Geographic and temporal trends in cadmium and lead concentrations in *Mytilus edulis* (blue mussel) along the Atlantic coast of Europe

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 (Tchounwou et al., 2012; WHO|Cadmium n.d.; WHO|Lead, n.d.)). Increasingly, research indicates that there may be no “safe” level of heavy metal exposure for humans, and so reducing levels of these neurotoxic, carcinogenic metals is a continuing battle for global human health. Heavy metals are also toxic to other organisms and have been associated with significant decreases biodiversity in natural systems (Johnston and Roberts, 2009). Cadmium and lead are two heavy metals which are naturally occurring in the environment but concentrations have increased due to human activities including fossil fuel combustion, industrial manufacturing, mining activities, and others (Tchounwou et al., 2012). Therefore, monitoring the spatial and temporal distribution of these heavy metal pollutants is essential for identifying pollution hot spots, avoiding the consumption of contaminated foods (like marine mussels), and assessing the rate of athropogenic heavy metal loading in natural systems.

Mytilus edulis (blue mussel) are marine bivalves which are capable of bioaccumulating heavy metals in their soft tissues. These organisms have been used for decades as a bioindicator for heavy metal pollution in marine environments (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. 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 monitoring programs throughout Atlantic 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 such as the banned use of tetraethyl leaded gasoline (Tchounwou et al., 2012); however, this decrease 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: Variable descriptions and statistics for ICES DOME monitoring data

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

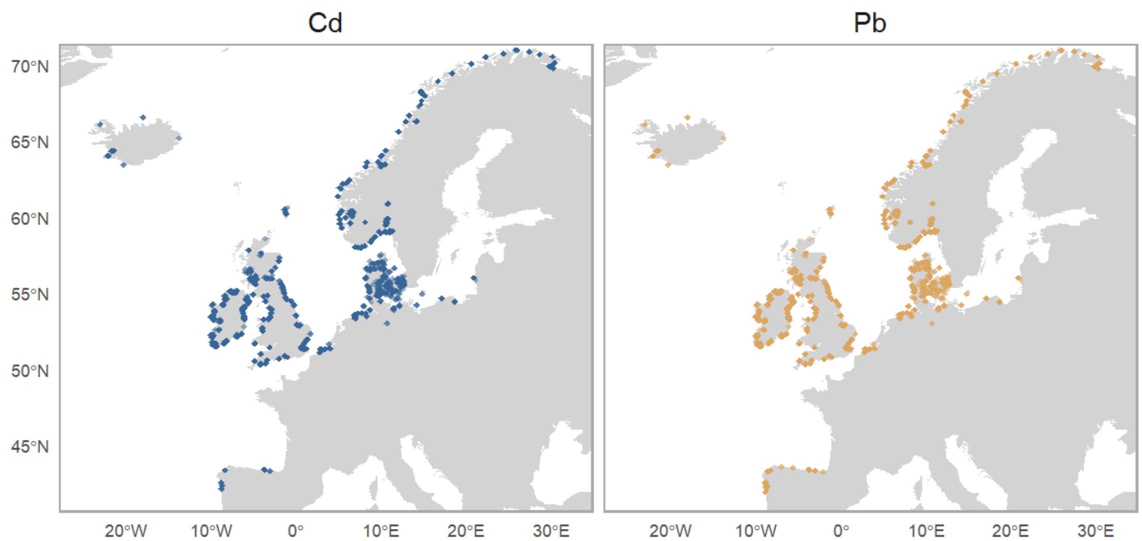

Figure 1: Monitoring locations of ICES DOME database records for cadmium and lead concentrations *Mytilus edulis* from 1990 to 2018

Sample locations are highly clustered near European Atlantic and North Sea shorelines (Fig 1). Geographic spread of the sample sites are similar for cadmium and lead (Fig 1). The total number of samples during the study period varied significantly by country. For both metals, Norway 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 (Fig 2). 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. Other coastal European nations including France, Sweden, Finland, Estonia, Lithuania, and Poland did not have any sampling records in this database.

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, the temporal trend is less obvious; a monthly time series analysis was used to determine whether a there is a monotonic trend after 1990. For both metals, April, May, June, and July were most likely to have missing sample data during the study period and September through November were the most compete months with at least one record for every year in the dataset (Table 2).

