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University of Turku

Faculty of Science
Department of Biology
Biology
Doctoral Programme in Biology, Geography and Geology (BGG)

Supervised by

Professor Veijo Jormalainen University of Turku Finland	Dr. Igor Eulaers Norwegian Polar Institute Norway

Reviewed by

Dr. Anne Sørensen Naturhistoriska riksmuseet Sweden	Professor Peter Haglund Umeå University Sweden

Opponent

Professor Jussi Kukkonen
University of Eastern Finland
Finland

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ABSTRACT

Anthropogenic contamination is a wide-spread environmental problem. In addition to persistent, bioaccumulative and toxic legacy contaminants, emerging contaminants with unknown environmental behaviour are causing concern. Many contaminants biomagnify in food webs, meaning that highest concentrations of contaminants are found in the apex species. Trophic dynamics of contaminants may differ depending on the properties of the contaminant, environment, and species composing a particular food web. In this thesis, I examine the current state of metal and organohalogen contamination in the Finnish Baltic coastal food webs, focussing on three avian sentinel species: the white-tailed eagle (*Haliaeetus albicilla*), the great cormorant (*Phalacrocorax carbo*), and the common eider (*Somateria mollissima*). In Chapter I, I investigate whether nestlings of white-tailed eagles and great cormorants act as sentinels for metal contamination associated with acidic sulphate soils. Compared to the control areas, I find higher concentrations of aluminium and cobalt in white-tailed eagles and of copper and manganese in great cormorants from the acidic sulphate soil areas. These results indicate that acidic sulphate soils are a source of certain metals for white-tailed eagle and great cormorant nestlings; there are, however, differences in metal exposure between these species. In Chapter II, I study the trophic transfer of mercury (Hg) between the benthic and pelagic food chains of the Archipelago Sea food web. The rate of trophic magnification of Hg was higher in the pelagic food chain than in the benthic food chain when the homeotherm birds were excluded from statistical models. In Chapter III, I investigate organohalogen contamination in the Archipelago Sea food web. I find that several organohalogen compounds biomagnify in the food web. Concentrations were generally highest in bird species, particularly the white-tailed eagle. As with the Hg models, statistical fit of the trophic magnification models of organohalogen compounds increased when removing birds from the data. In addition to the legacy contaminants, I also found emerging contaminant perfluoroethylcyclohexane sulfonate in the white-tailed eagle, great cormorant, and two fish species. This thesis demonstrates that different ecological and ecophysiological traits of the species play a role in the contaminant exposure and food web dynamics of the contaminants.

KEYWORDS: contaminantion, metals, organohalogen contaminants, food web, benthic, pelagic, stable isotopes, trophic dynamics, biomagnification

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TIIVISTELMÄ

Ympäristön kemiallinen saastuminen on vakava maailmanlaajuinen ongelma. Pysyvien, eliöihin kertyvien ja myrkyllisten, laajalle levinneiden haitta-aineiden lisäksi huolta aiheuttavat uudet haitta-aineet, joiden käyttäytymistä ympäristössä ei vielä tunneta. Monet haitta-aineet rikastuvat ravintoverkossa, jolloin korkeimmat pitoisuudet löytyvät ravintoverkon huipulla olevista lajeista. Haitta-aineiden käyttäytyminen ravintoverkossa riippuu aineen kemiallisista ominaisuuksista sekä ympäristön ja kyseisen ravintoverkon koostavien lajien piirteistä. Tässä väitöskirjassa tutkin metallien ja orgaanisten halogeeniyhdisteiden aiheuttamaa saastumista Suomen rannikon ravintoverkoissa keskittyen kolmeen ilmentäjälajiin: merikotkaan (*Haliaeetus albicilla*), merimetsoon (*Phalacrocorax carbo*) ja haahkaan (*Somateria mollissima*). Luvussa I tutkin ilmentävätkö merikotkan ja merimetson pesäpoikaset happamista sulfaattimaista peräisin olevia metallipäästöjä. Merikotkan poikasissa alumiinin ja koboltin pitoisuudet, ja merimetson poikasissa kuparin ja mangaanin pitoisuudet olivat koholla happamien sulfaattimaiden läheisyydessä syntyneissä ja kasvaneissa poikasissa verrattuna kontrollialueiden poikasiin. Tulokset osoittavat, että happamat sulfaattimaat ovat joidenkin metallien lähde merikotkan ja merimetson poikasille. Metalleille altistumisessa oli kuitenkin eroa lajien välillä. Luvussa II tutkin elohopean kulkeutumista Saaristomeren ravintoverkon benttisessä ja pelagisessa ravintoketjussa. Elohopea rikastui nopeammin trofiatasoa kohden pelagisessa ravintoketjussa verrattuna benttiseen ravintoketjuun, kun tasalämpöisiä lintuja ei sisällytetty mukaan rikastumismalleihin. Luvussa III tutkin orgaanisia halogeeniyhdisteitä Saaristomeren ravintoverkossa. Useat tutkituista aineista rikastuivat ravintoverkossa. Useimpien aineiden pitoisuudet olivat korkeimmat linnuissa, etenkin merikotkassa. Samoin kuin elohopean rikastumismalleissa, halogeeniyhdisteiden rikastumismallien selitysaste parani, kun lintuja ei sisällytetty malleihin. Tavanomaisten, Itämeren ympäristössä jo vuosia esiintyneiden haitta-aineiden lisäksi löysin perfluorietyylisykloheksaani sulfonaattia merikotkista, merimetsoista ja kahdesta kalalajista. Väitöskirja korostaa eri ekologisten ja ekofysiologisten ominaisuuksien roolia lajien haitta-ainealtistuksessa ja haitta-aineiden käyttäytymisessä ravintoverkoissa.

ASIASANAT: haitta-aineet, metallit, orgaaniset halogeeniyhdisteet, ravintoverkko, benttinen, pelaginen, stabiilit isotoopit, rikastuminen

Table of Contents

Abbreviations

Al	aluminium
BMF	biomagnification factor
Cd	cadmium
CHB	chlorinated bornane
CHL	chlordane
Co	cobalt
Cr	chromium
Cu	copper
DDE	dichlorodiphenyldichloroethylene
DDT	dichlorodiphenyltrichloroethane
dw	dry weight
GLMM	general linear mixed model
HCB	hexachlorobenzene
HCH	hexachlorocyclohexane
Hg	mercury
Li	lithium
lw	lipid weight
MeHg	methyl mercury
Mn	manganese
N	nitrogen
Ni	nickel
OCP	organochlorine pesticide
PBDE	polybrominated diphenyl ether
PCB	polychlorinated biphenyl
PFAS	per- and polyfluoroalkyl substances
PFBS	perfluorobutanesulfonic acid
PFCAs	perfluoroalkyl carboxylic acids
PFDA	perfluorodecanoic acid
PFDoA	perfluorododecanoic acid
PFDS	perfluorodecanesulfonic acid
PFECHS	perfluoroethylcyclohexane sulfonate

PFHpA	perfluoroheptanoic acid
PFHxA	perfluorohexanoic acid
PFHpS	perfluoroheptanesulfonic acid
PFHxS	perfluorohexanesulfonic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctanesulfonic acid
PFOSA	perfluorooctanesulfonamide
PFNA	perfluorononanoic acid
PFSAs	perfluoroalkyl sulfonic acids
PFTeA	perfluorotetradecanoic acid
PFTrA	perfluorotridecanoic acid
PFUnA	perfluoroundecanoic acid
POPs	persistent organic pollutants
psu	practical salinity unit
TEF	trophic enrichment factor
THg	total mercury
Tl	thallium
TP	trophic position
TMF	trophic magnification factor
ww	wet weight
Zn	zinc
6:2 Cl-PFAES	6:2 chlorinated polyfluorinated ether sulfonate
8:2 Cl-PFAES	8:2 chlorinated polyfluorinated ether sulfonate

List of Original Publications

This dissertation is based on the following original publications, which are referred to in the text by their Roman numerals:

I Vainio, R. K., Eulaers, I., Laaksonen, T., Vasko, V. & Jormalainen, V. (2020) White-tailed eagle (*Haliaeetus albicilla*) and great cormorant (*Phalacrocorax carbo*) nestlings as spatial sentinels of Baltic acidic sulphate soil associated metal contamination. *Science of the Total Environment* 718: 137424

II Vainio, R. K., Jormalainen, V., Dietz, R., Laaksonen, T., Schulz, R., Sonne, C., Søndergaard, J., Zubrod, J. P. & Eulaers, I. (2022) Trophic dynamics of mercury in the Baltic Archipelago Sea food web: Impact of Ecological and ecophysiological traits. *Environmental Science and Technology* 56: 11440-11448.

III Vainio, R. K., Vorkamp, K., Bossi, R., Jormalainen, V., Dietz, R., Laaksonen, T., Schulz, R., Sonne, C., Zubrod, J. P. & Eulaers, I. Legacy and emerging contaminants in the Baltic Archipelago Sea food web: concentrations, profiles, and biomagnification. Unpublished manuscript.

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1 Introduction

Anthropogenic chemical contamination is one of the most severe environmental problems, affecting both aquatic and terrestrial environments across the globe (McCrink-Goode, 2014). Many contaminants can be transported long distances from the original source of contamination: contaminants originating from human activities have been found in the most remote parts of the world, including polar areas (Xie et al., 2022) and deep-sea trenches (Jamieson et al., 2017). Anthropogenic contaminants include a wide variety of inorganic and organic elements and compounds with differing chemical properties that determine their behaviour and fate in the environment. Effects of contaminants in the environment range from subcellular effects such as oxidative stress (Regoli & Giuliani, 2014) to population- and ecosystem-level consequences including, for example, population declines caused by increased mortality, and changes in nutrient cycling and primary production (Johnston et al., 2015; Shore & Taggart, 2019). Contaminants with persistent, bioaccumulative, and toxic properties are particularly problematic for biota. A contaminant is bioaccumulative when the intake of the contaminant by an organism exceeds the excretion of the contaminant and the contaminant accumulates into the tissues of organisms over time. Many bioaccumulative contaminants are biomagnifying in food webs, meaning that concentrations increase along the trophic level of the species, highest concentrations being found in the apex species. Exposure to multiple contaminants at a time increases the risk of unpredictable interactions of contaminants, and often increases their toxicity in biota (Rodea-Palomares et al., 2015).

1.1 Contaminants of concern

1.1.1 Metals

Metals occur in the environment naturally, and some —such as zinc (Zn), iron (Fe), and sodium (Na)— are essential to organisms. Metals become contaminants, however, when they are released in amounts that exceed the environmental background levels. Metals are released into the environment from natural sources (e.g., volcanic activity), but are released in larger amounts from human sources such as industrial activities and burning of fossil fuels (Järup, 2003).

Sometimes naturally occurring metals are released into the environment in harmful amounts as an indirect consequence of human actions. Such unintentional leaching of metals occurs from acidic sulphate soils, which are typically found in coastal areas (Dent and Pons, 1995). While undisturbed acidic sulphate soils do not generally contribute to metal contamination, human activities such as agricultural land modification may expose the soils to atmospheric oxygen, forming sulphuric acid and lowering of the pH of the soil (Fältmarsch et al., 2008). Metals present in the soils are mobilised in these acidic conditions and are eventually flushed to waterbodies with run-off. Acidic sulphate soil effluents that contain high concentrations of metals have been found to cause changes in feeding behaviour, histology, and development of invertebrates (Vuori and Kukkonen, 1996; Wilson and Hyne, 1997; Dove and Sammut, 2007a, b), as well as deterioration of invertebrate communities (Wallin et al., 2015). Acidic sulphate soil effluents are also harmful to fish, and have even been found to cause mass fish kills (Fältmarsch et al., 2008; Russell et al., 2011; Toivonen and Österholm, 2011). The effects of acidic sulphate soil effluents on higher trophic level species, however, were previously unstudied.

