Drivers and biological effects of mercury and organo-halogenated chemicals on Arctic predators

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“Science knows no country, because knowledge belongs to humanity, and is the torch which illuminates the world.”

Louis Pasteur

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TABLE OF ABBREVIATIONS

AMAP	Arctic Monitoring and Assessment Program
CECs	Chemicals of emerging concern
DDT	Dichlorodiphenyltrichloroethane
EC	European Commission
EU	European Union
Hg	Mercury
MeHg	Methyl mercury (organic form of Hg with one methyl group)
OHC	Organo-halogenated chemicals
PBT	Persistent, bioaccumulative and toxic
PCB	Polychlorinated biphenyls
PFAS	Per- and polyfluoroalkyl substances
PFOS	Perfluorooctane sulfonic acid
POP	Persistent organic pollutants
REACH	Regulation on the registration, evaluation, authorisation and restriction of chemicals
THg	Total mercury (includes all inorganic and organic mercury forms)
SPV	Subjects per variable
UN	United Nations

SUMMARY

Pollutants have been identified as one of the major environmental threats to humankind, wildlife and ecosystems, including the Arctic. Organo-halogenated chemicals (OHCs) and mercury (Hg) are transported from anthropogenic sources via long-range transport to the Arctic, where many of these pollutants biomagnify within Arctic food chains, leaving in particular apex predators at the top of food chains vulnerable to potential health effects due to their hazardous properties. From a risk assessment perspective, top predators are valuable sentinels because they integrate chemical exposure across large areas and long time spans and, owing to their position at the top of food chains, reflect pollutant trends and ecosystem fluctuations. Thus, monitoring chemical exposure and understanding its impacts on Arctic predators is crucial for developing science-based recommendations for targeted conservation and management initiatives. The overall objective of the thesis was to assess the exposure of key Arctic marine and terrestrial mammal, bird and fish species to OHCs and Hg as well as their biological effects at the individual and population level. Specifically, we investigated dietary drivers of mercury exposure in two sentinel predator species inhabiting the circumpolar Arctic, namely the Arctic subspecies of the grey wolf (*Canis lupus arctos*) and the Arctic fox (*Vulpes lagopus*).

In Chapter I, total mercury levels were analysed in the fur of 30 Arctic grey wolves sampled between 1869 – 1998 in Greenland and High Arctic Canada. Diet was evaluated with the help of stable isotope analysis by determining dietary carbon sources (as inferred by δ^{13}C) and trophic level (as inferred by δ^{15}N). As expected, we observed high dietary variation of marine and terrestrial food sources at various trophic positions. Variability in Hg burden in the wolves was significantly driven by biomagnification (δ^{15}N) rather than by carbon source (δ^{13}C) or study site.

In Chapter II, various pathogens and Hg levels were evaluated as potential drivers of the Arctic fox population decline during 1970 – 1980 on the distinct, small Mednyi Island belonging to the Russian Commander Islands. We chose a study design that allowed the comparison of Hg fur levels from historical specimens from the Commander Islands representative for the population pre-crash period on Mednyi Island, contemporary Mednyi foxes sampled in 2011, and two unrelated and geographically remote ecotypes represented by inland and coastal Icelandic Arctic fox populations. As expected, Hg levels were significantly higher in foxes inhabiting coastal habitats regardless of geographic location than in those from inland sites and were highest during the pre-crash period in the Mednyi population. This was most likely because the Mednyi population depends solely on marine vertebrates that have been shown to be highly

contaminated. Our large-scale health assessment using serological and DNA based pathogen screening techniques suggested a low prevalence of pathogens in contemporary foxes, although it does not allow a proper assessment of the health status of the Mednyi Arctic foxes during or before the crash period. Most likely a complex interplay of stressors explains the high cub mortality observed on Mednyi Island, including high mercury body burden, which renders particularly young foxes vulnerable to infectious diseases.

Chapter III evaluates whether fur can be used as a minimally-invasive sampling matrix to reliably determine total Hg levels in soft tissues, specifically in liver and kidney of Arctic foxes. Associations between Hg levels in the fur, liver and kidney of 35 Arctic foxes sampled in 2011-2012 on Iceland were investigated. Observed total Hg concentrations varied considerably among tissues, with liver generally showing higher levels than fur and kidneys, and significant linear and sex-independent relationships based on regressions allowed to reliably extrapolate mercury liver and kidney concentrations from fur levels. Measurements in ecotoxicological studies frequently use different tissues (usually liver), which hinders cross-study comparison. Thus, the derived regression equations improve direct comparison of Hg levels among fur and soft tissues reported for Arctic canids.

Chapters I-III show that Arctic wolves from Greenland and High Arctic Canada tend to have relatively low Hg concentrations (< 5 µg g^{-1} dry weight), whereas coastal Arctic fox populations from Iceland and the Commander Islands had up to 7-fold higher Hg fur levels, while coastal populations had 3-fold higher Hg levels than inland ones. While the health status of the Arctic wolves was most likely not affected, the mercury levels in some of the Arctic foxes exceed putative thresholds for Hg-mediated toxic health effects. Apart from trophic magnification as major driver, the observed inter-population and intra-population variability in Hg levels likely results from a combination of varying ecosystem conditions, changes in emission patterns and biological factors. Although the results of Chapter I-III are limited in terms of conclusive evidence because historic study material and/or sample sizes were scarce and relatively small, respectively, they do provide novel tools and information on temporal and spatial variation in Hg pollution in these understudied Arctic canids.

In Chapter IV, exposure levels and health impacts of mercury and OHCs were evaluated in key Arctic marine and terrestrial mammals, birds and fish, using population-specific exposure data published between 2010 and 2019. Various pollutant induced health effects were summarized by each endpoint ranging from molecular to individual and population level effects. We identified quantifiable effects on vitamin metabolism, immune functioning, thyroid and steroid hormone balances, oxidative stress, tissue pathology, and reproduction. On this basis we calculated risk quotients in order to estimate critical body burdens specifically as regard to Hg and polychlorinated biphenyls (PCBs) shown to impair reproduction functions in vertebrates. The outcome was that most Arctic marine mammal species are at no or low risk in terms of health effects or reproductive impairments mediated by Hg or PCB exposure. However, for some species at high marine trophic levels, such as polar bear, narwhal and hooded seal, a segment of the population had body burden indicating a high or severe risk of suffering health effects and reproduction impairments. While bird Hg and PCB concentrations were also above toxicity benchmarks in many areas of the marine environment, terrestrial mammals were not at risk – with the exception of the Arctic foxes from Iceland and Commander Islands analysed in Chapter I – III.

Overall, the thesis demonstrates the usefulness of fur samples for monitoring Hg and carbon and nitrogen stable isotopes in Arctic canids. It provides a practical tool for cross-study comparisons of Hg across different tissue types and fur, which will help with interpreting exposure risks of Arctic canids in future studies. It further suggests that absolute exposure to pollutants may be less important than indirect contamination via the feeding ecology and feeding opportunities of canid predators which may in turn affect population health and stability. Although the release of numerous, hazardous OHCs and Hg into the environment have been limited and regulated for a long time, the thesis emphasises that certain Arctic predator species are still highly exposed, which may pose a potential threat to their populations and the integrity of their ecosystems. Since our understanding and assessment methods of the specific risks and chronic impacts of pollutants on wildlife populations are still limited, chemical risk assessment should in future be up-scaled to population level effects.

The results of the thesis will complement the existing data that form the basis for science-based recommendations for conservation management and policy measures and were already considered by the *Arctic Monitoring and Assessment Program*. Finally, to improve the effectiveness of the regulation of the release of pollutants into the environment and to support conservation initiatives for Arctic predators, particularly in view of climate change as a future stressor, further investigations are needed to improve our understanding of the mechanisms and interplay of drivers of pollution and its effects on Arctic wildlife populations and ecosystems.

ZUSAMMENFASSUNG

Schadstoffe stellen derzeit eine der größten Umweltbedrohungen für Menschen, Wildtiere und Ökosysteme einschließlich in der Arktis dar. Halogenierte organische Chemikalien (OHC) und Quecksilber (Hg) werden von anthropogenen Quellen über weite Entfernungen in die Arktis transportiert, wo sich viele von ihnen in den arktischen Nahrungsketten anreichern und dadurch die Gesundheit von Spitzenprädatoren gefährden aufgrund ihrer schädlichen Wirkungen. Aus regulatorischer Perspektive sind Spitzenprädatoren wertvolle Indikatoren für Schadstoffbelastung, da sie große Flächen und Zeiträume abdecken und als Endglieder der Nahrungskette besonders gut Trends der Schadstoffbelastung und Fluktuationen innerhalb von Ökosystemen abbilden. Daher ist die Überwachung der chemischen Belastung und das Verständnis ihrer Auswirkungen auf arktische Prädatoren von entscheidender Bedeutung für die Entwicklung wissenschaftlich fundierter Empfehlungen für gezielte Schutz- und Managementinitiativen.

Das übergeordnete Ziel dieser Doktorarbeit war es den Grad der Belastung mit OHC und Hg sowie deren biologische Wirkungen auf verschiedene arktische marine und terrestrische Säugetier-, Vogel- und Fischarten auf Organismen- und Populationsebene zu untersuchen. Insbesondere mögliche nahrungsbedingte Einflüsse auf die Quecksilberbelastung wurden in zwei arktischen Prädatoren näher untersucht, dem Polarwolf als arktischen Unterart des Wolfes (*Canis lupus arctos*) und dem Polarfuchs (*Vulpes lagopus*).