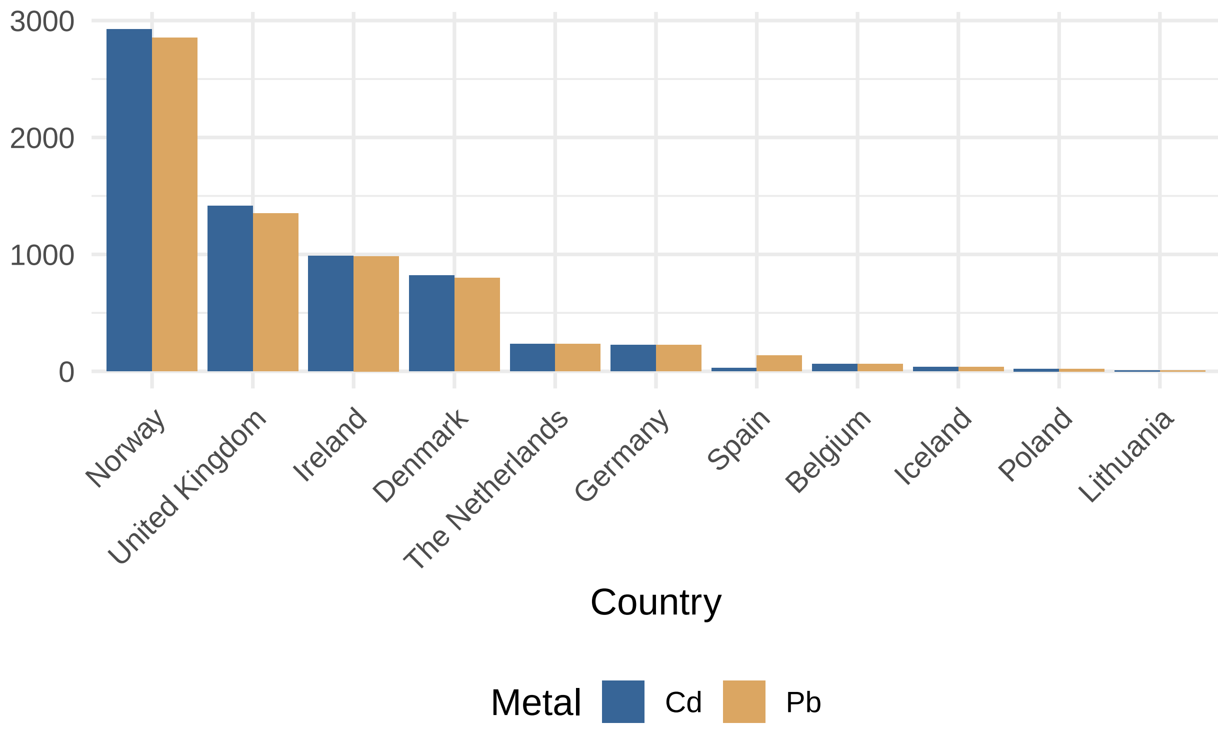

Figure 2: Number of *Mytilus edulis* cadmium and lead monitoring records by country between 1990 and 2019.