Mercury (Hg), while also released from natural sources, is released into the environment mostly from human sources, including mining, coal burning, and smelting. Hg also can be transported over long distances from the original source of emission via atmospheric currents (Driscoll et al., 2013). The Hg concentrations currently exceed the natural background levels in most parts of the world (Amos et al., 2013). In the environment Hg is problematic due to its bioaccumulation tendency and toxicity, causing symptoms such as impaired motor control and ataxia, changes in behaviour and cognition, reduced immune functioning, and reproductive failure (Burger and Gochfeld, 1997; Wolfe et al., 1998; Burgess and Meyer, 2008; Crump and Trudeau, 2009; Swaddle et al., 2017). For biota, Hg is especially problematic in its organic form methyl-Hg (MeHg), which is more toxic and bioaccumulative than inorganic forms of Hg (Chételat et al., 2020).

1.1.2 Organohalogen contaminants

Organohalogen contaminants are compounds that contain chlorine, bromine, or fluorine atoms (Kodavanti and Logathan 2017). Many organohalogen contaminants are categorised as persistent organic pollutants (POPs). Some of the most notable groups of organohalogen POPs are polychlorinated biphenyls (PCBs), which were used in electronics and lubricating fluids (Erickson and Kaley, 2011); organochlorine pesticides (OCPs); polybrominated diphenyl ethers (PBDEs), which are used as fire retardants in industrial and consumer products (Lee and Kim, 2015); and per- and polyfluoroalkyl substances (PFAS), including perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs), which are used in vast variety of industrial and consumer products in purposes such as surfactants and lubricants (Glüge et al. 2020).

Most chlorinated and brominated POPs (e.g., PCBs, OCPs, and PBDEs) are lipophilic, accumulating particularly into lipid-rich tissues of organisms (La Merrill et al., 2013). PFAS, on the other hand, tend to bind to proteins (Butt et al., 2010; Shi et al., 2018). The range of toxic effects caused by organohalogen contaminants is as wide as the range of compounds, and includes developmental and histological changes, reproductive impairment, neurotoxicity, carcinogenicity, and endocrine disruption (Ulbrich & Stahlmann, 2004; Fisk et al., 2005; Ahrens & Bundschuh, 2014; Akortia et al., 2016; Jayaraj et al., 2016).

Due to their persistent, bioaccumulative and toxic properties, use and production of many harmful past legacy POPs has been banned or restricted for decades under both national and international legislation, including the Stockholm Convention on Persistent Organic Pollutants, initiated in 2004 (Stockholm Convention, 2019). Environmental concentrations of many legacy POPs have consequently decreased (Sturludottir et al., 2014; Sun et al., 2020; White et al., 2021; Xie et al., 2022). Because of their persistency, however, these contaminants are still found in biota globally. In addition, new emerging contaminants, used partly to replace these legacy contaminants, are causing concern due to their potential toxicity and unknown behaviour in the environment (Sauvé & Desrosiers, 2014). While no single definition for term ‘emerging contaminant’ exists, they are often defined as recently discovered, non-monitored chemicals, with potential to enter the environment and cause adverse effects in biota (Chacón 2022).

1.2 Contaminants in food webs

Food web dynamics of contaminants depend on the physiochemical properties of the environment and the contaminant, and the characteristics of the species within the particular food web (Haffner et al., 1994). A food web consists of interconnected food chains, which in turn describe the transference of energy and matter from one trophic level to the next one, i.e., “who eats who”. Trophic magnification factors (TMFs) and biomagnification factors (BMFs) have traditionally been used to study the dynamics of contaminants in food webs and food chains (Borgå et al., 2012). TMF measures the average fold increase in the contaminant concentration per trophic level. TMF is derived from the slope of a linear regression on the logarithmic contaminant concentration and the trophic position (TP) of the species, while the intercept is thought to represent the variation in the basal contamination of the studied food chain or food web (Borgå et al. 2012). The TP of the species is often estimated using bulk stable isotopes of nitrogen $\delta^{15}N$, as the heavier ^{15}N isotope typically tends to enrich from diet to consumers (Cabana & Rasmussen, 1994; Post, 2002). BMFs, in turn, measure the fold increase in contaminant concentration from prey to predator. Multiple different formulas for BMF calculations are currently in use (Franklin, 2016); some normalize for the $\delta^{15}N$ derived TP, while others do not. Regardless of the method of derivation of magnification factors, contaminant is considered to be biomagnifying or bioaccumulative when TMF or BMF > 1.

Using these methods, many contaminants, including Hg and certain organohalogen contaminants, have been found to bioaccumulate and biomagnify in food webs (for example, in Hop et al., 2002; Nfon et al., 2008; Wu et al., 2009; Skarphedinsdottir et al., 2010; and Lavoie et al., 2013). Depending on the characteristics of a food web, the trophic dynamics of contaminants can differ among species with different ecologies. The rate of bioaccumulation and biomagnification has been found to be different in the pelagic and benthic parts of the food web in many aquatic systems. These differences may be attributed to variations in bioavailability of contaminants at the base of the food web or in the metabolism and excretion rates of the species (Lavoie et al., 2010; Muto et al., 2014; Ciesielski et al., 2016; Romero-Romero et al., 2017; Fan et al., 2017). Ecophysiological traits such as thermoregulatory strategy can also affect contaminant bioaccumulation. Homeotherm species have been found to accumulate higher concentrations of contaminants compared to poikilotherm species due higher energy requirements and consequently higher intake of contaminants through their diet (Borgå et al., 2012).

Biomagnification of contaminants in food webs poses increased risk to apex species of food chains, as they tend to accumulate potentially toxic concentrations of contaminants into their tissues. Due to their high trophic position and consequent high exposure to bioaccumulative contaminants, apex species such as birds have been used as sentinel species for monitoring contamination levels of their respective food webs (Helander et al., 2008; Treu et al., 2022). Due to differences in contaminant dynamics between compartments of a food web (e.g., benthic and pelagic food chains), apex species feeding from different parts of the food web thus also reflect differences in the contaminant dynamics of the food chain below them. Apex species can also act as early indicators of biomagnifying emerging contaminants in ecosystems (Treu et al., 2022).

1.3 Contaminants in the Baltic Sea

The Baltic Sea is a coastal sea located in northern Europe, and is vulnerable to contamination due to its shallowness and large drainage area inhabited by 85 million people (Reusch et al. 2018). The Baltic Sea has a long history with contamination, having received high inputs of contaminants over long time from for example industrial and agricultural diffuse and point sources, riverine input, and long-range atmospheric deposition (HELCOM 2018a). The Baltic Sea also receives high concentrations of metal contamination from Europe's largest acidic sulphate soil occurrences in Sweden and Finland (Roos & Åström, 2006).

In the past, Baltic apex species such as the white-tailed eagle (*Haliaeetus albicilla*), ringed seal (*Pusa hispida*), and grey seal (*Halichroerus grypus*) have suffered through population decline caused by contamination (Sonne et al., 2020). Despite decrease in concentrations of many legacy contaminants in Baltic Sea during the last decades (Airaksinen et al., 2014; Nyberg et al., 2015; Sun et al., 2019, 2020), monitoring data shows that concentrations of many legacy contaminants, including metals and organohalogen contaminants, are still elevated in the Baltic Sea basins (HELCOM, 2018b). Simultaneously, unregulated emerging contaminants have been found in the Baltic biota (de Wit et al., 2020). The exact sources and food web behaviour of many emerging contaminants in the Baltic Sea are largely unknown.

1.4 Aims of the thesis

The aim of this thesis is to evaluate the current state and trophic dynamics of legacy and some emerging contamination in a Northern Baltic coastal ecosystem, paying special attention to three avian sentinel species: the white-tailed eagle, the great cormorant (*Phalacrocorax carbo*), and the common eider (*Somateria mollissima*). Foci of the chapters included in this thesis are summarized in **Table 1**. In Chapter **I**, I investigate the white-tailed eagle and great cormorant nestlings as sentinels for acidic sulphate soil associated metal contamination. In Chapter **II**, I investigate the trophic dynamics of Hg in the Baltic Archipelago Sea food web. In Chapter **III**, I investigate the concentrations, profiles, and biomagnification of organohalogen contaminants, including PCBs, OCPs, PBDEs, and PFAS.

Table 1. Main research questions, studied contaminants, and main methods used in each Chapter.

Chapter	Main research questions	Target contaminants	Main methods
I	Do nestlings of white-tailed eagles and great cormorants in proximity to acidic sulphate soils have higher concentrations of metals associated with acidic sulphate soils in their blood than nestlings from control areas?	Acidic sulphate soil associated metals: - aluminium (Al) - cadmium (Cd) - chromium (Cr) - cobalt (Co) - copper (Cu) - lithium (Li) - manganese (Mn) - nickel (Ni) - thallium (Tl) - zinc (Zn)	Comparing concentrations of acidic sulphate soil associated metals of white-tailed eagle and great cormorant nestlings from acidic sulphate soil and control areas
II	Do trophic dynamics of Hg differ between the benthic and pelagic food chains of a Baltic coastal food web?	Mercury (Hg)	Comparing trophic magnification factors and biomagnification factors between benthic and pelagic food chains
III	How do concentrations, profiles, and biomagnification of different organohalogen contaminants differ between invertebrates, fish, and birds of a Baltic coastal food web?	- Polychlorinated biphenyls (PCBs) - dichlorodiphenyltrichloroethane and related compounds (DDTs) - hexachlorocyclobenzene (HCB) - hexachlorocyclohexanes (HCHs) - chlordane and related compounds (CHLs) - chlorinated bornanes (CHBs) - polybrominated diphenyl ethers (PBDEs) - per- and polyfluoroalkyl substances (PFAS)	Comparing concentrations, profiles, biomagnification factors, and trophic magnification factors of contaminants for bird, fish, and invertebrate species

2 Materials and Methods

2.1 Study area

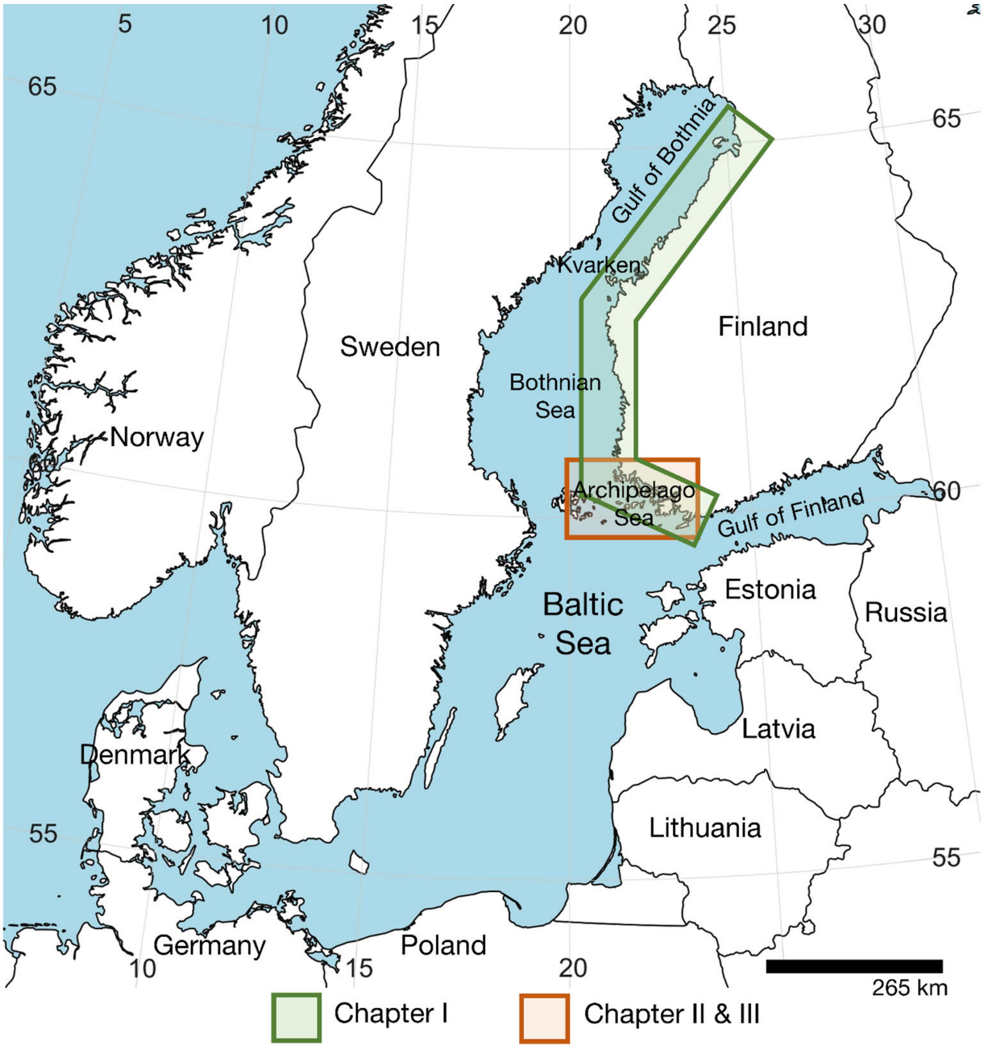


Figure 1. Map of the Baltic Sea and the study areas.