In Kapitel I analysierten wir den Gesamtquecksilbergehalt im Fell von 30 Polarwölfen, die zwischen 1869 und 1998 in Grönland und in der kanadischen Hocharktis gesammelt wurden. Zudem ermittelten wir die Nahrungsökologie der Wölfe anhand von stabilen Isotopen, indem die Kohlenstoffquellen der Nahrung (abgeleitet aus $\delta^{13}C$) und die Nahrungsnetzebene (abgeleitet aus $\delta^{15}N$) bestimmt werden. Wie erwartet, fanden wir bei den Wölfen eine hohe Variabilität der Beutearten, die sich sowohl von marinen als auch terrestrischen Nahrungsquellen ernährten. Die Variabilität der Hg-Belastung der Wölfe wurde maßgeblich durch die Anreicherung in den Nahrungsnetzen (δ15N) und weniger durch die Kohlenstoffquelle (δ13C, terrestrisch versus marine) oder die geographische Lage bestimmt.

In Kapitel II wurden verschiedene Krankheitserreger und Hg-Gehalte als mögliche Erklärungsfaktoren für den Populationsrückgang der Polarfüchse zwischen 1970 und 1980 auf der kleinen, abgelegenen Mednyi-Insel untersucht, die zu den russischen Kommandeur Inseln gehört.

Dafür wählten wir ein Studiendesign, das einen Vergleich der Hg-Fellkonzentrationen zwischen heutigen Polarfüchsen aus dem Jahr 2011 und historischen Polarfüchsen aus der Zeit des Populationsrückganges auf Mednyi erlaubte. Zudem wurden die Quecksilbergehalte der Füchse von den Kommandeur Inseln mit zwei geografisch weit entfernten Polarfuchspopulationen auf Island verglichen, die entweder in Inland- oder an der Küstenhabitaten lebten. Wie erwartet waren die Hg-Konzentrationen bei den Füchsen der Küstenhabitate signifikant höher im Vergleich zu den Inlandfüchsen unabhängig von der geographischen Lage, am höchsten jedoch in der Zeit vor dem Zusammenbruch der Mednyi-Population. Dies ist sehr wahrscheinlich darauf zurückzuführen, dass sich diese Fuchspopulation ausschließlich von marinen Wirbeltieren ernährte, die ebenfalls stark kontaminiert waren. Ein groß angelegtes Gesundheitsmonitoring der Fuchsproben, das auf serologischen und DNA-basierten Pathogen-Screening-Techniken basiert, deutet auf eine geringe Prävalenz von Krankheitserregern bei den heutigen Füchsen hin, lässt jedoch keine Rückschlüsse auf den Gesundheitszustand der Tiere während oder vor der Zeit des Populationseinbruchs zu. Höchstwahrscheinlich erklärt ein komplexes Zusammenspiel verschiedener Stressfaktoren einschließlich einer erhöhten Hg-Exposition, die die Jungfüchse besonders anfällig für Infektionskrankheiten und Zoonosen machen, die beobachtete hohe Jungtiersterblichkeit auf Mednyi in den Siebziger Jahren.

Kapitel III untersuchte, ob Fell als minimal-invasive Probenmatrix verwendet werden kann, um den Hg-Gehalt in Leber und Niere von Polarfüchsen zuverlässig zu bestimmen. Dafür wurden Korrelationen zwischen den Hg-Gehalten im Fell, in der Leber und in den Nieren von 35 Polarfüchsen untersucht, die zwischen 2011 und 2012 auf Island beprobt wurden. Obwohl die beobachteten Hg-Konzentrationen in den verschiedenen Geweben stark variierten, wobei die Leber im Allgemeinen höhere Werte aufwies als das Fell und die Nieren, konnte eine signifikante lineare und geschlechtsunabhängige Korrelation zwischen den Quecksilberwerten im Fell, Leber und Nieren nachgewiesen werden. Ökotoxikologischen Studien verwenden oft unterschiedliche Gewebe (in der Regel Leber), was einen studienübergreifenden Vergleich der Hg-Konzentrationen erschwert. Daher können die die abgeleiteten Regressionsgleichungen den direkten Vergleich der Hg- Gehalte in Fell und Organen von Polarfüchsen in zukünftigen Untersuchungen erleichtern.

Kapitel I-III zeigen, dass Wölfe aus Grönland und der kanadischen Hocharktis relativ niedrige Hg-Fellkonzentrationen aufwiesen (< 5 µg g^{-1} Trockengewicht), während Küstenpopulationen der Polarfüchse aus Island und den Kommandeur Inseln bis zu siebenfach höher waren. Dabei waren die Hg-Fellkonzentrationen von Polarfüchsen aus Küstenregionen dreifach höher als die von Füchsen aus dem Landesinneren. Während der Gesundheitszustand der untersuchten Wölfe höchstwahrscheinlich nicht beeinträchtigt wurde, überschritten die Quecksilberwerte in einigen der untersuchten Polarfüchsen die mutmaßlichen Schwellenwerte für Hg-vermittelte toxische Gesundheitseffekte. Abgesehen von der Anreicherung in Nahrungsnetzten als Treiber war die beobachtete Variabilität der Hg-Konzentrationen zwischen und innerhalb der beobachteten Populationen wahrscheinlich das Ergebnis einer Kombination aus unterschiedlichen Ökosystembedingungen, Änderungen der Emissionsmuster sowie weitere abiotische und biotische Faktoren. Auch wenn die Ergebnisse der Kapitel I-III aufgrund des heterogenen jedoch einzigartigen historischen Studienmaterials und/oder des relativ geringen Stichprobenumfangs teilweise begrenzt bleiben, liefern sie neue Informationen über die zeitliche und räumliche Variation der Hg-Belastung in den beiden bisher wenig untersuchten Spezies.

In Kapitel IV wurden die Schadstoffexposition und die gesundheitlichen Auswirkungen von Hg und OHC in marinen und terrestrischen Säugetieren, Vögeln und Fischen der Arktis beschrieben anhand einer Metaanalyse der Literatur zwischen 2010 und 2019. Verschiedene schadstoffinduzierte Gesundheitseffekte wurden dafür für jeden Endpunkt zusammengefasst, die von molekularen bis hin zu individuellen und populationsbezogenen Effekten reichten. Wir haben quantifizierbare Auswirkungen auf den Vitaminstoffwechsel, die Immunfunktion, den Schilddrüsen- und Steroidhormonhaushalt, den oxidativen Stress, die Gewebepathologie und die Reproduktion festgestellt. Zudem berechneten wir die Risikoquotienten, um die kritische Körperbelastungen für die Hg und polychlorierte Biphenyle (PCBs) abzuschätzen, die potentiell die Reproduktion in Vertebraten beeinträchtigen können. Dies ergab, dass für die meisten arktischen Meeressäugerarten kein oder nur ein geringes Risiko für gesundheitliche Auswirkungen durch Hg- oder OHC-Exposition bestand. Bei einigen Arten hoher trophischen Ebenen, wie Eisbär, Narwal und Mützenrobben, bestand jedoch für einen Teil der Population ein hohes oder schwerwiegendes Risiko für die Beeinträchtigung der Gesundheit- und der Reproduktion. Während die Hg- und OHC-Konzentrationen bei Vögeln in vielen Bereichen der Meeresumwelt ebenfalls über den Schwellenwerten lagen, waren terrestrische Säuger mit Ausnahme der in Kapitel I-III analysierten Polarfüchse aus Island und den Kommandeursinseln nicht gefährdet.

Insgesamt zeigt diese Arbeit den Nutzen von Fellproben, um Quecksilber und stabilen Kohlenstoff und Stickstoffisotopen in Polarfüchsen und Wölfen zu bestimmen. Sie bietet ein praktisches

Instrument für den studienübergreifenden Vergleich von Hg in verschiedenen Gewebetypen und Fell, was wiederum die Interpretation von Expositionsrisiken arktischer Caniden in zukünftigen Studien verbessert. Die Ergebnisse deuten darauf hin, dass die absolute Schadstoffexposition weniger wichtig ist als die Ernährungsökologie und das Nahrungsangebot von Polarfüchsen und Wölfen, was wiederum die Gesundheit und Stabilität der Populationen beeinflussen könnte. Obwohl zahlreiche gefährliche OHC und Hg seit langem reguliert sind, verdeutlicht die Arbeit, dass bestimmte Prädatorarten der Arktis immer noch stark exponiert sind, was eine potenzielle Bedrohung für ihre Populationen und die Integrität der Ökosysteme darstellt. Da unser Verständnis und unsere Bewertungsmethoden für die spezifischen Risiken und chronischen Auswirkungen von Schadstoffen auf Wildtierpopulationen noch sehr begrenzt sind, sollte die ökotoxikologische Risikobewertung in Zukunft stärker Effekte auf Populationsebene berücksichtigen. Die Ergebnisse der Arbeit erweitern die Wissensbasis, die die Grundlage für wissenschaftlich fundierte Empfehlungen für Schutzmaßnahmen bilden und bereits vom *Arctic Monitoring and Assessment Program* berücksichtigt wurden. Um die Effektivität des Chemikalienmanagements und das Schutzinitiativen für arktische Prädatoren zu verbessern, insbesondere im Hinblick auf den Klimawandel als zukünftigen Stressor, sind weitere Untersuchungen notwendig, die die Mechanismen und das Zusammenspiel von Schadstoffbelastung und deren Auswirkungen auf arktische Wildtierpopulationen und Ökosysteme stärker beleuchten.