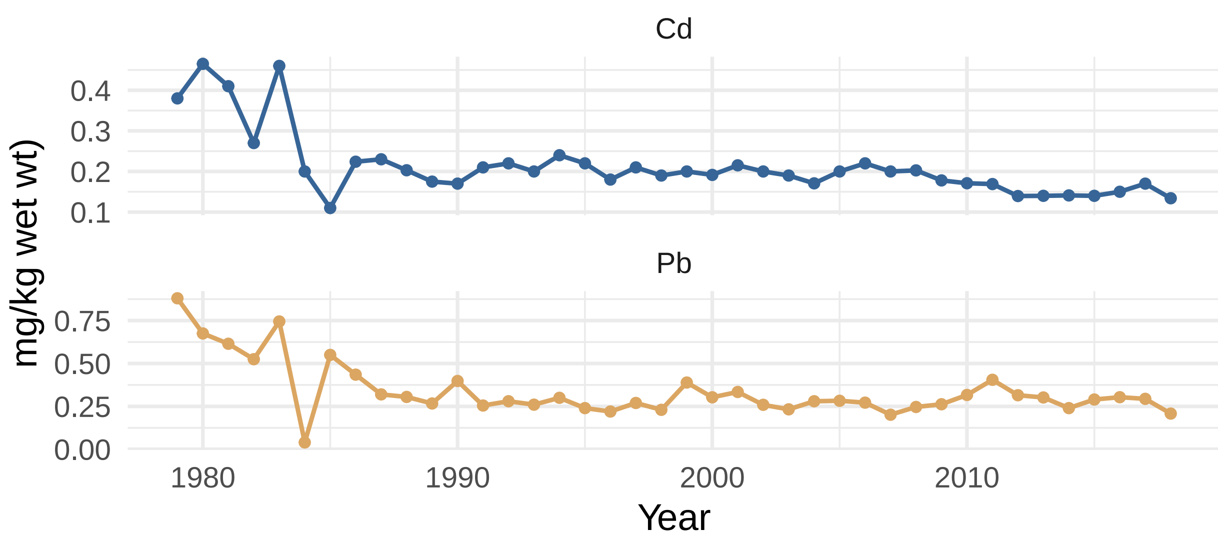

Figure 3: Yearly median concentrations of cadmium and lead in *Mytilus edulis* ICES monitoring data from 1979 to 2018. Notice the stark decrease from 1979 to the late 1980s and the leveling off after 1990.

Table 2: Sampling percentage for each month during the analysis study period 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 measurement 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). This results in considerable left censoring of the data which can affect interpretation of statistical analyses.

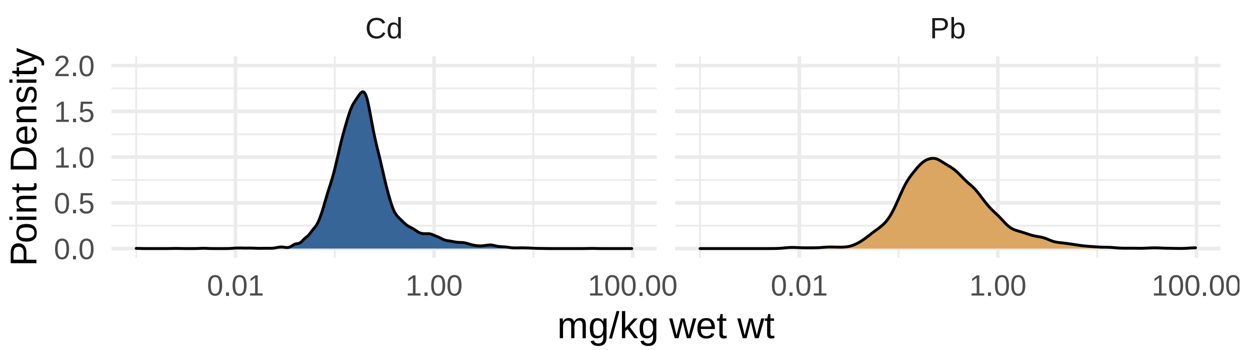

Figure 4: Distribution of cadmium and lead concentrations in *Mytilus edulis* sample records from 1990 to 2019.