The current Baltic Sea (**Figure 1**) formed following the last glaciation period, after going through several phases with variable salinity conditions. The sea is characterised by brackishness, shallowness, and a large drainage area with high human population density (HELCOM 2018a). During the Littorina Sea phase 7500–4000 years ago, acidic sulphate soils were formed in the sea's anoxic benthic sediments (Fältmarsch et al., 2008). In Finland, acidic sulphate soils are found at the western and southern parts of the coast; hotspots for contamination are in the Kvarken region of the Gulf of Bothnia (Roos & Åström, 2006). The Baltic Sea is connected to the Atlantic Ocean only through the Danish Straits, and it receives high inflow of fresh water through rivers and precipitation. Consequently, the salinity of the Baltic Sea is low compared to oceans, and its salinity decreases from the southern sub-basins, which have a surface salinity of 7–8 practical salinity units (psu), to the northern sub-basins, which have a surface salinity of 0–2 psu (HELCOM, 2018a). Due to lower salinity compared to oceans, the Finnish Baltic coast is inhabited by both marine and freshwater species (Vuorinen et al., 2015).

2.2 Focal study species and their food web trophic structure

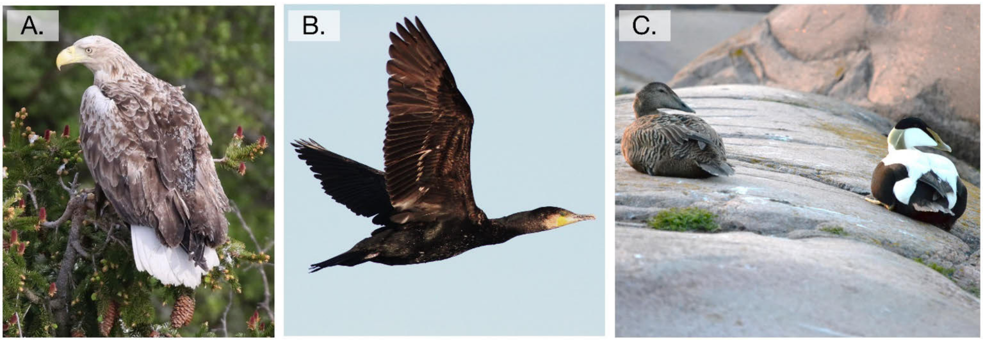


Figure 2. White-tailed eagle (A), great cormorant (B), and common eider female (C, left), and male (C, right). Pictures taken by Toni Laaksonen (A and B) and Céline Arzel (C).

2.2.1 White-tailed eagle

The white-tailed eagle (**Figure 2A**) is an apex avian species that breeds across the whole Baltic region, including all parts of the Finnish coast. This species is not migratory, but individual white-tailed eagles can roam long distances from the breeding areas, particularly when they are young. The white-tailed eagle is an opportunistic predator with diet consisting largely of other bird species, including the common eider and the great cormorant, fish, and terrestrial mammals (Sulkava et al., 1997; Ekblad et al., 2016). The Baltic white-tailed eagle population declined in the 1900s due to persecution and reproductive failure caused by environmental pollutants such as Hg, dichlorodiphenyltrichloroethane (DDT), and PCBs (Koivusaari et al., 1980, Högmander et al., 2020). The productivity of Baltic white-tailed eagles remained low during 1960s and 1970s: in 1972, for instance, only five white-tailed eagle chicks were observed in the whole of Finland (Vösa et al., 2017). Due to extensive conservation efforts and restrictions on the use of POPs, however, the Finnish white-tailed eagle population has recovered in recent decades, with more than 600 chicks being born in 2022 (Sääksisäätiö, 2022). As a consequence of its high trophic position, the white-tailed eagle is vulnerable to trophic magnification and bioaccumulation of environmental pollutants. Productivity of the species is therefore used by the Helsinki Commission (HELCOM) as an indicator of the state of the Baltic Sea (HELCOM, 2018c).

2.2.2 Great cormorant

The great cormorant (**Figure 2B**) is a colonial bird species added to the list of bird species breeding in Finland in 1996 (Lehikoinen, 2006). Its population has increased to 24,300 breeding pairs as of 2021 in Finland (Finnish Environmental Institute, 2021). Great cormorants breed in colonies along the entire Finnish coast and migrate during the winter to the Mediterranean and Central Europe (Saurola et al., 2013). Great cormorants are piscivorous, their diet including multitude of fish species, such as perch (*Perca fluviatilis*), roach (*Rutilus rutilus*), and viviparous eelpout (*Zoarces viviparus*) (Lehikoinen 2005; Lehikoinen et al. 2011).

2.2.3 Common eider

The common eider (**Figure 2C**) is a diving duck species that breeds along the Finnish coast in the Gulf of Finland, Archipelago Sea, and Bothnian Sea (Saurola, 2013). The Finnish common eider population spends their winters in southern Baltic Sea and the North Sea. The diet of common eiders consists mostly of blue mussels (*Mytilus edulis*) but may include also other prey items, mainly invertebrates (Bagge et al., 1973; Öst & Kilpi, 1998). The Baltic common eider population has been on a decline for the last two decades (Ekroos et al., 2012) and, per criteria by the International Union for Conservation of Nature, the species is currently categorised as endangered in Finland (Hyvärinen et al., 2019).

2.2.4 Trophic structure

The Baltic Sea food web comprises benthic, pelagic, and benthopelagic species (**Figure 3**). Benthic species inhabit the sea bottom, while pelagic species inhabit mainly the water column. In contrast to fully benthic or fully pelagic species, benthopelagic species feed from both benthic and pelagic sources and can move between these two environments. Movement between the benthic and pelagic environments and diets can also happen during different ontogenic phases of a species: for example, many fish are planktivorous during their larval and juvenile phases but switch to a macroinvertebrate or fish diet when they grow larger (Lankov et al., 2010, Jacobson et al., 2019). Primary producers, including phytoplankton and macroalgae, are at the base of the Baltic food web. These species are followed by invertebrates, fish, and predatory birds and mammals that feed from aquatic sources.

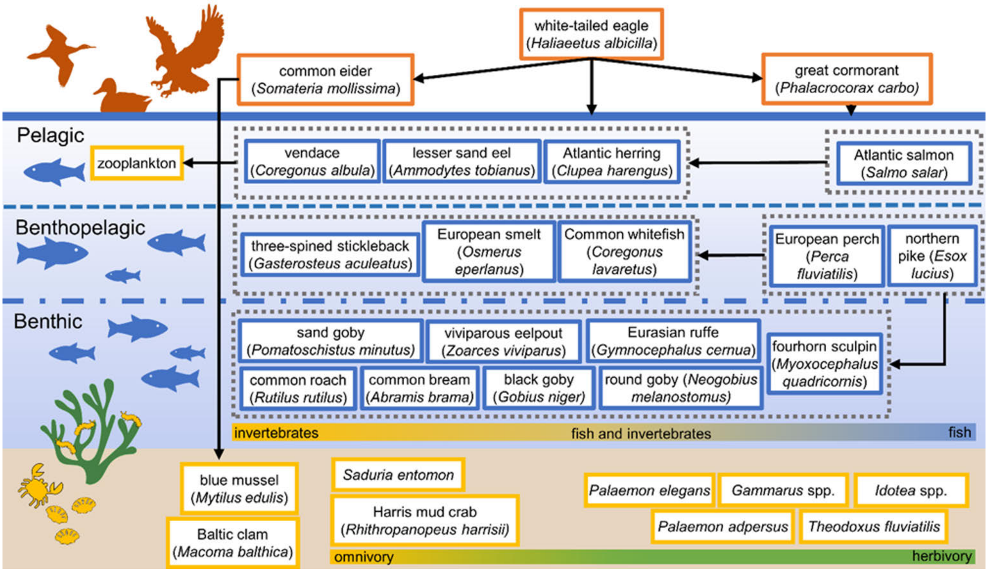


Figure 3. Study species and a simplified trophic structure of the food web for Chapters **II** and **III**. Yellow boxes indicate invertebrates, blue boxes fish, and orange boxes birds. The gradient bars indicate diet composition of invertebrate and fish species.

2.3 Chapter I

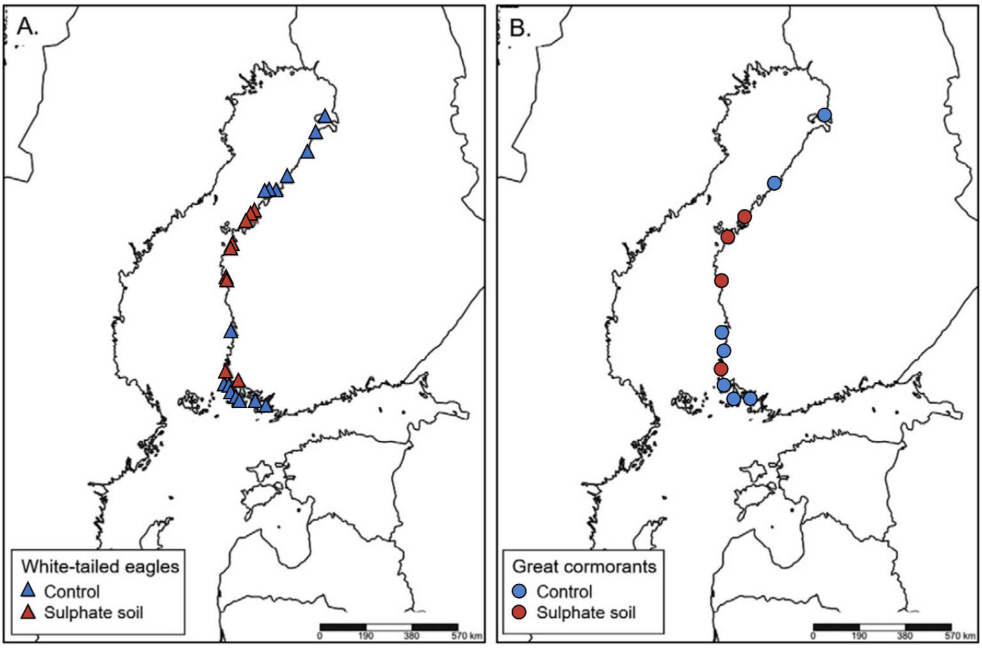


Figure 4. Sampling locations for A. white-tailed eagle territories and B. great cormorant colonies for Chapter **I**.