1. GENERAL INTRODUCTION

1.1 TERMINOLOGY

In ecotoxicological literature the terms pollutant, hazardous chemical and contaminant are often inconsistently used. While contamination describes simply the presence of a substance where it should not be or at concentrations above background, (chemical) pollution is contamination that results or can result in adverse biological effects to organisms (Chapman, 2007). Thus, all pollutants are contaminants, but not all contaminants are pollutants because substances introduced into the environment may be more or less bioavailable to organisms depending on their chemical form, modifying factors in the environment, the environmental compartment they occupy, and the reactions (behavioural and physiological) of exposed biota (Chapman, 2007; Chapman et al., 2003). Accordingly, in this thesis I use the term pollutant for chemicals proven as being hazardous to organisms based on laboratory data or field studies. In contrast, the term contaminant hereafter relates to all anthropogenic chemicals which were shown to be present in different environmental media and/or biota but lack ecotoxicological data to evaluate potential hazards. While hazard refers to the intrinsic adverse properties of a chemical like toxicity, persistency or bioaccumulation, risk considers hazards in conjunction with exposure and describes the probability of an adverse outcome (KEMI, 2020). Exposure to a chemical means how and in which concentration an organism get in contact with the chemical (KEMI, 2020). The term chemicals of emerging concern (CECs), is hereafter used for chemicals which may vary widely in terms of their properties and functions but share two common characteristics: they are unregulated, and regulators and the scientific community cannot guarantee that they are not hazardous to organisms either due to lack of effect data or due to inconclusive ecotoxicological assessment outcome (Diamond and Burton, 2021).

1.2 THE ARCTIC – ONE OF THE FASTEST CHANGING ECOSYSTEMS IN THE ANTHROPOCENE

The Arctic is the northernmost region of the planet, consisting of the Arctic Ocean, adjacent seas, and parts of Alaska (United States), Canada, Russia, Finland, Norway, Sweden, Iceland and Greenland (Denmark). Land within Arctic regions has seasonally varying snow and ice cover, with treeless permafrost containing tundra dominating, whereas Arctic seas contain seasonal sea ice in many places. Arctic regions are characterized by a harsh climate and extreme variation in light and temperature, short summers, extensive snow and ice cover in winter and large areas of permafrost (AMAP, 1997). The Arctic can be roughly divided into three regions, the High Arctic, which appears as polar desert, the Low Arctic, which corresponds to the tundra, and the adjacent Sub-Arctic regions between 50°N and 70°N latitude (AMAP, 1997; Meltofte, 2018, Figure 1). The tundra of the Low Arctic gradually turns into the sub-Arctic zone, with richer vegetation and wildlife. In general, the Low Arctic has much more vegetation than the High Arctic, where large lowland areas may be almost devoid of vegetation (Meltofte, 2018).

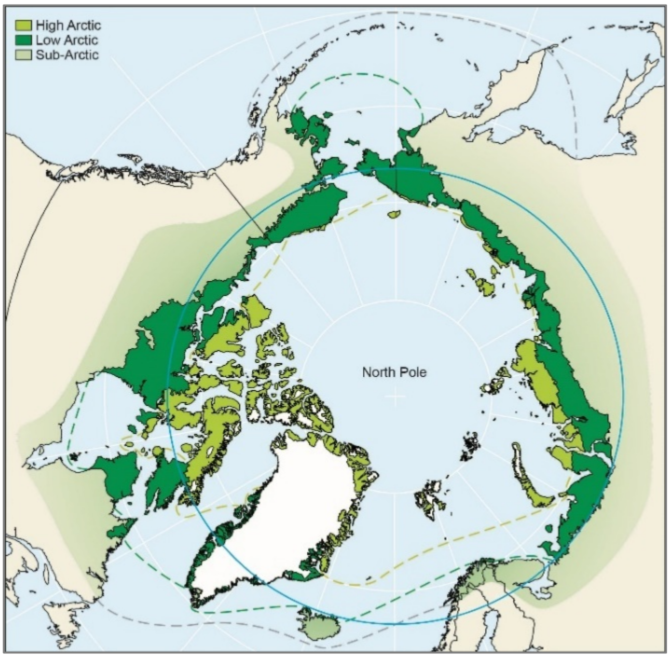

Figure 1. Map of the top of the northern hemisphere with the High (light green) and Low Arctic zones (dark green) together with a tentative demarcation of the sub-Arctic (beige). Dashed lines indicate continuation of the three zones in the ocean. The Arctic Circle is shown as blue line. Figure taken with permission from CAVM Team (2013) and Meltofte (2013).

Arctic biota contain more than 21.000 species of mammals, birds, fish, plants and fungi (Thomas, 2020) and have a relatively low mammalian diversity. The vegetation is composed of plants such as dwarf shrubs, graminoids, herbs, lichens, and mosses, which all grow relatively close to the ground, forming the tundra (AMAP, 1997; Thomas, 2020). Herbivores on the tundra include for instance the Arctic hare (*Lepus arcticus)*, lemming (*Lemmus ssp.*), muskox (*Ovibos moschatus)* and caribou (*Rangifer tarandus)*, which are preyed upon by arctic foxes (*Vulpus lagopus*) and Arctic wolves (*Canis lupus arctos*). In contrast, polar bears (*Ursus maritimus*) feed predominantly on marine prey species, e.g., ringed seals (*Pusa hispida*, Thiemann et al., 2008). Most birds only come north during the summer and few are true terrestrial birds, such as the rock ptarmigan (*Lagopus* *muta*) which occurs even in the High Arctic zone (AMAP, 1997). Marine mammals seasonally occurring in the Arctic include several species such as ringed seal, harbour seal (*Phoca vitulina*), hooded seal (*Cystophora cristata*), narwhale (*Monodon monoceros*), beluga whale (*Delphinapterus leucas*), pilot whale (*Globicephala melas*) and killer whale (*Orcinus orca)*.

The Arctic is warming at a faster rate than any other area in the world, and this rapid warming has profound effects on Arctic environments (AMAP, 2021a; Tseng, 2021), with cascading effects on individuals, populations and communities (Dietz et al., 2021). Rapid warming causes precipitous sea-ice loss, with consequences on the distribution and dietary ecology of species (Kortsch et al., 2015; Overland and Wang, 2013). Increasing temperatures, the retreat of sea ice, thawing permafrost, changing sea ice regimes, shrinking of glaciers and changes in precipitation patterns can all affect how pollutants distribute within the Arctic environment and subsequently affect Arctic ecosystems (Hung et al., 2022). For instance, changes in sea ice cover affect the exchange of mercury between seawater and the atmosphere as well as the amount of mercury that can be stored or released by the sea ice in summer (Schartup et al., 2022). Also changes in food web composition will contribute to changes in environmental partitioning and accumulation of many pollutants (Hung et al., 2022). Multiple stressors in the Arctic environment include pollution, climate change, shipping, and pathogens. All play a significant role in the reshaping of Arctic biodiversity and ecosystems (Thomas, 2020; Townhill et al., 2022) and can thus affect populations in terms of their size and viability. Yet assessments of the potential combined effects of human activities are limited by incomplete data, particularly with respect to pollutants (Townhill et al., 2022).

1.3 ARCTIC CHEMICAL POLLUTION

Chemical pollution has become one of the Anthropocene's greatest environmental challenges (UN, 2019). By now, humanity has likely exceeded safe thresholds of planetary boundaries for chemical pollution, with humans and wildlife in Europe being exposed to a wide range of types of chemical pollution (Persson et al., 2022; Rockström et al., 2009). These pollutants include plant protection products, biocides, pharmaceuticals, industrial chemicals and personal care products together with numerous transformation products, which pose substantial threats to human and environmental health as well as to biodiversity. The same holds true also for the Arctic, where exposure to pollutants such as mercury has been recognized as a health concern in humans and Arctic wildlife for several decades (Dietz et al., 2019; Dietz et al., 2022; Rigét et al., 2019; Sonne et al., 2021).

Mercury (Hg) in the Arctic

The thesis focusses in particular on mercury (Hg) exposure and related health implications in Arctic predators. Mercury is a global pollutant because of its ability to undergo long-range transport from source regions to remote parts of the world such as the Arctic, its ubiquitous presence in aquatic ecosystems and food webs (Fitzgerald et al., 1998), and is also detrimental to the health of human populations worldwide (Barst et al., 2022; Basu et al., 2022; Eagles-Smith et al., 2018; Karagas et al., 2012). In the Arctic, elevated concentrations of Hg exceeding toxicological thresholds have been reported in both humans and wildlife (AMAP, 2011; Dietz et al., 2013b).

There are several chemical forms of Hg. Elemental mercury (Hg^0) is a heavy, shiny, silver liquid and the only metal that is liquid at room temperature. Mercury falls into two general groupings − inorganic (Hg^0, Hg^{2+}, Hg^+) and organic forms, predominantly methyl mercury (MeHg). Studies on environmental concentrations of Hg usually measure total mercury, which is the sum of all Hg forms in a sample (elemental, inorganic and organic). Methylmercury is formed by methylation through bacterial processes in sediments and the water column of large water bodies (Hamdy and Noyes, 1975) such as large lakes and oceans. The potential for Hg to transfer between chemical forms presents a challenge to predicting Hg trends and toxicity within ecosystems because processes of oxidation, reduction and methylation are strongly affected by environmental conditions such as solar radiation and intensity of organic carbon processing (Brown et al., 2018). New analytical techniques, such as Hg stable isotope characterization, provide novel insights into sources and transformation processes of Hg and its forms.

