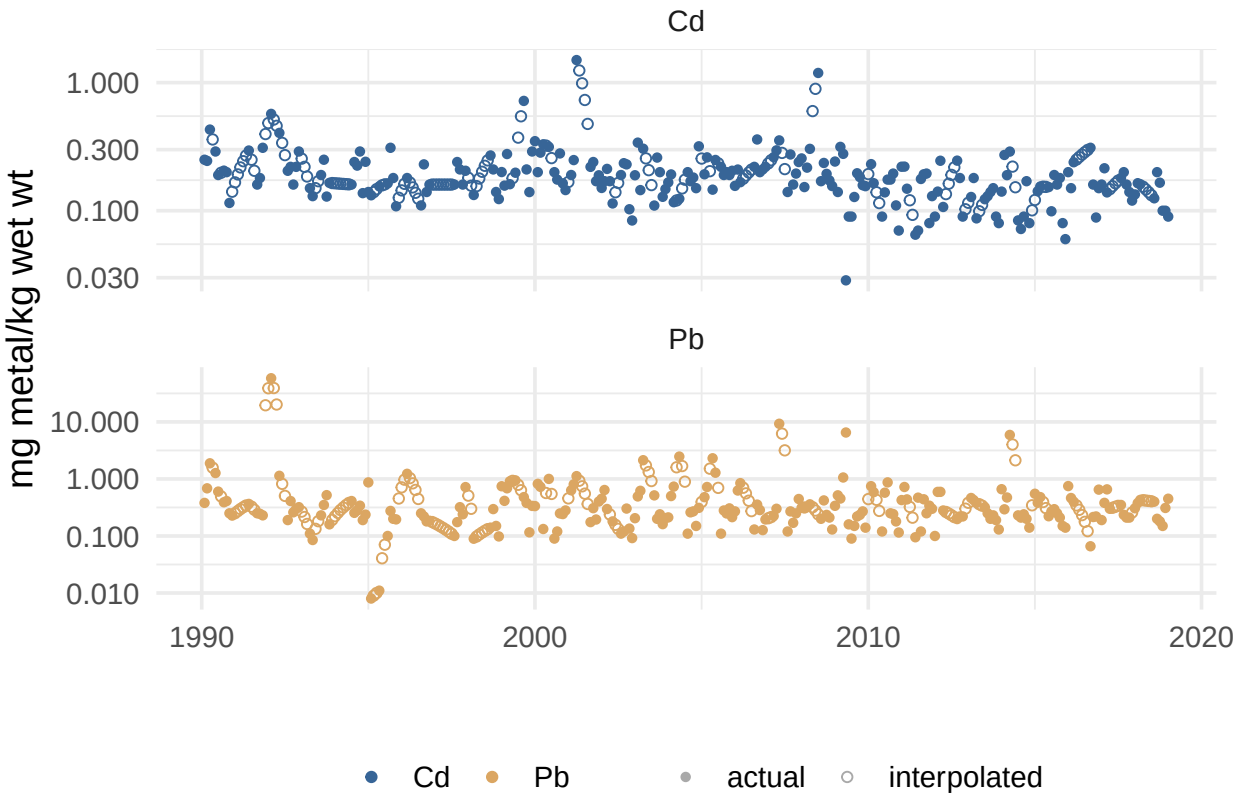

Figure 6: Concentrations of cadmium and lead in *Mytilus edulis* in ICES study region from 1990 to 2018.