2.3.1 Sample collection and preparation

Blood samples of 16 white-tailed eagle and 31 great cormorant nestlings were collected in May and June 2017 and 2016, at locations shown in **Figure 4**. From each white-tailed eagle nest, one nestling was sampled, while from each great cormorant colony three nestlings were sampled, except for one colony, where only one nestling was sampled. Nestlings were captured from the nest, measured, ringed, and sampled for 5 ml of blood. Samples were centrifuged to separate plasma and erythrocytes. After separation, samples were stored at -18 °C until chemical analysis was carried out.

2.3.2 Chemical analysis

I investigated erythrocyte concentrations of Al, Cd, Co, Cr, Cu, Li, Mn, Ni, Tl and Zn in chemical analysis. These metals have been associated with acidic sulphate soil effluents in Finland (Åström, 2001; Sohlenius & Öborn, 2004; Fältmarsch et al., 2008; Nordmyr et al., 2008b, a; Nyberg et al., 2012; Nystrand & Österholm, 2013; Wallin et al., 2015; Nystrand et al., 2016). Chemical analysis was carried out at ALS

Scandinavia AB, Luleå, Sweden, using an Inductively Coupled Plasma Mass Spectrometer. Full details of these methods are reported in Rodushkin et al. (2000, 2001).

2.3.3 Data analysis

I assigned each white-tailed eagle nest or great cormorant colony into one of two groups, control or sulphate soil, based on data regarding the magnitude of metal contamination caused by acidic sulphate soils in the closest river or estuary (Roos & Åström, 2005, 2006; Saarinen et al., 2010; Nyberg et al., 2012; Beucher et al., 2014; Wallin et al., 2015, Finnish Institute of Geology, 2019). I measured the distance of each sampling point (nest or colony) to the closest rivers and assigned sampling points in proximity to notably contaminated estuaries and rivers to the sulphate soil group. Metal concentrations in the water column decrease with increasing distance from the estuary due to dilution and precipitation (Åström et al., 2012; Nystrand et al., 2016). I therefore assigned sampling points further than 15 km from the closest estuary to the control group.

I used a general linear mixed model (GLMM) to assess differences between nestling erythrocyte concentrations in sulphate soil and control areas. I also included standardized latitude and interaction of soil type and standardized latitude as fixed factors. Interaction was removed from the models when not significant ($p > 0.05$). For great cormorant models, I added colony as a random effect to control for the intra-colony dependency in metal concentrations. I checked assumptions of normality and heteroscedasticity both visually and using Levene's and Shapiro-Wilk tests, and identified metals that did not fulfil the model assumptions (Al, Cd, Co, Cr, and Ni for white-tailed eagles and Al, Cd, Co, Cr, Mn, Ni, and Tl for great cormorants). In these cases, I used a log-normal transformation. The Ni concentrations in white-tailed eagles showed neither normal distribution nor heteroscedasticity, with or without data transformation, due to a likely outlier; GLMM for Ni of white-tailed eagles was therefore run without the outlier. Another putative outlier appeared in the white-tailed eagle Cr data; results for Cr are presented without the outlier. Marginal means of metal concentrations for the two soil types with their 95 % confidence limits were derived. For log-normal transformed data, I back-transformed values to original scale.

I used Spearman correlations to test for spatial correlations in metal concentrations in great cormorant and white-tailed eagle nestlings. For calculation of these correlations, each sampling point was associated with the nearest sulphate soil or the control area and the mean concentrations of each metal for both species were calculated separately. I also tested for intraspecific correlations between metals.

2.4 Chapters II and III

2.4.1 Sample collection and preparation

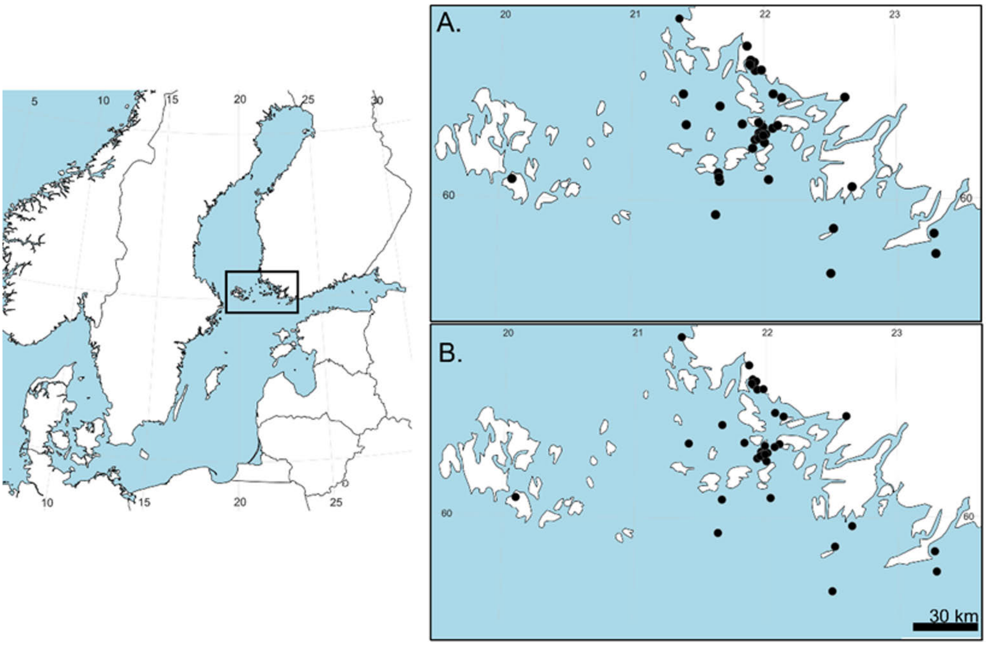


Figure 5. Map of sampling locations for A. Chapter **II** and B. Chapter **III**.

I collected samples of white-tailed eagles, great cormorants, common eiders, and 17 fish and 10 invertebrate species in the Archipelago Sea (**Figure 5**) during years 2017–2019, except for the white-tailed eagle, for which samples were collected during 2013–2019. The study species and simplified trophic structure of the food web are presented in **Figure 3**. The white-tailed eagle samples were carcasses collected by the Natural History Museum of Finland, while the samples of great cormorants and common eiders were shot by hunters. Fish samples were caught using fishing nets or were obtained from local fishers and fish markets. Invertebrates were collected either by hand, or by using bottom trawler, sediment samplers, and trap cages.

After collection, samples were processed according to size and species (**Table 2**). Before the Hg (**II**) and stable isotope analyses, sub-samples were freeze-dried at -50 °C until at stable mass, gravimetrically determined for dry-matter, and homogenised into fine powder using a mortar and pestle. For analyses of organohalogen compounds (**III**), I formed one composite sample per fish and invertebrate species. The composite samples for fish consisted of a maximum of 10 individuals, while the composite samples of invertebrates consisted of samples of multiple individuals collected at different locations, using equal mass of each individual or sample to form the composite sample. For birds, both the liver and muscle of each five individuals per species, were analysed separately. From each sample, a subsample was separated and prepared for stable isotope analysis as described above. All samples were stored at -20 °C before the chemical analyses. Due to insufficient sample mass, organochlorine and organobromine compounds were not analysed from the Harris mud crab. *Palaemon adpersus* was not analysed for organohalogen contaminants.

Table 2. Methods of sample preparation.

Method of preparation	**Species**
Individuals dissected for liver and muscle	common eider, great cormorant, white-tailed eagle
Individuals dissected for fillet	Atlantic herring, Atlantic salmon, common bream, common roach, common whitefish, Eurasian ruffe, European perch, fournhorn sculpin, northern pike
Individuals homogenized whole (gastrointestinal tract removed)	black goby, European smelt, round goby, three-spined stickleback, viviparous eelpout, vendace
Pooled sample of individuals homogenized as whole	*Gammarus* spp., *Idotea* spp., lesser sandeel, *Palaemon adpersus*, *Palaemon elegans*, *Saduria entomon*, sand goby, *Theodoxus fluviatilis*, zooplankton
Pooled sample of homogenized soft tissue	Baltic clam, blue mussel, Harris mud crab

2.4.2 Chemical analyses

The Hg analysis (**II**) was done at the Trace Element Laboratory at the Department of Ecoscience, Aarhus University, Denmark. Contents of total Hg (THg) were determined in freeze-dried and homogenized samples. Analytical details of the Hg analysis are described in Section 2.2. of Chapter **II**.

Analyses of PCBs, OCPs, PBDEs, and PFAS (**III**) were carried out in the Department of Environmental Science, Aarhus University, Denmark. Analytical details of analyses of PCBs, OCPs, and PBDEs are described in Section 2.3. of Chapter **III**. The details extraction method for the liver samples for PFAS analysis is described in Bossi et al. (2005), and for other tissues in Ahrens et al. (2009). Instrumentation details are described in detail in Section 2.4. of Chapter **III**.

The stable isotope analyses for Chapters **II** and **III** were performed at the Stable Isotope Lab of the University of Koblenz-Landau, Germany. The ratios of stable nitrogen isotopes (^{15}N:^{14}N) were determined in freeze-dried and homogenized samples. Details on the stable isotope analyses are provided in Sections 2.3. and 2.5. of Chapters **II** and **III**, respectively.

2.4.3 Data analysis

Based on literature about their diets, I categorised the species under investigation into benthic and pelagic food chains (**II**). Species feeding from both benthic and pelagic sources were categorized as benthopelagic. I calculated the TPs of the pelagic and benthic fish and invertebrates following Post (2002):

$$\mathrm{TP}_{\mathrm{consumer}} = 2 + \frac{\delta^{15}\mathrm{N}_{\mathrm{consumer}} - \delta^{15}\mathrm{N}_{\mathrm{baseline}}}{\Delta^{15}\mathrm{N}} \tag{1}$$

where I used the trophic enrichment factor (TEF) $\Delta^{15}N$ of 3.40, as recommended by Borgå et al. (2012), and where $\delta^{15}N_{baseline}$ is the $\delta^{15}N$ value of the specified benthic or pelagic primary consumer species (assumed TP = 2).

I used *Gammarus* spp. and zooplankton, respectively, as benthic and pelagic baseline species. As the benthopelagic species feed from both benthic and pelagic sources, I calculated the TPs of benthopelagic species also following additional two-source model following Post (2002):

$$\mathrm{TP}_{\mathrm{consumer}} = 2 + \frac{\delta^{15}\mathrm{N}_{\mathrm{consumer}} - (\delta^{15}\mathrm{N}_{\mathrm{baseline\_B}} \times \alpha) + \left(\delta^{15}\mathrm{N}_{\mathrm{baseline\_P}} \times (1-\alpha)\right)}{\Delta^{15}\mathrm{N}} \tag{2}$$

where the $\delta^{15}N_{baseline\_B}$ and $\delta^{15}N_{baseline\_P}$ represent the $\delta^{15}N$ values of benthic and pelagic baseline species, respectively, and α is the proportion of feeding from the first food chain (here benthic). As the actual proportions in which the benthopelagic species feed from benthic and pelagic food chains are unknown, I set $\alpha = 0.50$.