Previous research indicates that MeHg biomagnification occurs in Arctic food webs via uptake by primary producers and subsequent consumption by invertebrates, fish, birds and marine mammals and terrestrial carnivores (e.g. Atwell et al., 1998). This is because particularly MeHg is lipophilic and therefore bioaccumulates in tissues of organisms due to the high affinity to protein thiol groups, following absorption in the gut, but is slowly eliminated from the body (Hammerschmidt and Fitzgerald, 2006; Harris et al., 2007). Under most conditions, in fish and wildlife uptake of MeHg occurs primarily through the diet (Scheuhammer et al., 2007). Because of biomagnification of MeHg, long-lived predatory animals feeding in aquatic and terrestrial food chains are at great risk for elevated dietary MeHg exposure, accumulation, and toxicity, while species that feed primarily on fish or on other piscivorous species, are in many cases at highest risk (Chételat et al., 2020; Scheuhammer et al., 2007). Accordingly, marine mammals such as seals, beluga, and polar bears have generally higher Hg body burden than terrestrial mammals and freshwater fish (Fisk et al., 2003; Muir et al., 1999).

Mercury has been released for a long period dating back even to Roman times (López-Costas et al., 2020). More pertinent to the present, Hg has been released in large quantities from multiple sources during the past 60 years. Mercury released by historical human activities and now deposited in soils and the oceans acts as a reservoir by being available for re-emission to air, thereby increasing atmospheric mercury concentrations and maintaining them at higher levels than would be the case if current emissions were the only source (UN, 2019b). For instance, recent modelling indicates that mining since the 16th century accounts for about two-third of all anthropogenic mercury currently in the oceans (UN, 2019b). The remaining third of anthropogenic mercury input to oceans has come since then, mainly from coal combustion and other industrial activities. Of global aquatic Hg releases, around 50% are estimated to occur in China and India, where Hg drains into the West Pacific and North Indian Oceans (Obrist et al., 2018). At present, the main sources of global Hg release (60%) are mercury production, small-scale gold mining and coal combustion as well as municipal waste incineration (Beckers and Rinklebe, 2017; Driscoll et al., 2013; Obrist et al., 2018; Streets et al., 2019, 2017, UN 2019). Over 98% of atmospheric Hg is emitted outside the region and is transported to the Arctic via long-range air and ocean transport (Dastoor et al., 2022). Since Hg also occurs naturally in the earth's crust and in the environment, it is also released by degassing through volcanoes and wildfires (Pirrone et al., 2010). Consequently, each year an estimated 4400 tons of Hg in its various chemical forms are emitted to water, air and soil by all sources (UN, 2019). The Arctic has been considered as a sink for Hg deposition, with the highest deposition observed in the European part of the Arctic, downwind of major anthropogenic source regions (Ariya et al., 2004) .

Regulation of Hg emission and temporal changes of Hg concentrations in the Arctic

Because of the high Hg concentrations in the Arctic and elsewhere worldwide, anthropogenic releases of Hg are now globally regulated by the Minamata Convention on Mercury, which was ratified in 2017 (www.mercuryconvention.org). The Convention signalled a commitment by government signatories from around the world to reduce the use and environmental release of Hg in order to protect human health and the environment. Anthropogenic activities have cumulatively increased environmental mercury concentrations globally over the past centuries (UN, 2019b). Bearing in mind the uncertainties in natural and anthropogenic emission estimates, and the many deficiencies in our understanding of the processes and flux rates governing Hg transport and fate between the air, soil and ocean compartments, the best information currently available suggests that the increase in atmospheric Hg concentrations has driven a ~ 310% increase in average deposition rates to the Earth's surface (AMAP/UN Environment, 2019) . Surface marine waters have shown a 230% increase in Hg concentrations above natural levels. The increase in surface soils (~15%) has been much lower (Basu et al., 2022). Global models on Hg emissions and cycling predict that Hg clearance rates will be slow relative to the rate of anthropogenic emission reductions in future due to long resident times in deep waters, sediment and soil, such that removal of anthropogenic Hg from the world's oceans and marine food chains will take many decades to centuries (reviewed by AMAP/UN Environment, 2019). This emphasizes the need for prompt and ambitious action to reduce mercury concentrations worldwide, including the Arctic.

Temporal trends of mercury in Arctic wildlife species were found to vary by region and by the magnitude of the time period chosen for analysis (AMAP, 2021a; AMAP/UN Environment, 2019; Gamberg et al., 2005; Morris et al., 2022; Rigét et al., 2011). For instance, the recent AMAP analysis of 110 time series of Hg concentrations revealed 14 decreasing and 17 increasing significant changes in various Arctic biota species including freshwater, terrestrial and marine mammals, birds and fish (AMAP, 2021a; Morris et al., 2022). Furthermore, numerous examples of time-trend datasets in Arctic and non-Arctic biota did not agree with the trends in regional atmospheric deposition or concentrations over recent decades (reviewed by AMAP/UN Environment, 2019). The exact reasons for the large variability and lack of consistency between trends in Hg emissions and biota levels remained unclear but might be related to changes in food availability, the deposition and concentrations of Hg in air over time (MacSween et al., 2022), transport and fate of Hg, and the impacts of climate change on abiotic and biotic systems (Chételat et al., 2022; Dastoor et al., 2022; Jonsson et al., 2022; McKinney et al., 2022). As Arctic wildlife is exposed to Hg and MeHg primarily through their diet, environmental changes and drivers, that affect Arctic food web structure and composition particularly affect Hg biota levels and trends over time (AMAP, 2021a; Hudelson et al., 2019; Jonsson et al., 2022; Lehnherr, 2014; Loseto et al., 2015; McKinney et al., 2022; Morris et al., 2022; Schartup et al., 2020). More specifically, Hg concentrations in Arctic biota are generally controlled by interlinked processes which act on one or both of the primary regulating factors. These are the size of the abiotic pool of Hg available for bioaccumulation, and the transfer of MeHg through food webs (St. Louis et al., 2011; Kirk et al., 2012; Lehnherr, 2014; Loseto et al., 2008). The size of the abiotic Hg pool is mainly determined by the atmospheric delivery of inorganic Hg to Arctic aquatic ecosystems, riverine, and oceanic sources, sea ice cover and methylation efficiency within Arctic ecosystems (Lehnherr, 2014).

Organo-halogenated chemicals (OHCs) in the Arctic

The use of an increasing number of chemicals over the last century has resulted in high environmental emissions and wildlife exposures (Chiaia-Hernández et al., 2020; Hollender et al., 2019; Persson et al., 2013). By now over 350.000 chemicals and mixtures of chemicals have been registered for global production and use, up to three times more than previously estimated (Wang et al., 2020). In most cases, anthropogenic contaminants reach the marine and freshwater environment from land-based sources, such as industrial activities, urban and riverine inputs. Many pollutants like per- and polyfluoroalkyl substances (PFAS), UV-filters or aromatic hydrocarbons are also released directly into the marine environment, e.g. by ship activities (Tornero and Hanke, 2016) and into the air, e.g. by direct emissions from paper mills (Dionne and Walker, 2021) or through volatilization during waste water treatment (Shoeib et al., 2016; Wang et al., 2012).

Organo-halogenated chemicals (OHCs) are a large class of natural and synthetic chemicals that contain one or more halogens (fluorine, chlorine, bromine, or iodine) combined with carbon and other elements. The groups of OHCs include for instance polychlorinated biphenyls (PCBs), organochlorine pesticides and many industrial chemicals such as brominated flame retardants and PFAS. Many OHCs are classified as persistent organic pollutants (POPs) which are regulated under the Stockholm Convention ("The new POPs under the Stockholm Convention", 2022). This is because they only slowly degrade or do not degrade at all in the environment, particularly under cold conditions (Letcher et al., 2018), accumulate in organisms and food chains, are transported to remote areas via long range transport in the air and freshwater or seawater, and adversely affect human and environmental health (de Wit et al., 2010, 2020). Since within the circumpolar Arctic there is very little direct production and use of OHCs, they typically do not originate from local sources in the Arctic but from global industrial and anthropogenic use, such as agricultural pest control, textiles, or firefighting foams (de Wit et al., 2010). Accordingly, POPs and their precursors, degradation products and metabolites are carried into the Arctic from more southerly latitudes via long-range atmospheric transport, ocean currents and rivers (Braune et al., 2015, 2005; de Wit et al., 2006; Letcher et al., 2018). This transport has been shown to depend on both the physical–chemical properties of the chemicals and the environment they encounter (Wania and Mackay, 1996). For instance, Wania (2003) predicted that those persistent substances which are both relatively volatile and water soluble or hydrophobic are most likely to accumulate in Arctic ecosystems. Some aquatic pollutants enter the Arctic via local wastewater and waste from settlements, riverine nutrient inputs caused by thawing permafrost and erosion (Tank et al., 2012), emissions from increasing tourism and shipping, commercial fisheries, and chemical and waste emissions from resource exploitation including mining and the extraction of minerals, oil and gas (AMAP, 2018). As with methyl mercury, the lipophilic pollutants to which many OHCs belong, bioaccumulate in Arctic organisms and biomagnify through food webs, reaching levels of concern for the health of exposed wildlife, particularly for top predators (Dietz et al., 2019; Gabrielsen, 2007; Sonne, 2010; Vorkamp and Rigét, 2014). This is mainly because Arctic wildlife species rely on energy-rich fatty tissues as their main energy source, which is prone to contain high levels of lipophilic pollutants and contaminants (Butt et al., 2010; Dietz et al., 2013b; Houde et al., 2011).