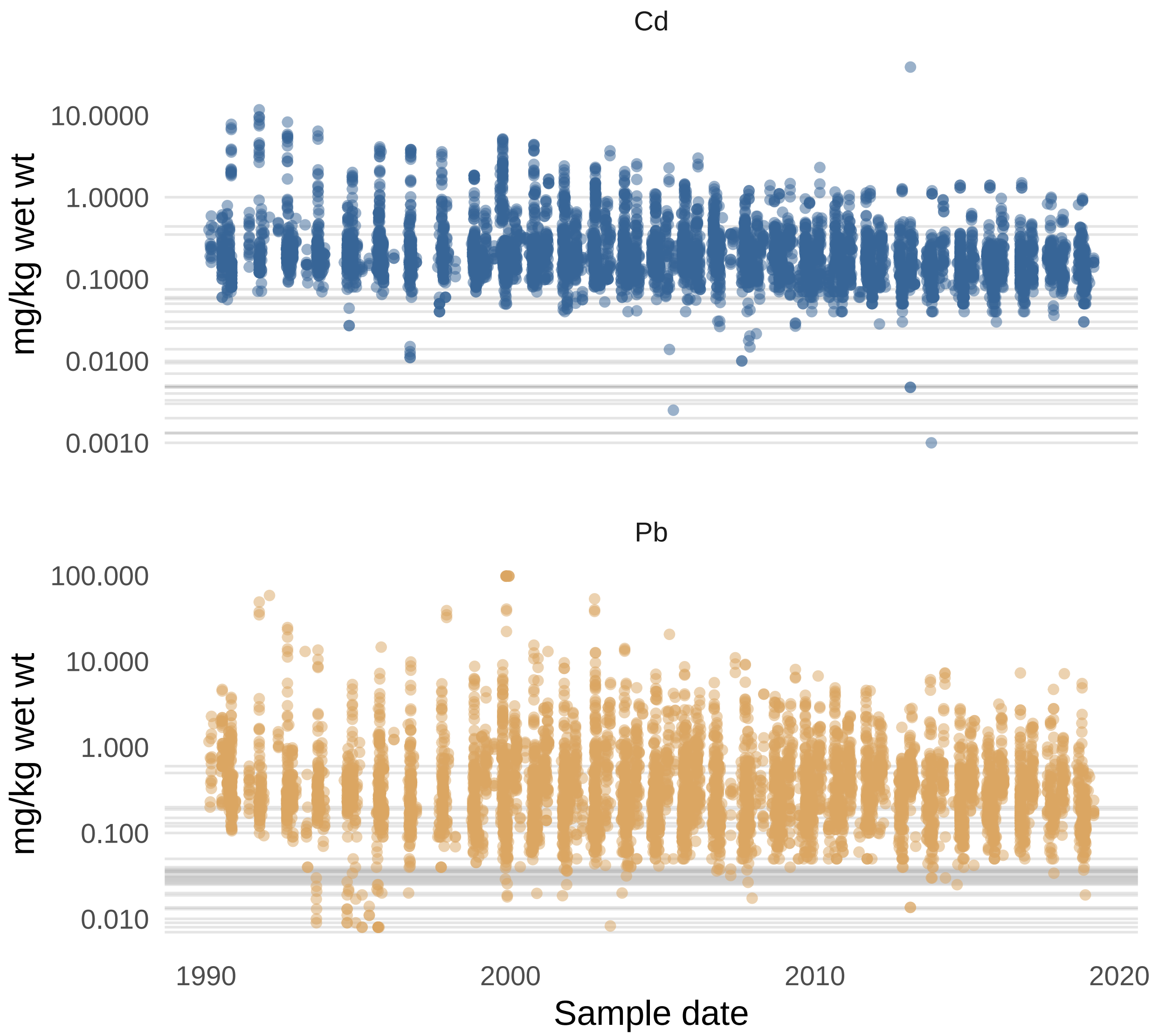

Figure 5: Concentrations of cadmium and lead in ICES *Mytilus edulis* monitoring samples for all sample dates from Feb 1990 to Jan 2019. Grey lines indicate the various instrument detection limits reported in each the data set.

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 based on the monthly median for each month between February 1990 to January 2019. Months with no sample data were interpolated using a linear interpolation (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 some seasonal variability; higher decreasing trends were seen in November, December, and January as compared to other months of the year (Seasonal Sen;s Slope, $p < 0.05$). Lead showed no significant monotonic trend over the study period (Seasonal Mann-Kendall; S = -284; $p = 0.125$).