As the TEF of birds differs from those of fully aquatic species due to metabolic differences, I used TEF of 2.40 for birds. This value has been found to represent the trophic enrichment between bird muscle tissue and their prey more appropriately than the TEF of 3.40 (Mizutani et al., 1991). I calculated the TP of the benthic common eider and the TP of the white-tailed eagle and great cormorant using a one-source model based on Hop et al. (2002):

$$\mathrm{TP}_{\mathrm{bird}} = 3 + \frac{\delta^{15}\mathrm{N}_{\mathrm{bird}} - (2.4 + \delta^{15}\mathrm{N}_{\mathrm{baseline}})}{\Delta^{15}\mathrm{N}} \tag{3}$$

For the benthopelagic white-tailed eagle and great cormorant, I calculated the TPs also using a two-source model adapted from Hop et al. (2002) and Post (2002):

$$TP_{bird} = 3 + \frac{\delta^{15}N_{bird} - [2.4 + (\delta^{15}N_{baseline\_B} \times \alpha) + (\delta^{15}N_{baseline\_P} \times (1\text{-}\alpha))]}{\Delta^{15}N} \quad (4)$$

For the Hg data, I used a linear mixed model for both one-source and two-source models for benthic and pelagic food chains. I used log_{10}-transformed Hg concentration (dry weight or dw) as the dependent effect and TP as the independent effect. Due to the unbalanced samples sizes among the species, I incorporated species as a random effect in the model. I assessed the trophic magnification of Hg in both benthic and pelagic food chains using TMFs derived from the slope (b) of each GLMM as

$$TMF = 10^{b} \quad (5)$$

I also calculated TMFs for sum concentrations of organohalogen contaminant groups: ∑PCBs, ∑DDTs, HCB, ∑HCHs, ∑CHLs, ∑CHBs, ∑PBDEs, ∑PFCAs and ∑PFSAs. For each contaminant group, I used a linear regression coefficient of log_{10}-transformed sum contaminant concentration on TP. For birds I used the mean sum concentration of a species for each contaminant group. Similar to the Hg model, I derived the TMF for each contaminant group from the slope of the regression using Equation 5. I compared the goodness of fit for the Hg models using marginal R^2_m, representing total variance explained by fixed effects, and conditional R^2_c, representing the proportion of variance explained by both fixed and random effects (Nakagawa et al. 2017). For the organohalogen contaminant models, I used adjusted R^2 to assess the goodness of the fit.

I also assessed the biomagnification of Hg and organohalogen contaminants by calculating BMFs using two different formulas, both commonly used in literature:

$$BMF_{TP} = \frac{[Contaminant_{predator}]/[Contaminant_{prey}]}{TP_{predator} - TP_{prey}} \quad (6)$$

$$BMF_{R} = [Contaminant_{predator}]/[Contaminant_{prey}] \quad (7)$$

I assessed the risk of Hg-exposure-associated negative health effects for each functional group (birds, fish, and invertebrates) and their component species using risk categories. For invertebrates and fish, I used threshold values suggested by Dietz et al. (2021), while for birds I used those suggested by Dietz et al. (2019). The risk categories, along with their corresponding concentration thresholds, are provided in **Figure 9**. These risk categories evaluate the risk of negative health effects, such as impaired motoric control and ataxia, behavioural and cognitional changes, and reduced function of the immune system and reproductive failure (Burger & Gochfeld, 1997; Wolfe et al., 1998; Burgess & Meyer, 2008; Crump & Trudeau, 2009; Swaddle et al., 2017). I included only species with sample size ≥ 5 in the assessment. For birds, I used liver concentrations, while for fish I used either muscle or whole fish concentrations depending on the species and the tissue analysed. As the risk thresholds for fish suggested by Dietz et al. (2021) were established for muscle tissue, I adjusted these values using the average tissue conversion factors reported by Boalt et al. (2014) and Rüdel et al. (2020) to fit fish analysed as a whole. Further, because the suggested thresholds for invertebrates reported by Dietz et al. (2021) are valid for bivalve soft tissue only, I included blue mussel as the only invertebrate to the risk assessment. It is important to note that these latter risk thresholds for bivalves are not indicative for a health risk for the bivalves but rather for their consumers (Dietz et al. 2021).

I assessed the detection frequencies, profiles, and concentrations of different organohalogen compound groups (PCBs, OCPs, PBDEs, PFCAs, and PFSAs) between the studied species. I also used the Kruskal-Wallis H test, and post-hoc Mann Whitney U test to evaluate the difference in sum concentrations of PCBs, OCP groups, PBDEs, PFCAs, and PFSAs between the bird species.

3 Results and discussion

3.1 Nestlings as spatial sentinels for acidic sulphate soil associated metal contamination on the Finnish coast

In the Chapter **I**, I found concentrations of multiple metals to be elevated in the white-tailed eagle and great cormorant nestlings from the sulphate soil areas as compared to the control group (**Figure 6**). This finding suggests that acidic sulphate soils are a source of contamination of certain metals for white-tailed eagle and great cormorant nestlings on the Finnish coast, and that nestlings can be used as sentinels of those metals. There were, however, differences in which elements were elevated in the sulphate soil areas compared to the control for these two species. In white-tailed eagles, concentrations of Al and Co were higher in the sulphate soil areas. In great cormorants, concentrations of Cu and Mn were elevated in sulphate soil areas compared to the control, while concentrations of Zn were found to be higher in the control areas compared to the sulphate soil areas. There were also between-species differences in the metal correlations. Out of several statistically significant correlations between different metals, only correlations between Al and Cr, as well as Co and Li, occurred in both species. These between-species differences in which elements were elevated in the sulphate soil areas compared to control, and lack of between-species correlations of metals despite spatial overlap in the nest and colonies, are probably related to differences in the diet of these species. The nestling diet of white-tailed eagles on the Finnish coast consists mostly of birds, such as ducks and gulls, and fish species, such as pike and Cyprinids (Sulkava et al., 1997; Ekblad et al., 2016); great cormorants, in contrast, feed their chicks mainly viviparous eelpout, roach, and perch (Lehikoinen, 2005). As different prey species inhabit different environments and feed from different sources, they may also be exposed to different contaminants. For example, benthic species might be more exposed to and accumulate contaminants that have precipitated into the bottom sediments than species in the pelagic environments (Simpson & Batley, 2007). Such differences in the contaminant burden in the prey species are reflected in the contaminant profile and levels in their predators.

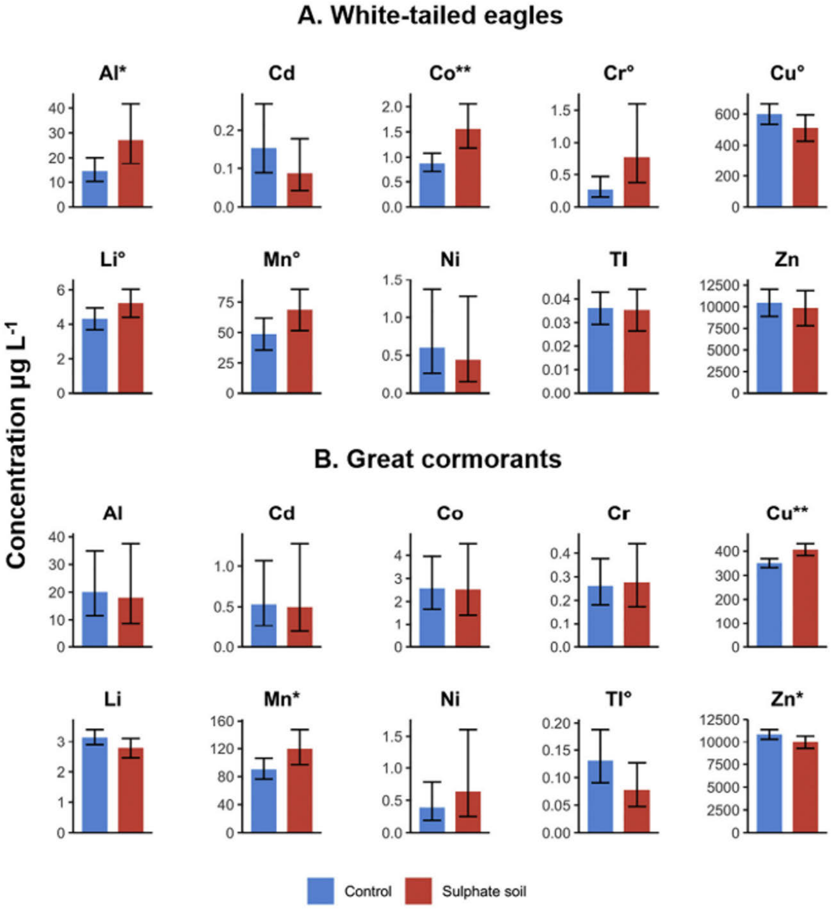


Figure 6. Estimated marginal means and their 95% confidence intervals for erythrocyte metal concentrations in control and sulphate soil areas for A. white-tailed eagle nestlings (n = 24) and B. great cormorant nestlings (n = 31). Results for Co and Li concentrations in great cormorant nestlings without the interaction between latitude and soil type are included for comparison. °: $p \leq 0.10$, *: $p \leq 0.05$, **: $p \leq 0.01$.

For many metals, however, I did not find differences in the concentrations between nestlings from control and acidic sulphate soil areas, indicating that sulphate soils are not the primary source of contamination of these metals for the nestlings. In order for contaminants to end up in the bird nestlings, contaminants must be first available for uptake and accumulate into the prey species. Concentrations of many acidic sulphate soil associated metals —for example Al, Co and Cu— have been found to decrease in estuarine water with increasing distance from the river mouth due to changes in the water chemistry (Åström et al., 2012; Nystrand et al., 2016). As their concentrations decrease, metals are less available for uptake by organisms. Metals might not also be absorbed through the dietary tract into the tissues of the prey, or they might be excreted efficiently, and would thus not end up into the tissues of their predators. Further, depending on abiotic and biotic factors of the system under investigation, metals can biodilute in the food chain (as in Campbell et al., 2005; Nfon et al., 2009; and Hu et al., 2021), meaning that metals are not transferred through the food chain to the apex species. In these cases, the highest concentrations of metals are found in the species at the bottom of the trophic structure. If such biodilution is occurring with the acidic sulphate soil associated metals on the Finnish coast, metals might be present in elevated concentrations for species that are in the bottom of the food web, but not in the apex species.

There were also latitudinal gradients in the concentrations of many of the studied metals in both of the study species (**I**). In white-tailed eagles, concentrations of Cd and Mn were higher in the southern parts of the coast compared to the northern parts, while concentrations of Tl were higher in the north compared to the south. Concentrations of Tl were also higher in the north compared to the south in great cormorants, as were concentrations of Zn. In great cormorants there was also a correlation between soil type and latitude for Co and Li. The concentrations of both metals increased in great cormorant nestlings with latitude, and this increase was found to be steeper in nestlings from the sulphate soil areas compared to the control areas. These latitudinal trends in the concentrations of metals probably reflect differences in the sources and quantity of emissions in different parts of the coast. Differences in the contamination patterns of different metals also suggest that these metals have different sources into the Baltic Sea. For Cd, riverine input into the Baltic is largest into the Gulf of Finland from Russia; atmospheric emissions, in contrast, are largest in the more southern parts of the Europe, with deposition decreasing from the southern parts of the Baltic to the northern parts (HELCOM, 2018d). These contamination patterns may explain the latitudinal trend of Cd concentrations in white-tailed eagle nestlings, as well as similar latitudinal trends of other metals. Metals with the opposite trend (i.e., decreasing concentrations north to south) are possibly indicative of higher inputs of metals to the northern parts of the Baltic Sea. Differing chemical and physical properties in different parts of the coast may also affect the bioavailability of contaminants. An increase in salinity and decrease in pH, for example, decreases the concentrations of many acidic sulphate soil associated metals in water column (Nystrand et al., 2016).

Concentrations of the studied acidic sulphate soil associated metals in blood of the nestlings were generally low, or were similar to levels reported to be below toxic thresholds in blood samples in other bird species (Dolan et al., 2017; Fenstad et al., 2017; Maia et al., 2017). Some caution should be taken, however, when comparing the concentrations in Chapter **I** with other studies, as erythrocytes were used here instead of plasma or whole blood concentrations. Moreover, while concentrations were similar to those reported in earlier studies, the nestlings were exposed to several metals simultaneously, as demonstrated by several positive correlations between different metals in both species. Simultaneous exposure to different metals poses a risk of possible additive and interactive effects between metals, as metals can work both antagonistically and synergistically together (Pan et al., 2015). As the studied individuals were nestlings, it is hard also to project the effects of long-term exposure to metals in proximity to acidic sulphate soil areas for adult birds, as metals tend to accumulate into tissues of birds with age (Berglund et al., 2011; Maia et al., 2017). This accumulation may lead to elevated concentrations in individuals' tissues over the lifespan.