Regulation of OHC emission and temporal changes of OHC concentrations in the Arctic

Governmental and intergovernmental organizations such as the European Commission (EC, 2020, 2019) and the United Nations Environment Programme (UNEP, 2017) have developed strategies and enacted legally binding regulations and multilateral agreements to control and manage chemical pollution to foster a toxic-free environment. After authorisation of most POPs beginning in the 1930s, national legislation and voluntary substitutions by the industry have phased out a minor part of POP emissions in the 1970s. This was followed by continuous global regulation through the United Nations Stockholm Convention on POPs starting in 2004 (UNEP, 2004). New POPs recently added to the Stockholm list include Perfluorooctane Sulfonic Acid (PFOS) and short-chained chlorinated paraffins ("The new POPs under the Stockholm Convention.," 2022). Over the last two decades, the concentrations of highly toxic polychlorinated biphenyls (PCBs), organochlorine pesticides and PFASs have remained essentially unchanged or even increased, e.g. in polar bears inhabiting pollutant hotspots, such as Greenland and the Hudson Bay and its hinterland (reviewed by: Hung et al., 2022; Muir et al., 2019; Sonne et al., 2021; Wong et al., 2021). A general decrease in concentrations of many legacy substances, including POPs, has been documented in Arctic biota, which has been traditionally interpreted as a consequence of emission history and regulation (Andersen et al., 2015; de Wit and Muir, 2010; Dietz et al., 2013a; Rigét et al., 2019, 2011). However, temporal changes in Arctic wildlife are often not consistent, particularly with regard to non-regulated OHCs, which has in part be attributed to a complex interplay of several factors, including climate change, fluctuations in emission patterns of chemicals, changes in food web structures and species interactions and other factors (AMAP, 2016; Braune et al., 2005; Brown et al., 2018; Houde et al., 2011; Derek Muir et al., 2019).

Apart from POPs thousands of new chemicals are now in commerce that are predicted to have long-range transport and persistence properties similar to some of the legacy POPs (de Wit et al., 2019; Muir et al., 2019). Adverse effects of certain emerging chemicals on humans and wildlife at different trophic levels have already been documented for some of these compounds also present in Artic environments (de Wit et al., 2019; Sonne et al., 2021, 2020) such as PFASs (Ankley et al., 2021; Chen et al., 2021) or chlorinated paraffins (González-Rubio et al., 2020; Vorkamp et al., 2019). As with mercury, top predators are at particular risk to suffer health effects due to biomagnification along the food webs they are part of (AMAP, 2018; Sonne et al., 2021; Vorkamp and Rigét, 2014).

1.4 MONITORING CHEMICALS AND DIETARY TRACERS IN ARCTIC TOP PREDATORS

Top predators such as raptors and marine and terrestrial mammals are excellent indicator species for persistent bioaccumulative chemicals because (i) they integrate chemical signatures across space and time, including entire biological communities, (ii) have relatively high and easily measured chemical concentrations and (iii) are consumed by people or can be compared with people who consume the same kind of wild foods (Burger and Gochfeld, 2004; Elliott and Elliott, 2013). In that sense arctic top predators are sentinels for humans as they consume some of the same foods that we do, such as fish, and act as potential vectors for pollutant transfer to humans if they are harvested for human consumption (Sonne et al., 2017). This was highlighted in the 2018 UN Global Mercury Assessment (UN, 2019), which demonstrated that *'Arctic populations who consume fish and marine mammals'* were one of four priority groups of concern (Basu et al., 2018). Thus, studying and understanding the effects of global change and pollution on Arctic wildlife is crucial for developing science-based recommendations to protect both people and vulnerable Arctic species by targeted conservation and management initiatives. Some apex mammalian predators such as the polar bear and the Arctic fox are commonly used as sentinel species to detect such environmental changes or document the spatial-temporal changes in pollutant exposure because of their relatively long lifespans and the integration of chemical exposure across their food chains over larger geographical areas and time periods (Burger and Gochfeld, 2001; Harley et al., 2016). Population declines of top predators have been demonstrated to be amongst the most tangible impacts of chemical pollution, and have driven public pressure to enact treaties aimed at reducing such pollution (Best et al., 2010; Bierregaard et al., 2014). Most ecotoxicological studies have focused on marine mammal species, whereas there are few insights into contaminant levels and effects on Arctic terrestrial wildlife (AMAP, 2018; Dietz et al., 2019; Dietz et al., 2022) from a few ecotoxicological studies on Arctic foxes (Andersen et al., 2015; Fuglei et al., 2007; Hallanger et al., 2019; Pedersen et al., 2015).

In this thesis, we use Artic wolves and Arctic foxes as sentinels representing circumpolar predators which feed across different ecosystems to evaluate the degree and drivers of mercury burden in the two model species, which inhabit the yet poorly documented terrestrial Arctic environment (Chapter I – III, (Bocharova et al., 2013; Treu et al., 2022a, 2018). We then go one step further by gathering reported exposure levels of OHCs and Hg and related pollutant induced health effects on various Arctic key species, including fish, birds and mammals (Chapter IV, Dietz et al., 2019).

Arctic wolves (Canis lupus ssp.)

Figure 2. Arctic wolf from Daneborg, Greenland (with permission by Morten Petersen).

The grey wolf (*Canis lupus*) has one of the largest distribution ranges of all terrestrial mammals (Mech, 1970). Despite some taxonomic controversy regarding sub-species delineation, both cranial measures (Krizan, 2005) and genetic data (Carmichael et al. 2008) separate Arctic wolves from southern populations (Nowak 2003). Arctic wolves are distinguished from southern wolves by their smaller size, whiter coloration and narrower braincase (Mech, 2007, Figure 2).

Arctic wolves are concentrated along the northern and eastern coastlines of Greenland, as well as the northernmost parts of North America and the Canadian Arctic Islands (Nowak, 1995). As opportunistic carnivores, Arctic wolves feed mainly on muskoxen (*Ovibos moschatus*), caribou (*Rangifer tarandus*), lemmings (*Dicrostonyx groenlandicus*), Arctic hares (*Lepus Arcticus*), geese and other birds, insects and human-generated rubbish (Dalerum et al., 2018; Marquard-Petersen, 1998). The diet of wolves may vary spatially and temporally across the landscape in response to variation in the distribution, density and seasonal availability of local food resources (Dalerum et al., 2018; Lafferty et al., 2014). For example, studies from Alaska and coastal British Columbia demonstrated that wolves may augment their diet with non-ungulate prey such as seasonally abundant salmon (*Onchorynchus* spp., Adams et al., 2010). Arctic wolves are not threatened by human hunting or persecution; the greatest threat has been proposed to be climate change (WWF, 2021). In contrast to Arctic foxes and grey wolves from other areas, generally very little is known about the ecology and pollutant levels in Arctic wolves (Dalerum et al., 2018). For instance, for High Arctic environments, publications are restricted to three studies from Greenland (Marquard-Petersen, 2021, 2012, 2009), and two studies from the Hall Basin (Dalerum et al., 2018; Mech and Adams, 1999). Therefore, Chapter I studies mercury concentrations and potential dietary ecological drivers in High Arctic wolves as sentinels to assess the poorly documented terrestrial environment. Specifically, we investigated the link between total mercury concentrations and dietary proxies of trophic position ($\delta^{15}N$) and carbon source ($\delta^{13}C$) in the fur of 30 Arctic wolves collected between 1869 and 1998 in the Canada High Arctic and Greenland.

The Arctic fox (Vulpes lagopus)

Figure 3. Arctic fox pups in summer fur (with permission by Mike Boylan, U.S. Fish and Wildlife Service)

The Arctic fox is a medium-sized canid with a circumpolar distribution in the northern hemisphere (Berteaux et al., 2017, Figure 3), which breeds north of the tree line on the Arctic tundra in North America and Eurasia and on the alpine tundra in Fennoscandia (Macpherson, 1969). It was chosen as a climate change flagship species by the International Union of Conservation of Nature (IUCN), and it is decreasing in and retracting from the southern part of its range because of increased competition with red foxes, decreased prey abundance and habitat loss (Berteaux et al., 2017; Ims, 2009). In the wild, most individuals do not survive the first year. Adult lifespan may reach up to 11 years (Pagh et al. 2009).

Arctic fox populations vary dramatically in terms of threats to the viability of their populations. On the Russian Commander Islands (Mednyi and Bering Islands) located at the western end of the Aleutian Island Arc, Arctic fox populations have been isolated since the Pleistocene (Zalkin, 1944). On these islands habitat conditions differ sharply from those on the continent. The fox habitat on Bering Island is somewhat different; it is larger than Mednyi, inhabited by people and offers a greater variety of food sources (Zagrebel'nyi, 2000). On Bering Island, Arctic foxes also feed on terrestrial prey such as reindeer and voles, whereas on Mednyi Island foxes rely solely on marine food items as they are relatively stable throughout the year (Goltsman et al., 2005; Volodin et al., 2013). The foxes on Mednyi Island suffered a drastic decline in population size in the late 1970s but no such phenomenon was observed on Bering Island. After the bottleneck, the Mednyi population stabilised (Goltsman et al. 2011) at numbers much lower than before the decline, with only about 90 individuals in 2005 (Goltsman et al. 2005), in sharp contrast to up to 1000 individuals before the decline (Geptner and Naumov 1967), rendering this subspecies endangered (Goltsman et al. 1996).

In contrast to the Commander Islands, on Iceland two ecotypes of Arctic fox populations are observed, the inland and coastal populations (Hersteinsson and Macdonald, 1996). Whereas coastal Icelandic foxes mainly feed on sea birds and eggs, invertebrates and sea mammal carcasses, inland foxes usually feed on ptarmigans, migrating waders, geese, eggs and carrion (Hersteinsson and Macdonald, 1996). In Chapter II, we investigate pathogen and Hg related causes of the observed population decline on Mednyi Island in comparison to Arctic foxes on Iceland.