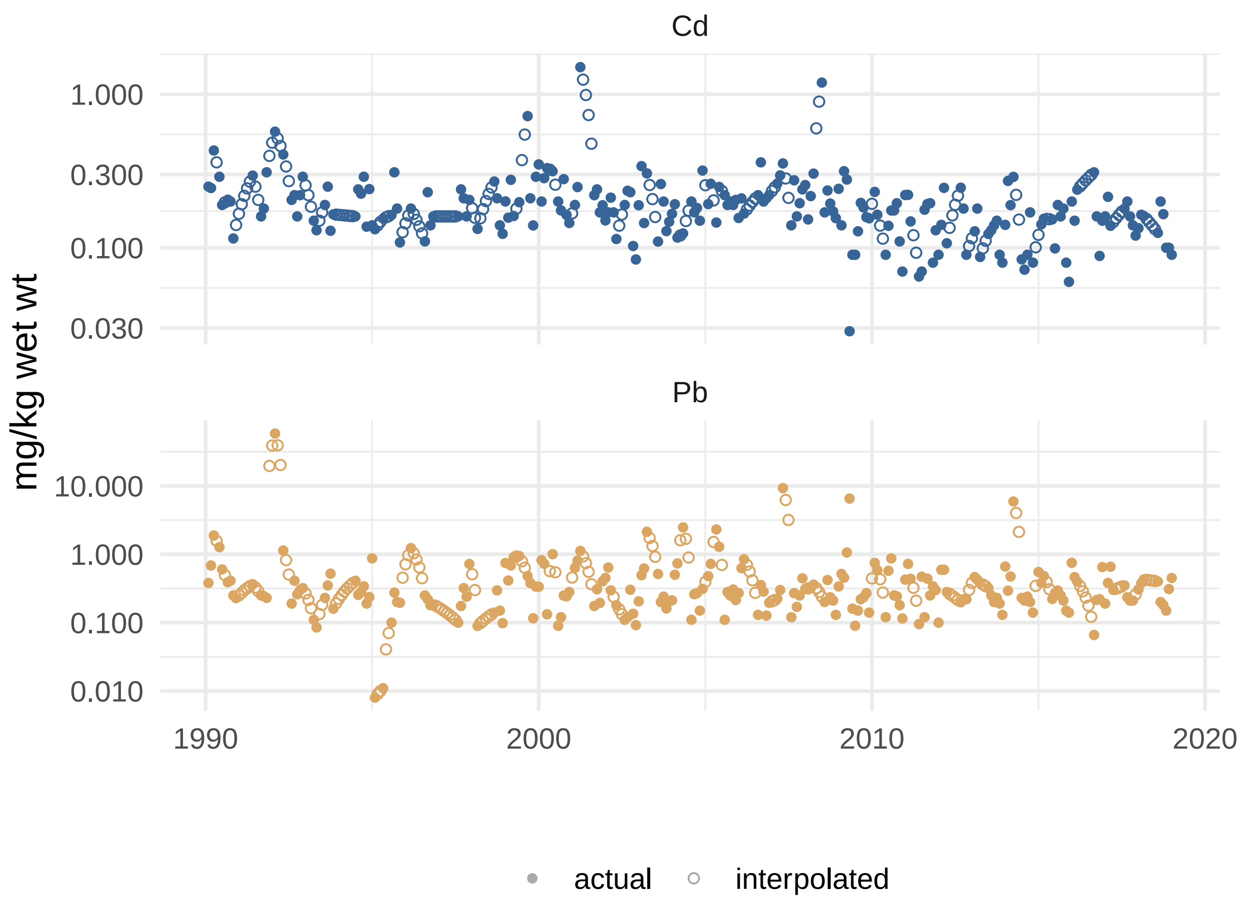

Figure 6: Monthly median cadmium and lead concentrations in *Mytilus edulis* from 1990 to 2018. Points symbolized with open circles are linearly interpolated.

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. Due to the non-normal, heterogenous variance, and unequal sample size of the data, a non-parametric group-wise comparison was used to compare concentration distributions by country.

Analyses for cadmium revealed that concentration 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).