3.2 Trophic dynamics of mercury and organohalogen contaminants in the Archipelago Sea food web

In Chapter **II** and **III** I found that Hg, ∑PCBs, ∑DDTs, HCB, and ∑CHL biomagnify in the Archipelago Sea food web, as demonstrated by values for TMFs and most BMFs being >1 **(Figure 7** and **Figure 8**).

When excluding birds from Hg trophic magnification models, I found the rate of trophic magnification of Hg to be higher in the pelagic food chain as compared to the benthic food chain. This increase was consistent regardless of whether one- or two-source model was used to derive the TP of the species or whether the $\delta^{15}N$ signal was used instead of TP (**II**). Lavoie et al. (2010) and Muto et al., (2014), similarly, have found higher TMFs of Hg in the pelagic food chain compared to the benthic food chain in other aquatic systems. The difference in trophic dynamics of Hg between the benthic and pelagic food chains may be related to different ratios of inorganic Hg and organic MeHg. As MeHg is more bioaccumulative than inorganic Hg (Chételat et al., 2020), higher proportions of MeHg in the pelagic food chain could lead to a higher rate of biomagnification along the food chain. However, as the speciation of Hg was not examined in Chapter **II**, this relationship cannot be confirmed. Although not reflected by the trophic magnification models with the birds included, the results demonstrate that avian predators feeding from different parts of the food web can be exposed to differing amounts and profiles of contaminants. In the case of Hg in the Archipelago Sea food web, predators feeding from the pelagic part of the food web might be at greater risk of Hg bioaccumulation, and thus toxic effects of Hg, than those feeding from the benthic food chain.

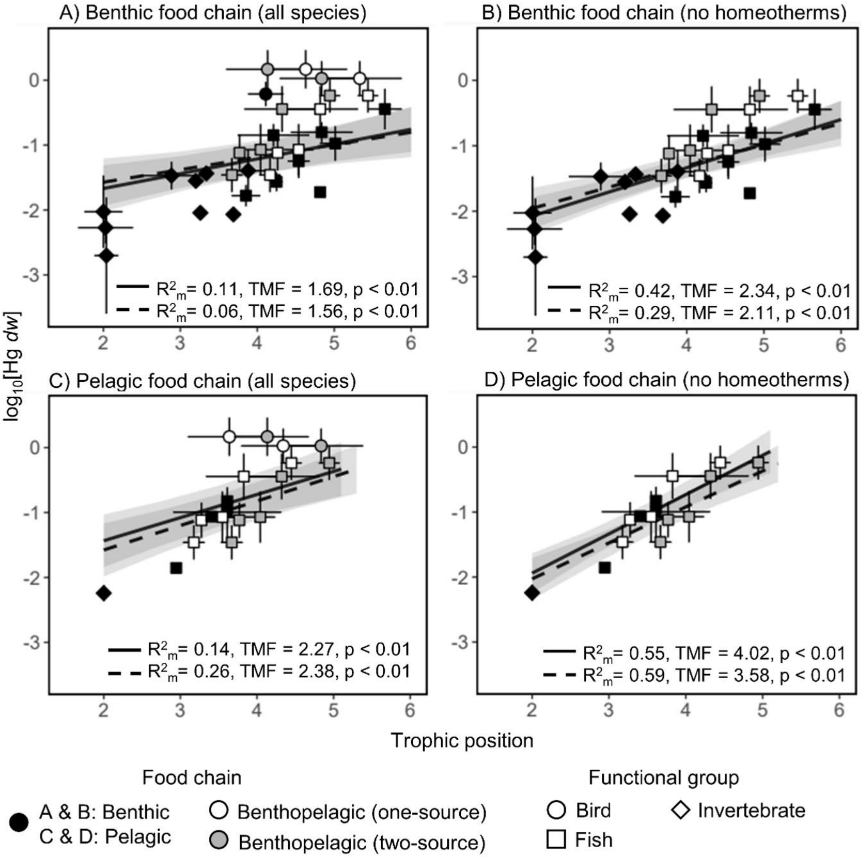


Figure 7. Linear regressions between $\log_{10}$[THg dry weight] (µg g^{-1}) and trophic position, R^2_m, trophic magnification factors, and the p-values of the slope for benthic (A, C) and pelagic (B, D) food chains. Solid lines illustrate regressions of the one-source model (Equations 1 and 3), while dashed lines illustrate regressions for the two-source model (Equations 2 and 4).

Based on TMFs, ∑PCBs, HCB, ∑DDTs, ∑CHLs, and ∑PFSAs biomagnify in the Archipelago Sea food web, rate of trophic magnification being highest for ∑PCBs. Based on TMFs, ∑HCHs, ∑PBDEs, and ∑PFCAs did not seem to biomagnify. BMFs for PCBs, DDTs, HCB, CHLs and PBDEs tended to be highest in birds (**III**), indicating higher biomagnification in the top of the food chain. BMFs for PFCAs and PFSAs showed a more variable pattern of biomagnification. These findings are consistent with other studies which show that lipophilic contaminants —including PCBs, DDTs, HCB, CHLs, and PBDEs— often biomagnify across different systems (Hop et al., 2002; Nfon et al., 2008; Wu et al., 2009; Hallanger et al., 2011; Fremlin et al., 2020), while PFAS have more variable magnification patterns between different environments (Miranda et al., 2022). As biomagnification tendencies can differ between congeners and compounds within a compound group (Nfon et al., 2008; Miranda et al., 2022), however, TMFs derived for the sum concentrations should be considered as rough estimates not accounting for individual variation between compounds.

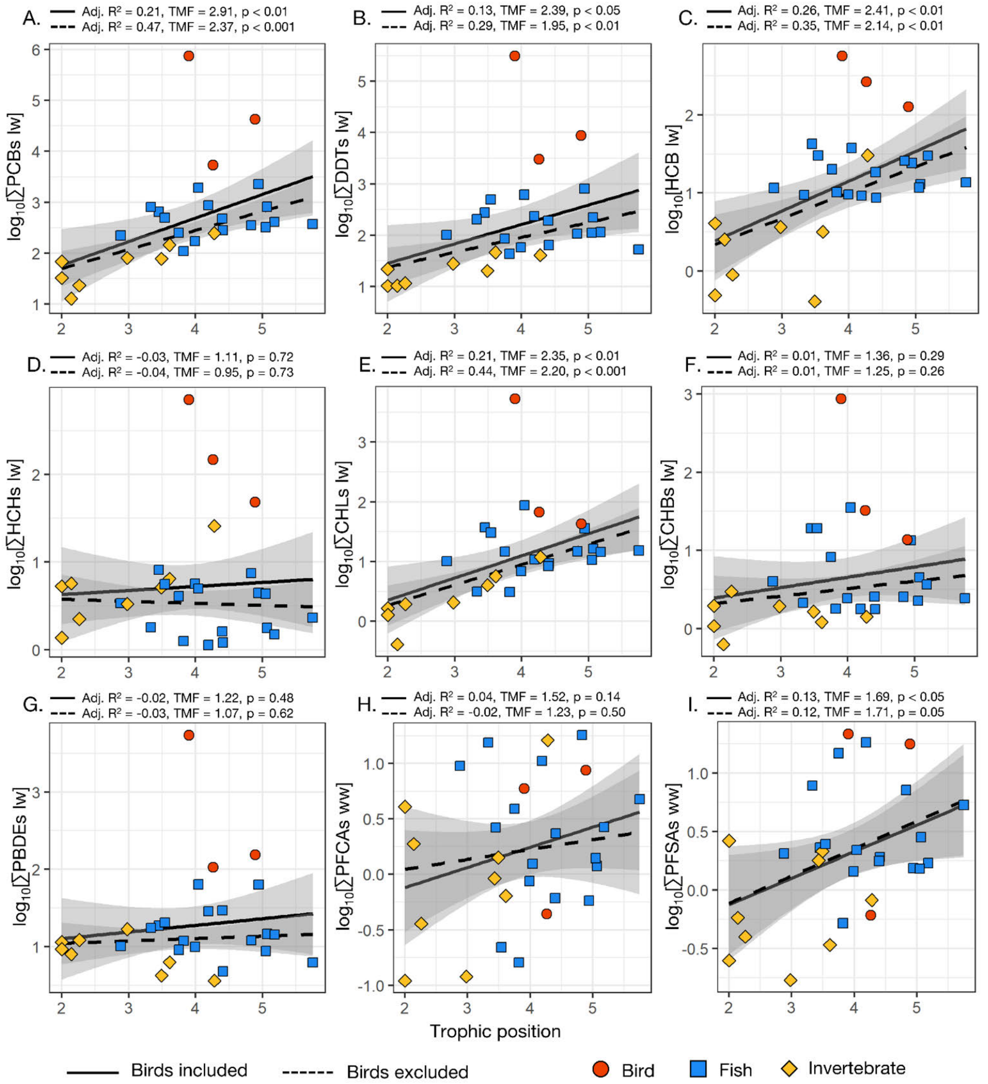
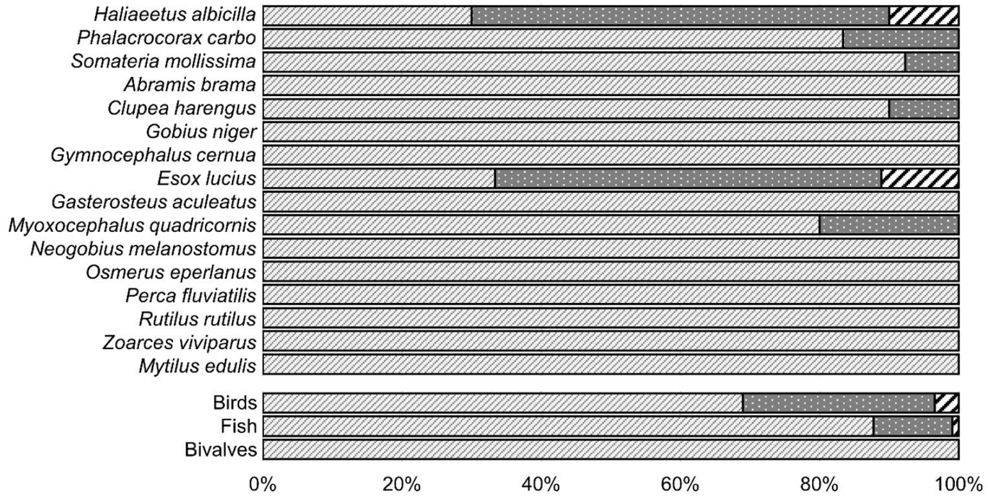


Figure 8. Linear regressions between $\log_{10}$-transformed concentrations of organohalogen contaminants and trophic position, adjusted R^2, trophic magnification factors, and p-values for the slopes. Solid lines illustrate regressions with birds included in the data, dashed lines regressions with birds excluded.