Use of carbon and nitrogen stable isotopes as dietary proxies

Stable isotope analysis is an approach to improve the overall understanding of feeding behaviour and food webs (Kelly, 2011, 2011) and to unravel animal diets, as stable isotopes in animal tissues and excreta reflect diets and yield insight into the environmental conditions experienced by the animal (e.g. West et al., 2006). Primary producers at the base of food webs often incorporate into the biological molecules that they manufacture with distinct carbon, nitrogen, hydrogen, phosphorus and sulphur isotopes (Newsome et al., 2010). Nitrogen isotopes ($^{15}N/^{14}N$) undergo strong fractionation as a consequence of an organism's metabolic processes such as digestion and assimilation of ingested prey (Kelly, 2011; West et al., 2006). This results in a stepwise ^{15}N enrichment along the food web in consumers in relation to ^{14}N by ~2 to 3.4 ‰ above that of their prey (Vanderklift and Ponsard, 2003), so the $^{15}N/^{14}N$ ratio can be used to assess trophic position of a species (Newsome et al., 2010) with the highest ratios found in top predators. The carbon isotopic ratio $^{13}C/^{12}C$ is less modified along the trophic levels. This allows the identification of direct or indirect producers, allowing researchers to distinguish different carbon sources from the terrestrial environment, freshwater systems and oceans (Kelly, 2000) and to study the species' distribution and habitat use (Torniainen et al., 2017). In the current work we use nitrogen and carbon stable isotopes to relate Hg exposure to dietary traces in Arctic wolves (Chapter I). Arctic foxes' dietary sources of Hg intake were evaluated in relation to reported prey items and habitat type (coastal versus inland) of the fox populations (Chapter II).

Fur as a minimally invasive sample matrix

Generally, different tissues provide information at various temporal scales (weeks to years), allowing researchers to study animal ecology and physiology at different temporal resolutions (Ramos and González-Solís, 2012). Since keratin in fur is quite resistant to biochemical and physical degradation (Crewther et al., 1965), both the sequestered Hg burden (Appelquist et al., 1984) and its stable isotope composition (Thompson et al., 1995) remain chemically stable in hair for some time. This allows the use of fur or feathers from archived specimens to retrospectively establish chemical exposure and dietary habits of an individual, and to investigate the relationship between them. Fur is recognised as the least invasive sample type for Hg exposure analysis in polar bears (Bechshoft et al., 2019; Cardona-Marek et al., 2009; Dietz et al., 2006) and some non-Arctic mammals such as red foxes, otters and seals (Dainowski et al., 2015; Eccles et al., 2019; Peterson et al., 2016 a,b). No such data on the feasibility of fur in relation to Hg levels in fur and inner organs are available for Arctic foxes. Such data would be highly valuable for a comparison of studies using different tissue types, particularly since Arctic foxes have been shown to be good model species in ecotoxicological studies and toxicokinetic models (Sonne, 2010; Harley et al., 2016). Thus, Chapter III tests whether fur reliably reflects soft tissue levels of Hg in Arctic foxes and whether the observed relationships are strong enough to reliably standardise Hg levels measured in different tissues by several studies on Arctic canids.

1.5 RISK ASSESSMENT AND BIOLOGICAL EFFECTS OF MERCURY AND OHCS ON KEY ARCTIC WILDLIFE SPECIES

The intention of global and European chemical regulations such as the Stockholm Convention (UNEP, 2004), or the Europe-wide regulation on Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH, EC, 2006) is to protect humans and the environment from hazardous substances. To ensure a high level of protection and safe use of chemicals, producers and importers in are responsible for performing health and environmental risk assessments and compile the information in dossiers based on guidance from different chemicals legislations depending on their intended use and the country of authorization. Environmental hazard and risk assessment are obligatory as part of the registration or approval process of a chemicals worldwide. Usually the intrinsic hazards and environmental fate are evaluated, e.g. acute and chronic toxicity and evaluation of persistence, bioaccumulation, and toxicity (PBT). In a further step a quantitative risk assessment is applied which is a product of the intrinsic hazards of a substance (e.g. acute or chronic toxicity) combined with an estimate of the environmental exposure. If the chemical under scrutiny poses an environmental or human risk, risk management measures need to be initiated. These are for instance restriction or specific provisions on the use of the respective chemical depending on the legal framework under which the chemical is regulated. However, ecotoxicological data and information on exposure essential for a thorough risk assessment are often of poor data quality as shown for the majority of REACH registration dossiers (Fantke et al., 2020; Springer, 2015) or are even lacking (EEA, 2019). Reasons are inter alia that environmental risk assessment is costly, time-consuming and cannot keep pace with the quantity of produced and imported chemicals (Treu et al., 2022b). Furthermore, current environmental risk assessment concepts are usually based on data generated under laboratory conditions, typically lacking information on the ecological and landscape like food web magnification (Brühl and Zaller, 2019; Schäfer et al., 2019; Topping et al., 2020; Weisbrod et al., 2009). This inevitably provides an incomplete picture and leads to the situation that true hazards and risks of chemicals under regulatory scrutiny are overlooked leaving humans and wildlife insufficiently protected against the exposure to hazardous chemicals.

In general, pollutants may have direct or indirect effects on wildlife at both the individual and population level. Direct effects on wildlife populations include direct mortality or reduced individual fitness, especially by lowering reproductive output, as shown for metals and many OHCs (Mason and Wren, 2001). For instance, Hoondert et al. (2021) estimated the effect of POPs and mercury on the population growth rate of nineteen polar bear subpopulations from Arctic regions. The assessment was based on a modelling approach as developed by Hendriks and Enserink (1996) using laboratory ecotoxicity data from mammals. This revealed that, on average, PCB concentrations in prey pose a large threat to polar bear subpopulations, with estimated population growth rates to become negative for most subpopulations.

Whereas less information is available on the effects of CECs (Sonne et al., 2021), the harmful effects of Hg and OHCs on Arctic wildlife health have recently become better recognised, as discussed in Chapter IV (Dietz et al., 2019). Harmful effects of mercury and OHCs comprise changes in immune functioning, hormone imbalances, oxidative stress, tissue pathology and behavioural, neurochemical and reproductive impairments (see reviews by Chételat et al., 2020; Desforges et al., 2016; Evers, 2017; Scheuhammer et al., 2015; Sonne, 2010; Dietz et al., 2019, 2013b; Dietz, 2022). For instance, several studies found significant effects of POPs and Hg on humoral and cellular immunity in Arctic species including canids, suggesting that pollutants may be impairing the ability of animals to competently respond to infectious pathogens (reviewed by Dietz et al., 2019; Letcher et al., 2010; Sonne, 2010). For instance, investigating the immunotoxicity in marine mammals using blubber-derived complex chemical ‘cocktails’ from polar bears and killer whales, Desforges et al. (2017) reported significant *in vitro* effects on lymphocyte proliferation, natural killer cell activity and phagocytosis in lymphocytes from cetaceans, seals and polar bears.

Substantial knowledge gaps remain, with regard to population level effects of pollutants on Arctic wildlife species, in particular with respect to the health impairment of top predators. For instance, climate change and other aspects of global change are likely to modify the bio-availability of chemicals in the environment (Tartu et al., 2022). However, it is unclear how top predators will be affected. To this end, field studies on single species often have limited explanatory power in terms of the direct interpretation of adverse cause-and-effect mechanisms. Examining correlations between biological endpoint measurements and contaminant levels using a large cross-species data set can offer a more comprehensive and powerful way to identify and assess pollutant-related biological effects in wildlife, but such a review on the Arctic has not yet been done. Thus, Chapter IV pulls together the knowledge on exposure status and related health effects of OHCs and mercury in a broad range of key Arctic marine and terrestrial mammal, bird and fish species by reviewing the literature published since 2010. This strives to substantiate the lines of evidence regarding pollutant related effects which will support future chemical risk assessments.

2. OBJECTIVES OF THE THESIS

2.1 OVERALL OBJECTIVE

The overall objectives of this thesis are (i) to investigate dietary drivers of mercury exposure in two sentinel predator species, the Arctic wolf and the Arctic fox, and (ii) to evaluate the exposure status of organo-halogenated chemicals and Hg as well as their biological effects in key Arctic marine and terrestrial mammal, bird and fish species at different trophic levels. The results have contributed to the knowledge basis for science-based recommendations regarding conservation managements and policy measures as part of the *Arctic Monitoring and Assessment Program*, with the ultimate goal of protecting Arctic wildlife and ecosystems in the future.

2.2 SPECIFIC OBJECTIVES AND HYPOTHESES

Chapter I: There is a general paucity of information on how dietary and other drivers relate to Hg exposure in terrestrial Arctic top predators, including the Arctic wolf. The aim of the first chapter was to investigate the Hg exposure and its drivers in this understudied sentinel species. Based on previous studies on Hg contamination in marine and terrestrial Arctic canids, we hypothesised that Arctic wolves are not at risk of Hg-associated compromised health and that individuals feeding on marine prey fed on prey of a higher trophic level and therefore exhibit higher Hg fur levels. To test this, the link between dietary sources, trophic magnification effects and Hg exposure was examined by means of stable isotope analysis and total Hg measurements in fur samples from 30 Arctic wolves sampled between 1869 and 1998 in the High Arctic areas of Canada and Greenland.