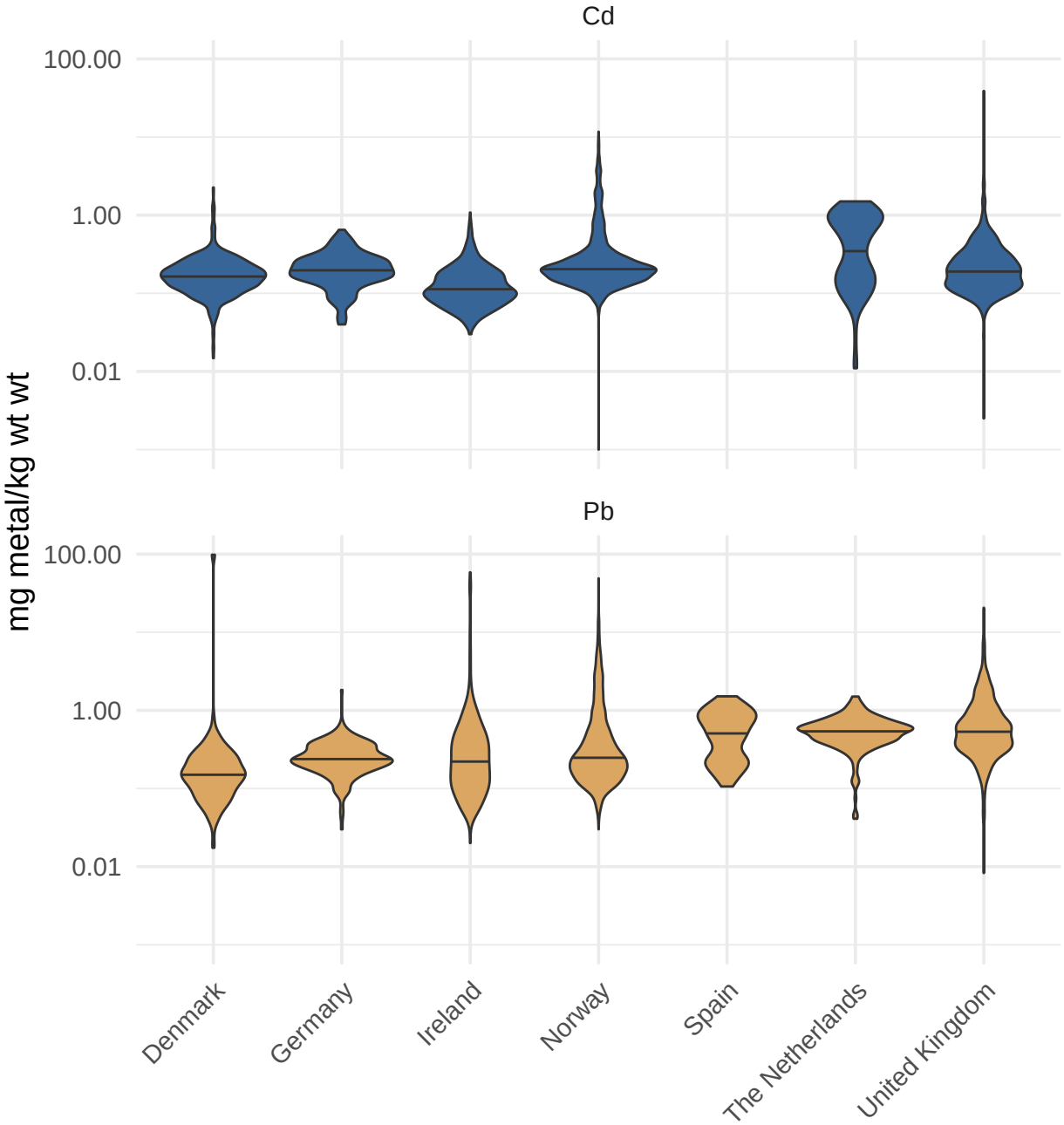

Figure 7: Distribution of cadmium and lead concentrations in *Mytilus edulis* for countries with more than 100 samples between 1990 and 2018.

5 Summary and Conclusions

6 References

Phillips, D. J. H. (1977). The use of biological indicator organisms to monitor trace metal pollution in marine and estuarine environments—A review. Environmental Pollution (1970), 13(4), 281–317. https://doi.org/10.1016/0013-9327(77)90047-7

WHO | Cadmium. (n.d.). WHO; World Health Organization. Retrieved April 20, 2020, from http://www.who.int/ipcs/assessment/public_health/cadmium/en/

WHO | Lead. (n.d.). WHO; World Health Organization. Retrieved April 17, 2020, from http://www.who.int/ipcs/assessment/public_health/lead/en/