Concentrations of Hg and most of the studied organohalogen contaminants in birds were higher in relation to their TP than those of fish and invertebrates, which decreased their fit to the linear trophic magnification models and affected the results of the magnification models overall. This effect was demonstrated by the higher R^2_m (**Figure 7**) and adjusted R^2 (**Figure 8**) of the significant slopes when birds were excluded from the models. Multiple studies have reported higher bioaccumulation of contaminants in homeotherm species, such as birds, as compared to poikilotherm species (Hop et al., 2002; Wang et al. 2013; Romero-Romero et al., 2017). These levels are attributed to higher energy requirements, which result in higher food consumption and greater intake of contaminants (Borgå et al., 2012). Further, while the TP derived using the $\delta^{15}N$ of common eider and great cormorant seemed to match the TP of species in dietary studies (Bagge et al., 1973; Öst & Kilpi, 1998; Lehikoinen, 2005; Lehikoinen, et al., 2011), the $\delta^{15}N$ based TP of the white-tailed eagle did not match the apex position the species has in the food web, with diet including high trophic level fish and other birds, such as common eiders and great cormorants (Sulkava et al., 1997; Ekblad et al., 2016). In fact, the mean TP of the 10 white-tailed eagles in Chapter **II** was lower than that of the great cormorant and ≤ 0.5 TPs higher than common eider. Similarly, the mean TP of five white-tailed eagles in Chapter **III** was lower than the TP of the other two bird species, many fish species, and even the invertebrate scavenger *Saduria entomon*, demonstrating the effect of individual variability on the TP. As trophic enrichment of $\delta^{15}N$ can vary between species, individuals, and tissues (Stephens et al., 2022), it is possible that the TEF of 2.40 used for birds does not reflect the actual trophic enrichment of $\delta^{15}N$ between the diet and tissues of the white-tailed eagle. Also, as white-tailed eagles can forage across large areas (Saurola et al., 2013), the result might be affected by a temporal and spatial mismatch between the $\delta^{15}N$ signal of the chosen isotopic baseline species and the $\delta^{15}N$ signal of the white-tailed eagles, as well as by individual variability in diet composition. While the Baltic white-tailed eagles mostly feed on fish and other birds, their diet can also include terrestrial prey, such as mammals (Sulkava et al., 1997), further introducing uncertainty to their TP determination. These results call for consideration of ecological and ecophysiological traits and their accommodation in the models for individual species when studying contaminant dynamics, as these traits can have direct effects on model outcomes.

3.3 Levels and profiles of mercury and organohalogen contaminants in the Archipelago Sea food web



	▨ No risk	░ Low risk	▨ Moderate risk	▧ High risk	■ Severe risk
Birds (liver) $\mu g\ g^{-1}$	< 1.40	1.4 - 7.3	7.3 - 22.7	22.7 - 30.5	> 30.5
Fish (muscle) $\mu g\ g^{-1}$	< 0.10	0.10 - 0.30	0.30 - 0.50	0.50 - 2.0	> 2.0
Fish (whole) $\mu g\ g^{-1}$	< 0.08	0.08 - 0.24	0.24 - 0.40	0.40 - 1.6	> 1.6
Bivalves (soft tissue) $\mu g\ g^{-1}$	< 0.01	0.01-0.05	0.05-0.15	0.15-0.40	> 0.4

Figure 9. Risk Category proportions of individuals of each species or functional group. Calculations follow thresholds suggested by Dietz et al. (2021) for fish and bivalves and Dietz et al. (2019) for birds. Risk Categories for bivalves indicate health effects for predators with bivalve diet, not for bivalves themselves.

Risk assessment for Hg-mediated health effects based on risk categories of the total mercury (THg) concentrations showed that most of the species under investigation were at no risk or low risk of Hg-associated negative health effects (**Figure 9**). All but one white-tailed eagle individuals, and all common eider and great cormorant individuals were at no risk or low risk. The remaining white-tailed eagle was at moderate risk of negative health effects caused by Hg. The Hg levels in fish indicated no or low risk, except for one northern pike, which was at moderate risk of Hg-mediated negative health effects. Assessment of blue mussel samples indicated no risk for species feeding on them. These results are consistent with another recent risk assessment that likewise reported fish and bivalve-eating species from the Baltic Sea to be generally at no or low risk of negative health effects (Dietz et al., 2021). Based on feather Hg concentrations, white-tailed eagles examined by Dietz et al. (2021)

were either at moderate or severe risk. In contrast, our risk assessment, which was based on liver concentrations, showed only one of 10 white-tailed eagles to be at moderate risk. Based on these results, Hg concentrations in the Archipelago Sea biota seem generally low.

The concentrations of organochlorine and organobromine contaminants generally increased from invertebrates to fish, and from fish to birds (**III**) (**Figure 10** and **Figure 11**). Legacy contaminants, including PCBs, DDTs, HCB, HCHs, CHLs, CHBs, and PBDEs, were found in all species groups; concentrations were highest in the white-tailed eagles. Congeners and compounds with high bioaccumulation tendency, such as PCB-153 and *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE), were found in most species, with proportions increasing along the trophic level. Contaminant profiles of the three bird species differed in some respects. For example, proportions of HCB and ∑HCHs were slightly higher in the benthic common eider compared to the white-tailed eagle and great cormorant, while the proportion of ∑DDTs was lower in the common eider compared to the other bird species. Similarly, the highest concentrations and proportions of PBDE-209, which is associated with benthic contamination (Lee & Kim, 2015), were found in the common eider; proportion of PBDE-209 of ∑PBDEs was, however, low in all three bird species.

Among PFAS, concentrations of perfluorodecanesulfonic acid (PFDS), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorotetradecanoic acid (PFTeA), and the emerging PFAS perfluorobutanesulfonic acid (PFBS), 6:2 chlorinated polyfluorinated ether sulfonate (6:2 Cl-PFAES), and 8:2 chlorinated polyfluorinated ether sulfonate (8:2 Cl-PFAES) were below the limit of detection in all species. As expected, perfluorooctanesulfonic acid (PFOS) dominated the PFAS concentrations in most species (Butt et al., 2010), followed by perfluorononanoic acid (PFNA). I found the cyclic PFAS perfluoroethylcyclohexane sulfonate (PFECHS) in the white-tailed eagle, great cormorant, Atlantic salmon, and Eurasian ruffe, with highest concentrations in the white-tailed eagle. A recent study found PFECHS for the first time in the Baltic biota, specifically in aquatic mammals and sea bird eggs (de Wit et al., 2020). PFECHS has been also found in Finnish riverine fish (Kärrman et al., 2019). The finding of PFECHS in Finnish white-tailed eagles, great cormorants, and fish together with the other studies indicate that this novel compound is widespread in the Baltic Sea environment. In other systems, PFECHS has been found to bioaccumulate into aquatic biota (Wang et al., 2016; Munoz et al., 2022) and birds (Szabo et al., 2022), and has been shown to biomagnify in a food web (Munoz et al., 2022). Concentrations of PFECHS increased in order of Eurasian ruffe < Atlantic salmon < great cormorant < white-tailed eagle (**III**), possibly indicating that biomagnification of this contaminant also occurs in the Baltic Sea food web.

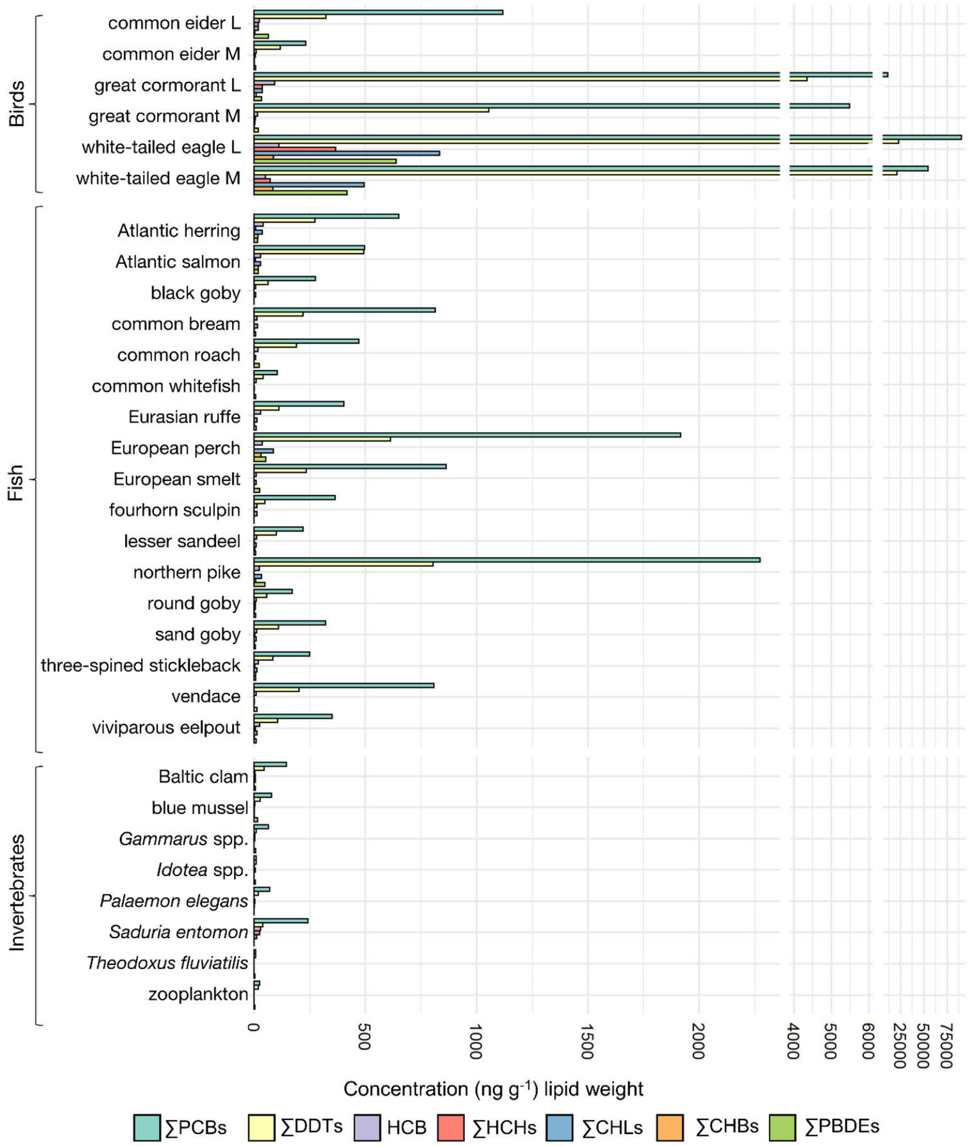


Figure 10. Concentrations (ng g^{-1} lipid weight) of PCBs, DDTs, HCB, HCHs, CHLs, CHBs, and PBDEs in the studied species. For the bird species, mean concentrations are reported. L: liver, M: muscle. Note axis cut points with change in scale.