Chapter II: As outlined above, the Arctic fox population on Mednyi Island declined dramatically between 1970 and 1980 for unknown reasons; this island had not been inhabited by people since the 1960s. The objective of the second chapter was to investigate possible pathogen and Hg related factors of this unexpected decline by dermining Hg levels in the foxes and screening for several pathogens that plausibly could cause mass mortality in canids. We hypothesised that Hg levels depend on the prey living in different fox habitats, specifically coastal habitats compared with inland territories, because of biomagnification of Hg when feeding on marine food items, overriding any possible effects of geographic origin (Commander Islands versus Iceland). We further assumed that as a consequence coastal foxes exhibit high mercury levels which partly contributed to the decline of the Mednyi fox population. Therefore, we chose a study design that allowed the comparison of Hg fur levels from historical specimens from the Commander Islands from the population pre-crash period on Mednyi Island, modern Mednyi foxes, and two unrelated and geographically remote ecotypes represented by inland and coastal Icelandic Arctic fox populations.

Chapter III: Although fur is recognised as the least invasive sample type for Hg exposure analysis in many wildlife species, measurements in previous ecotoxicological studies used different tissues (usually liver), which hinders cross-study comparison. Therefore, the third chapter aimed to determine whether fur can be used as a minimally-invasive sampling matrix to reliably determine Hg levels in soft tissues of Arctic foxes. We hypothesised that Hg levels in fur are related to those of other tissues and that these correlations are strong enough to reliably predict Hg concentrations in soft tissues, specifically liver and kidney. To test this, the links between Hg levels in the fur, liver and kidney of 35 Arctic foxes sampled in 2011-2012 on Iceland were investigated. Regression model equations were derived to extrapolate Hg soft tissue levels from Hg fur concentrations, which enabled a better cross-study comparison and improved risk assessment for Arctic canids. With these equations, we could compare the results of previous studies on Hg levels in different tissues of Arctic foxes from Iceland, Canada, Norway, Alaska and the Russian Arctic.

Chapter IV: Single field studies reporting contaminant concentrations of single substance, single wildlife species and one toxicological endpoint (effect) often have limited explanatory power for the direct interpretation of adverse cause-and-effect mechanisms. Examining correlative relationships between biological endpoint measurements and contaminant levels using a large set of studies across different species is a powerful approach to unravel chemical-related biological effects in wildlife. Thus, the fourth chapter took the analysis a step further and gathered the exposure status of various organo-halogenated chemicals and Hg as well as reported cumulative health effects in key Arctic marine and terrestrial mammal, bird and fish species reviewing literature published between 2010 and 2019.

3. CHAPTERS AND LIST OF PUBLICATIONS

This doctoral thesis is divided into four chapters, each of which is a research article published in the journal of 'Science of the Total Environment' or 'PLoS ONE'. The articles are provided in the journal format.

1. **Treu, G.,** Sinding, M.-H.S., Czirják, G.Á., Dietz, R., Gräff, T., Krone, O., Marquard-Petersen, U., Mikkelsen, J.B., Schulz, R., Sonne, C., Søndergaard, J., Sun, J., Zubrod, J., Eulaers, I., 2022. An assessment of mercury and its dietary drivers in fur of Arctic wolves from Greenland and High Arctic Canada. Science of The Total Environment. 838, 156171. https://doi.org/10.1016/j.scitotenv.2022.156171

   GT: conceptualisation, writing and editing of the original and final draft of manuscript, formal analysis, statistical analysis, visualisation.

Chapter I

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2. Bocharova*, N., **Treu**,* G., Czirják*, G.Á., Krone, O., Stefanski, V., Wibbelt, G., Unnsteinsdóttir, E.R., Hersteinsson, P., Schares, G., Doronina, L., Goltsman, M., Greenwood, A.D., 2013. Correlates between feeding ecology and mercury levels in historical and modern Arctic foxes (*Vulpes lagopus*). PLoS ONE 8, e60879. https://doi.org/10.1371/journal.pone.0060879.

   *shared first authorship, GT: conceptualisation, writing and editing of the original and final draft of the manuscript, mercury analysis, parts of the statistical analysis, visualisation.

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*GT: mercury analysis of samples from of Arctic foxes from Iceland and Norway, commenting and editing on the original and final draft of the manuscript.

Chapter IV

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4. CHAPTER I: AN ASSESSMENT OF MERCURY AND ITS DIETARY DRIVERS IN ARCTIC WOLVES FROM GREENLAND AND HIGH ARCTIC CANADA

Treu, G., Sinding, M.-H.S., Czirják, G.Á., Dietz, R., Gräff, T., Krone, O., Marquard-Petersen, U., Mikkelsen, J.B., Schulz, R., Sonne, C., Søndergaard, J., Sun, J., Zubrod, J., Eulaers, I., 2022. Science of The Total Environment 838, 156171.

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5. CHAPTER II: CORRELATES BETWEEN FEEDING ECOLOGY AND MERCURY LEVELS IN HISTORICAL AND MODERN ARCTIC FOXES (*VULPES LAGOPUS*)

Bocharova, N., Treu, G., Czirják, G.A., Krone, O., Stefanski, V., Wibbelt, G., Unnsteinsdóttir, E.R., Hersteinsson, P., Schares, G., Doronina, L., Goltsman, M., Greenwood, A.D., 2013. PLoS ONE 8.

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6. CHAPTER III: CORRELATIONS BETWEEN HAIR AND TISSUE MERCURY CONCENTRATIONS IN ICELANDIC ARCTIC FOXES (*VULPES LAGOPUS*)

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7. CHAPTER IV: CURRENT STATE OF KNOWLEDGE ON BIOLOGICAL EFFECTS FROM CONTAMINANTS ON ARCTIC WILDLIFE AND FISH

Dietz, R., Letcher, R.J., Aars, J., Andersen, M., Boltunov, A., Born, E.W., Ciesielski, T.M., Das, K., Dastnai, S., Derocher, A.E., Desforges, J.-P., Eulaers, I., Ferguson, S., Hallanger, I.G., Heide-Jørgensen, M.P., Heimbürger-Boavida, L.-E., Hoekstra, P.F., Jenssen, B.M., Kohler, S.G., Larsen, M.M., Lindstrøm, U., Lippold, A., Morris, A., Nabe-Nielsen, J., Nielsen, N.H., Peacock, E., Pinzone, M., Rigét, F.F., Rosing-Asvid, A., Routti, H., Siebert, U., Stenson, G., Stern, G., Strand, J., Søndergaard, J., Treu, G., Víkingsson, G.A., Wang, F., Welker, J.M., Wiig, Ø., Wilson, S.J., Sonne, C., 2022. Science of The Total Environment 829, 154445.

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8. MAIN FINDINGS

- Nitrogen stable isotope, not carbon source or region, explains variation in Hg exposure in Arctic wolves, suggesting that biomagnification is the major driver of observed Hg levels. Large ranges of nitrogen and carbon stable isotopic profiles reflect high dietary plasticity of terrestrial and marine food sources in Arctic wolves. These are likely related to fluctuations in food availability and dietary preference, but may be influenced by other factors such as shifts at the lowest trophic levels of food webs.
- Historical and modern fur samples confirm the usefulness of fur for monitoring Hg and carbon and nitrogen stable isotopes in Arctic canids and might be applied in both wildlife welfare and conservation contexts.
- In Arctic foxes, Hg levels increase not only with trophic position of prey but also with the proportion of marine prey in coastal ecotypes, as compared to inland foxes feeding mainly on terrestrial resources. Highest Hg concentration were found in the coastal ecotype as shown for the Mednyi Island, where foxes depend solely on marine vertebrates. Hg exposure may thus in part explain the decline of the Mednyi Island foxes in the 1970s which renders particularly young foxes vulnerable to infectious diseases. Our large-scale health assessment using serological and DNA based pathogen screening techniques suggested a low prevalence of pathogens in contemporary foxes, although it does not allow a proper assessment of the health status of the Mednyi Arctic foxes during or before the crash period. Most likely a complex interplay of stressors explains the high cub mortality observed on Mednyi Island.
- Mean Hg levels ± SD (µg g^{-1} dry weight) in fur of Arctic wolves from Greenland and High Arctic Canada were 1.41 ± 1.39 (n = 30). Coastal Arctic foxes from Iceland (10.58 ± 2.12, n = 16) and Commander Islands (contemporary foxes: 10.42 ± 2.45, n=12; historic foxes: 10.42 ± 1.31, n = 11) had nearly 3 times higher Hg fur levels than those from inland habitats (3.55 ± 1.00, n = 12). Whereas the Arctic wolves most likely did not have a compromised health, Hg concentrations in some of the Arctic foxes exceeded the putative thresholds for Hg-mediated toxic health effects previously estimated for non-target species or populations outside the Arctic (Dietz et al., 2022, 2019).
- Hg levels in fur reliably reflected concentrations in internal (liver and kidney) tissue in Arctic foxes. Significant relationships of total Hg levels among tissue types were observed between fur and kidney ($R^2 = 0.51$), fur and liver ($R^2 = 0.61$), as well as between liver and kidney ($R^2 =$

0.77). As demonstrated for Arctic foxes, regression model equations will be useful for future comparisons of Hg levels in tissues of Arctic canids and will support interpretation of exposure risks of species at individual and population levels.