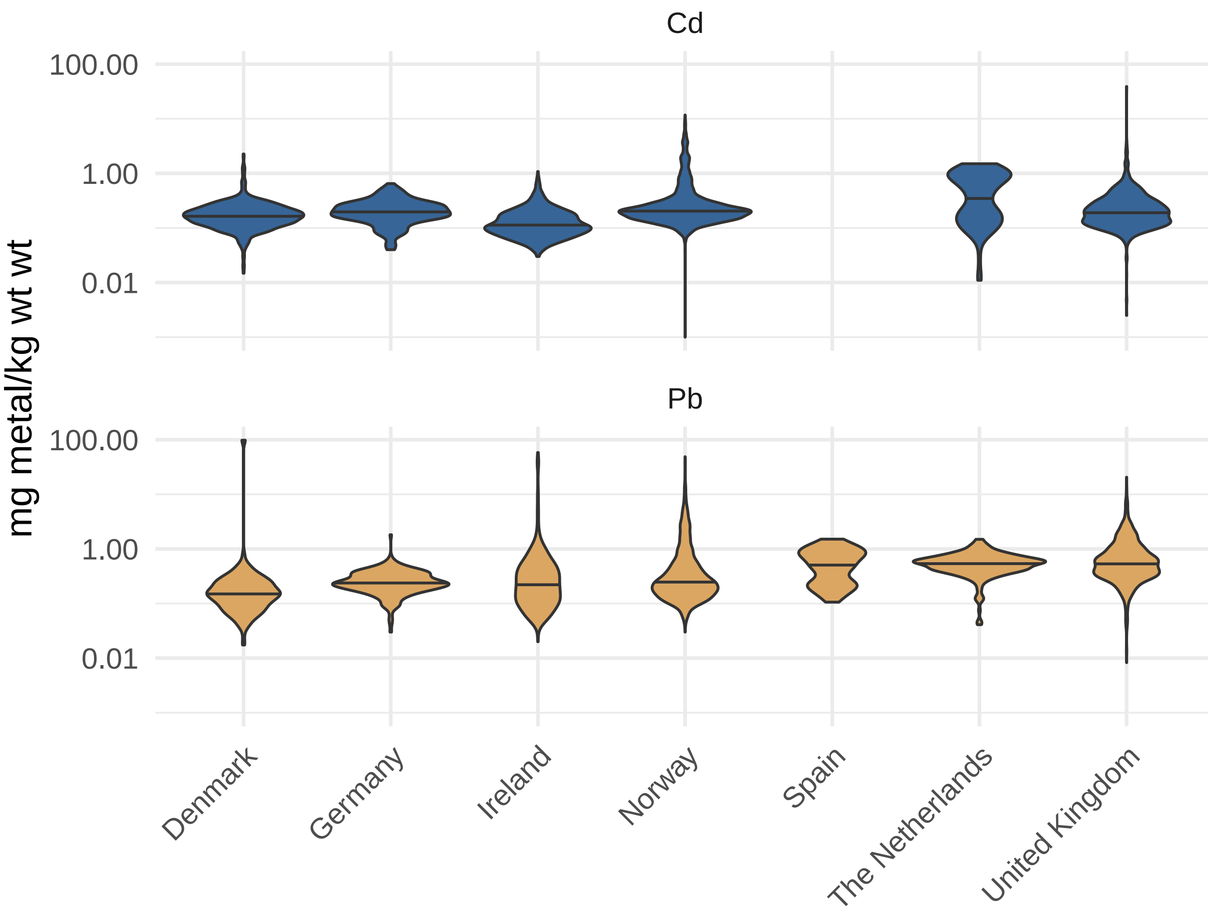

Figure 7: Distribution of cadmium and lead concentrations in *Mytilus edulis* for countries with more than 100 samples between 1990 and 2018.

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, $p < 0.05$). The Netherlands, Spain, and UK had the highest distribution of lead concentrations and were statistically similar to each other. Next highest lead concentrations were in Norway, then Germany and Ireland (not significantly different from each other, but were different from all other groups), and then the lowest lead concentrations were in Denmark (Dunn's Test, alpha = 0.05).

Table 3: 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	-	0.49
Norway	0.21	0.25
Germany	0.20	0.24
United Kingdom	0.19	0.54
Denmark	0.16	0.15
Ireland	0.11	0.22

5 Summary and Conclusions

Long term monitoring efforts for *Mytilus edulis* allow researchers to gauge our progress in reducing heavy metal contaminants in marine ecosystems. However, it is important to consider the limitations of using data which was collected over many years and from numerous testing programs. One of the limitations in this investigation is the wide array of sampling methods, analysis methods, and varying instrument detection limits used in the data collection process. Some of the analytical equipment used for monitoring efforts were not able to detect metal concentrations below 0.6 or even 1 mg/kg, while other testing facilities recorded metal concentrations well below these detection limits at 0.01 mg/kg or less. A high number of left-censored records could lead to an under or over-estimate of the true trends in metal concentrations over time. Therefore, it would beneficial to update metal testing procedures to use more sensitive tests that are able to precisely measure environmentally-relevant concentrations of cadmium and lead in marine organisms.

This analysis of heavy metals trends in the European Atlantic coastal region suggest that, as a whole, concentrations of cadmium have decreased by a small but significantly amount since 1990. Current median concentration of cadmium in *Mytilus eduils* remains well below the current United Nations Food and Agricultural Organization's (UN FAO) suggested maximum level of 2 mg/kg in marine bivalves for human consumption, but efforts are still needed to reduce cadmium concentrations to background levels (FAO, 2019). Lead concentrations, however, were not found to have decreased significantly over the same study period, and several countries were found to have much higher lead loads than others.

Cadmium and lead concentrations in *Mytilus edulis* were shown to vary regionally. The Netherlands was consistently shown to have the highest concentrations of both heavy metals. Spain and the UK also had significantly higher levels of lead bioaccumulation in their monitoring samples than the other countries included in this analysis. These results suggest that management efforts at the regional or national level may have direct implications for heavy metal loading in coastal environments. Future analysis should be done to identify identify major sources of cadmium and lead and identify effective management and policy efforts that can help reduce heavy metal releases to coastal environments.

6 References

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