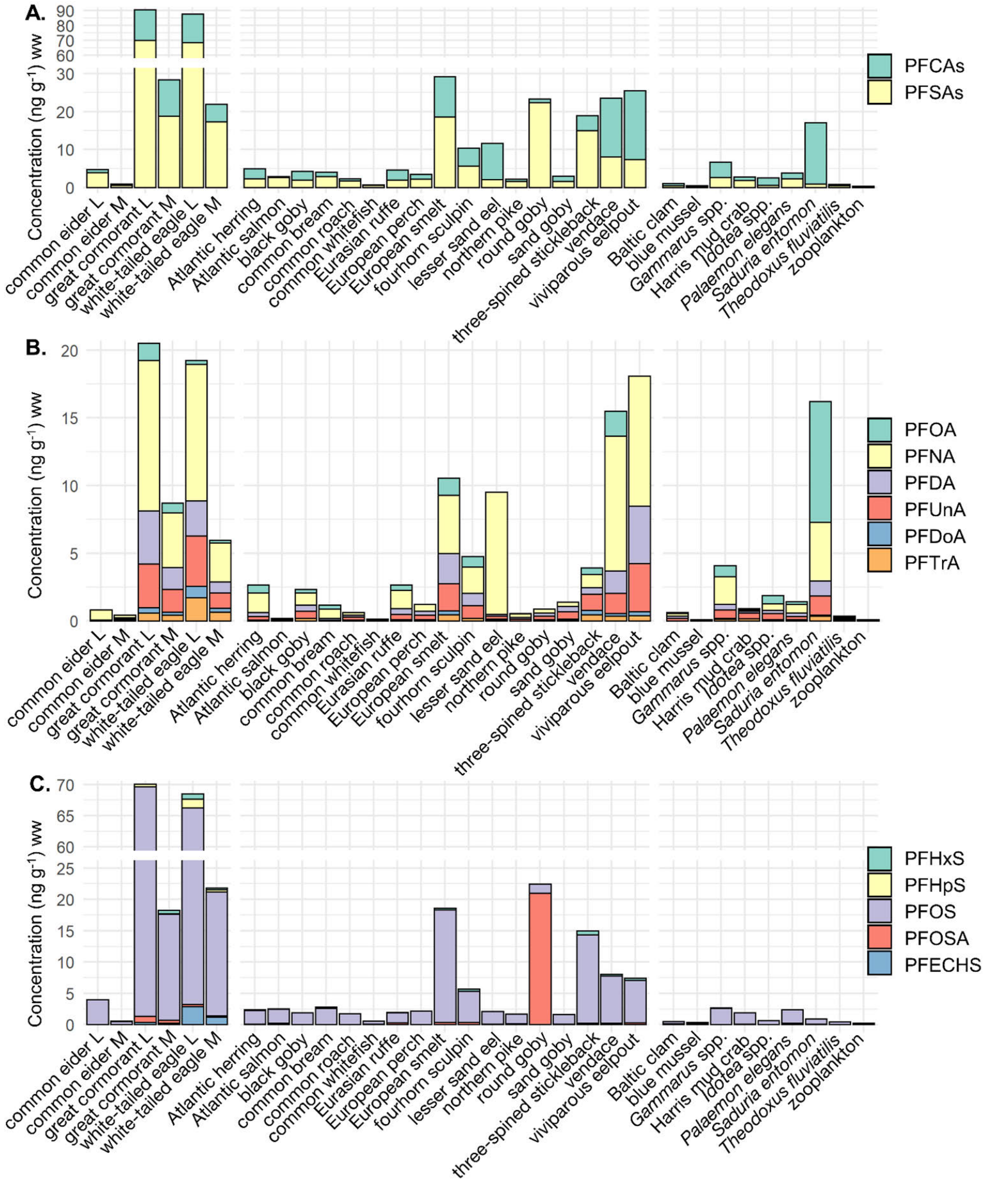


Figure 11. Concentrations (ng g^{-1} weight) of A. ∑PFCAs and ∑PFSAs, B. PFCAs, and C. PFSAs in the study species in the studied species. Sulfonamide PFOSA is grouped with the PFSAs in the figure. For the bird species, mean concentrations are reported. L: liver, M: muscle. Note axis cut points with change in scale in A and C.

Mean sum concentrations of most compound groups in white-tailed eagle clearly exceeded those in the other species under investigation, with concentrations being highest for ∑PCBs and ∑DDTs. One white-tailed eagle in particular had very high concentrations for both wet weight (ww) and lipid weight (lw) of organochlorine and organobromine compounds in both its liver and muscle tissue compared to other white-tailed eagle individuals. As the lipid percentages of the tissues of the contaminated white-tailed eagle were low compared to the other individuals of the species, it is possible that the contaminated individual had starved before its death, thus possibly releasing stored lipophilic contaminants from the adipose tissue (La Merrill et al., 2013).

Out of chlorinated and brominated compounds, the ww and lw concentrations of ∑PCBs, ∑DDTs, ∑CHLs, ∑CHBs, and ∑PBDEs, and ww concentrations of HCB and ∑HCHs differed between the bird species ($7.98 \leq \chi^2 \leq 11.58$, $0.003 \leq p \leq 0.02$). The ww and lw concentrations of ∑DDTs, ∑CHLs, ∑CHBs, and ∑PBDEs and ww concentrations of ∑PCBs, HCB, and ∑HCHs in muscle tissue were higher in white-tailed eagles than in the other two bird species ($0 \leq U \leq 2$, $0.008 \leq p \leq 0.03$), and ∑PCBs ww and lw were higher in the great cormorant than in the common eider ($0 \leq U \leq 2$, $0.008 \leq p \leq 0.03$). There were no differences, however, in the sum concentrations of ∑PFCAs ($U = 17$, $p = 0.42$) or ∑PFSAs ($U = 13$, $p = 1$) of white-tailed eagles and great cormorants, and concentrations of ∑PFCAs and ∑PFSAs were lower in the common eider than in the other bird species ($U = 0$, $p < 0.01$). These results indicate that white-tailed eagles and great cormorants are exposed to similar levels of the investigated PFAS in the Baltic environment, but that white-tailed eagles are exposed to higher concentrations of most of the studied chlorinated and brominated legacy POPs. This finding is probably related to ecological differences in diet composition. The fish species with the highest concentrations of PFAS were the size that are more caught by great cormorants than white-tailed eagles, possibly contributing to the lack of difference in the PFAS concentrations between the two species. Also, as the age of the studied white-tailed eagles is unknown beyond that they were mature individuals older than five years, it is possible that the differences in concentrations of chlorinated and brominated POPs between the white-tailed eagles and great cormorants is due to higher age of the white-tailed eagle individuals in the sample. However, as a similar difference in concentrations of white-tailed eagles and great cormorants was not observed for the bioaccumulative legacy PFAS such as PFOS ($U = 14$, $p = 0.84$), differences in species ecology seem more likely explanation.

On the other hand, common eiders seem to have lower exposure to PFAS than the two bird species, while the concentrations of chlorinated and brominated compounds (except for ∑PCBs) do not seem to differ between the common eider and the great cormorant, even though common eiders feed on species at lower trophic levels (Bagge et al., 1973; Öst & Kilpi, 1998; Lehikoinen, 2005, Lehikoinen, et al., 2011). This result could be indicative of higher exposure to certain legacy contaminants through their benthic diet. Overall, concentrations of most of the investigated legacy contaminants in the birds and in their diet in the Archipelago Sea were lower than those reported in 1990s and 2000s (Kiviranta et al., 2003; Burreau et al., 2004; Krone et al., 2006; Pikkarainen, 2007; Stephansen et al., 2012; Vuorinen et al., 2017). This finding is consistent with time-trend data of contaminant concentrations from the Baltic Sea (Airaksinen et al., 2014; Nyberg et al., 2015; Sun et al., 2020).

3.4 Ecological uncertainty related to birds as sentinels for contamination

As demonstrated by the findings in this thesis and by different monitoring schemes (Treu et al., 2022), birds can act sentinel species of food web contamination: their contaminant burden provides an overview of the contaminant burden of the food web below them. However, there are also several elements of uncertainty in using birds as sentinels for contamination. Firstly, except for nestlings, birds are rarely sedentary, and can move long distances during their lifetime. Birds can move long distances when foraging, while most foraging usually happens close to the territories or colonies of the species (Hentati-Sundberg et al., 2018, Krone et al., 2013, Haworth et al., 2010). Many birds, such as the common eider and great cormorant, are migratory and spend large portions of the year in areas outside their breeding areas; others, like the white-tailed eagle, can roam considerable distances, especially outside the breeding season (Saurola et al., 2013). As accumulation of contaminants into tissues such as muscle and liver tends to happen over time, and as most often the life histories of the studied individuals are unknown, it is difficult to differentiate where the contaminant burden originated. Contaminant concentrations in blood are often considered to represent recent contaminant exposure (Berglund, 2018) and can possibly provide a more conclusive picture of local contaminant levels. Contaminants can, however, be released into the bloodstream from stored deposits during starvation or fasting (La Merrill et al., 2013; Bustnes et al., 2012). In addition to spatial uncertainties, there can be also temporal uncertainties related to contaminant concentrations in birds. Due to bioaccumulation of contaminants and the long lifespan of birds, contaminants might have been stored in the internal tissues for long periods, meaning that concentrations in these tissues do not represent concentrations in the present environment.

Models of trophic dynamics of contaminants based on TMFs and BMFs often rely on fairly accurate determination of the TP of the species. While differences in the ecology, behaviour, and physiology of species make it impossible to make an exact calculation of the TP of an individual, using $\delta^{15}N$ provides a good proxy for trophic level estimations (Borgå et al., 2012). As I already discussed in the case of the TP of the white-tailed eagle, however, mismatches can occur between TP yielded using $\delta^{15}N$ and TP derived from dietary studies. This mismatch can be due to spatial and temporal mismatches between the $\delta^{15}N$ of the species and the baseline used to derive the TP, to variation in the diet of studied species and individuals, or to the selected TEF not representing the actual trophic enrichment of $\delta^{15}N$ between the predator and its prey. While accurate determination of TP is important for all species of the food web, more unknown variables are present in the case of mobile species such as birds. Inaccurate determination of TP for a species can possibly lead to inaccurate evaluation of trophic magnification and bioaccumulation tendencies of contaminants (Won et al., 2020, Elliott et al., 2021), which may have consequences for regulatory decisions made based on such models. In recent years, use of compound-specific isotope analysis has increased the accuracy of TP estimations compared to those done on bulk $\delta^{15}N$, and in turn has improved the accuracy of trophic magnification models based on these estimates (Won et al., 2020, Elliott et al., 2021).

4 Conclusions

In this thesis, I have studied the state of metal and organohalogen contamination in the northern Baltic Sea coastal food web, focussing on three bird species: the white-tailed eagle, great cormorant, and common eider. In Chapter **I**, I studied whether nestlings of white-tailed eagles and great cormorants act as spatial sentinels for acidic sulphate soil associated metal contamination. I found that nestlings in proximity to acidic sulphate soil areas had higher concentrations of some metals compared to nestlings from control areas, indicating that acidic sulphate soils are a source of certain metals for apex birds. The elevated metals varied between the species, however, likely reflecting differences in the diets of these species. In Chapters **II** and **III**, I studied the trophic dynamics, levels, and profiles of Hg and organohalogen contaminants, including some emerging contaminants, in the Archipelago Sea food web. As expected, Hg biomagnified in the studied food web, and the rate of trophic magnification was higher in the pelagic food chain compared to the benthic food chain when the bird species were excluded from the models. Similarly, TMFs revealed that ∑PCBs, ∑DDTs, HCB, ∑CHLs, and ∑PFSAs biomagnify in the food web; the fit of the models increased when birds were not included. Concentrations of Hg and most organohalogen contaminants in the birds were higher in relation to their TP, as derived using $\delta^{15}N$, than in the fish or invertebrates, which decreased the birds' fit to the linear magnification models. Other studies have similarly reported higher contaminant concentrations in homeotherm species compared to poikilotherm species. These results show that models for biomagnification can be improved by taking into account species traits and ecology.

Across the studies included in this thesis, contaminant concentrations and profiles varied between species. Concentrations were generally highest in the white-tailed eagles, a finding that is consistent with the apex trophic position of this species in the food web. The contaminant concentrations of acidic sulphate associated metals and Hg investigated in Chapters **I** and **II**, respectively, were generally low and not of concern. The concentrations of legacy POPs (**III**) were mostly lower than those reported in the Baltic Sea during last decades, which is consistent with monitoring data. Yet the results of this thesis show that bird species of the Finnish coastal food webs are exposed simultaneously to multiple contaminants, which increases the risk of adverse interaction effects of these contaminants. I also found PFECHS, an emerging PFAS, in the white-tailed eagle, great cormorant, and two fish species. This result shows that, in addition to legacy contaminants, emerging contaminants with largely unknown trophic dynamics are present in the Baltic environment. In the future, the trophic dynamics of such novel contaminants should be studied to better understand their behaviour in the Baltic Sea food web. These studies should consider the characteristics of species in order to better understand the risks that novel contaminants pose to species with differing ecological and ecophysiological traits, as these traits can affect both the outcomes of trophic magnification models and the level of contaminant exposure of the species.

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