- The literature review on exposure levels and effects of Hg and organo-halogenated chemicals on Arctic wildlife post 2000 revealed:
    - a wealth of information on biological effects in marine mammals and seabirds, and sentinel species such as the domestic dog races used to run sledges and the Arctic fox. However, information on other terrestrial vertebrates and fish remains scarce. Reported effects concerned vitamin metabolism, immunocompetence, hormones, oxidative stress, tissue pathology and reproduction.
    - depending on the species and population, some of the tissue burdens of PCBs and Hg post 2000 are high enough to exceed putative risk threshold levels that have been previously estimated for non-target species or populations outside the Arctic. For the polar bear, pilot whale, narwhal, beluga as well as the hooded seal, a proportion of the population is at high or severe risk for health effects mediated by Hg exposure, and bird Hg concentrations are above toxicity thresholds in many areas of the marine environment.
    - knowledge gaps remain with regard to the biological and toxicological effects of organo-halogenated chemicals and Hg in terrestrial predators in the Arctic, particularly with a view to chemicals of emerging concern, as well as on endpoints addressing population health.

9. GENERAL DISCUSSION

9.1 LEVELS AND DRIVERS OF MERCURY EXPOSURE IN ARCTIC CANIDS

In this thesis, Arctic wolves and Arctic foxes were used as sentinels to assess Hg exposure patterns in the understudied terrestrial Arctic environment. Chapters I–III allow the comparison of Hg levels (given as mean ± standard deviation in µg g^{-1} dry weight) between Arctic foxes and wolves from the western Aleutian Arc, Greenland, the High Arctic part of Canada and from Iceland from different time periods. Average Hg concentrations in Arctic wolves from Greenland and the Canadian High Arctic (1.41 ± 1.39, n = 30) tended to have low Hg fur concentrations as compared to the values of other terrestrial Arctic wildlife and Alaskan and Canadian wolves (Braune et al., 2015; Gamberg and Braune, 1999; McGrew et al., 2014). The Hg fur level of Arctic foxes were up to 7-fold higher in coastal and twice as high in inland populations as compared to the Arctic wolves. Coastal Arctic fox populations from Iceland (10.58 ± 2.12, n= 16) and the Commander Islands (contemporary foxes: 10.42 ± 2.45, n=12; historic foxes: 10.42 ± 1.31, n = 11) had up to 7-fold higher total Hg fur levels than inland populations (3.55 ± 1.00, n = 12) or those reported for Arctic foxes from Svalbard (Prestrud et al., 1994), the Canadian Arctic (Ballard et al., 2003; Dehn et al., 2006), and Alaska (Ballard et al., 2003; Dehn et al. 2006).

We compared Hg liver levels measured or estimated in Chapters I-III to the putative risk thresholds of Hg-mediated reproductive impairment proposed in Chapter IV (Dietz et al., 2022). These toxicity thresholds were derived by risk quotient calculations based on the ratio of measured Hg liver concentrations and estimated critical body residues. Hepatic total Hg threshold values were calculated from relatively low critical daily doses determined in the laboratory for mink (*Mustela vison*) and by using physiologically-based pharmacokinetic modelling (Nielsen et al., 2006). This yielded five risk categories for reproductive toxicity – no risk, low risk, moderate risk, high risk and severe risk. The comparison revealed that most Arctic foxes on Iceland (8 - 35 %) were at low to moderate risk of health and reproductive impairment, though 9 % of the adults were at severe risk. Among the Arctic foxes from the Commander Islands a large proportion of individuals (73% of the pre-crash period and 42% of contemporary foxes) were at moderate risk of reproductive impairment, whereas the remaining foxes fall into the low or no risk category. Unlike Arctic foxes and according to our hypothesis, observed Hg levels in Arctic wolves indicate they are at no risk (93 %) or at low risk (7 %).

When interpreting results of Hg analysis of historical material it is important to remember that mercuric chloride ($HgCl_2$ or sublimate) was sometimes used in the late 1800s and early 1900s during sampling, museum cataloguing or subsequent storage (e.g. Horton et al., 2009; Dietz et al., 2011). In this thesis, the treatment and storage processes of the fur samples of Arctic wolves and foxes at the different museums had not been reported to us. Since we followed recommendations for sample preparation to reduce external Hg contamination (Morton et al., 2002) and the observed Hg levels lied within or below the biologically plausible range of those reported in Arctic canids such as grey and Arctic wolves (Gamberg and Braune, 1999; McGrew et al., 2015) and Arctic foxes (reviewed in Treu et al., 2018; Hallanger et al., 2019), it is unlikely that external mercury contamination was relevant.

Dietary drivers of mercury exposure

The results of Chapters I are consistent with the predictions from our hypothesis and provide evidence that biomagnification (an increase in δ^{15}N) of Hg substantially and significantly explained the variation in Hg levels in Arctic wolves, whereas region and carbon source (δ^{13}C) were not relevant. Similarly, and as expected, Chapter II demonstrated that in Arctic foxes the strongest driver of total Hg concentration was coastal ecotype, with foxes that relied solely on marine vertebrates, as on Mednyi Island, reaching the highest Hg body burden. The observed large variability in carbon stable isotopes indicated that although some Arctic wolves fed on both marine and terrestrial prey, most individuals foraged mostly on terrestrial food items, which in turn may partly explain the observed low Hg fur levels.

The analysis of Chapter I and II partly relies on unique specimens opportunistically available at different museums and scientific collections, contributing to the spatial and temporal heterogeneity of the data. Furthermore, due to current lack of systematic sampling of Arctic wolves and strict conservation and hunting restrictions, only three recent samples (post 1992) could be obtained while the remaining wolves (*n*= 27) were collected before 1941. Due to the temporal and spatial heterogeneity of the Arctic wolf data temporal trend analysis of Hg was not reasonable from a statistic point of view. Furthermore, interpretation of the historical fur stable isotope data is a challenge, since similar information for other Arctic organisms lower in the food chain is usually unavailable but would be required, as the isotope composition of biological materials depend upon trophic position, food web structure and feeding behaviour (Horton et al., 2009). Such historical data or suitable specimens were not available for most Arctic organisms, including Arctic wolves.

The observed depleted δ^{13}C values found in some of the Arctic wolves might also stem from depletion of baseline δ^{13}C values in Arctic oceans related to the decline in sea ice over time and subsequent changes in isotopic fractioning in phytoplankton (de la Vega et al., 2019) rather than to terrestrial food sources. In order to provide a more refined estimate of Arctic predator food web patterns and to link them to pollutant levels, future studies would be needed that examine multiple isotopes, including sulphur and amino acid-specific stable isotopes of δ^{15}N and δ^{13}C (Elliott et al., 2021; Góngora et al., 2018; O'Donovan et al., 2018). The development of such new techniques and multidisciplinary approaches in this research opens a promising new spectrum of possibilities for effective conservation measures through the quantitative tracking of chemicals in food webs.

Still, our data allow some interpretation. The data are consistent with the predictions from the hypothesis that Hg exposure is determined by biomagnification within food chains across Arctic ecosystems and by the foraging strategies of Arctic canids, rather than by geographic region. Previous studies demonstrated that Arctic and non-Arctic mammals feeding at higher trophic levels are more polluted (e.g., review by Lavoie et al., 2013) as Hg biomagnification occurs in Arctic food webs via uptake by primary producers and subsequent consumption by invertebrates, fish, birds and marine and terrestrial carnivores (Atwell et al., 1998; Jæger et al., 2009; Ruus et al., 2015). Consistent with these studies, Chapter II show that Mednyi and coastal Iceland Arctic fox populations had higher mercury levels than those feeding on prey at a lower trophic level in inland habitats. Similarly strong links between mercury levels and feeding habits, as inferred by different stable isotopes or dietary analysis, have also recently been recognised in Arctic foxes from Svalbard (Hallanger et al., 2019), wolves from Alaska (McGrew et al., 2014) and other Arctic mammals, birds and fish (Atwell et al., 1998; Aubail et al., 2011; Burnham et al., 2018; Dietz et al., 2021; Hobson et al., 2002; Lehnherr, 2014).

Considering Hg loads in potential prey species of Arctic foxes and wolves, generally higher Hg levels are found in marine species such as seals (Fant et al., 2001; Riget et al., 2005) and seabirds (Braune et al., 2002; Jæger et al., 2009) than in terrestrial species such as the herbivorous reindeer (Riget et al., 2004) and geese (Braune and Malone, 2006). We found similar relations when measuring a small subset of Arctic fox prey species, i.e. bird and seal samples collected from carcasses in seal rookeries or food remains on Arctic fox den sites on Mednyi Island during 2010 and 2011. In northern fur seals (*Callorhinus ursinus*), Hg levels were variable and on average high. In the case of marine birds, the average Hg concentrations were lower than in seals. However, the available samples did not permit the statistical comparison of museum and recently-collected prey species.

As with Hg, similar relationships were found for POP levels, with increasing concentration in Arctic foxes from Svalbard when foraging at higher trophic levels and/or on marine prey (Fuglei et al., 2007; Hallanger et al., 2019). Dietary preferences also affect pollutant exposure levels to humans, as shown by the fact that people regularly consuming fish or the meat of whales and seals have higher blood and hair concentrations of POPs and metals (including Hg) than those eating other food (Dallaire et al., 2013; Kinghorn et al., 2006; Pontual et al., 2021; Tian et al., 2011). The results confirm the usefulness of fur samples for monitoring Hg and stable isotopes in Arctic canids, which might be applied in both wildlife welfare and conservation contexts. It further suggests that the absolute exposure to Hg and other pollutants may be less important than the feeding ecology and feeding opportunities of canid top predators, which may in turn influence population health and stability, as already demonstrated for POPs in Arctic foxes (Fuglei et al., 2007), and which may trigger population declines in predators such as polar bears (Hoondert et al., 2021) and killer whales (Desforges et al., 2018). In particular, PCB-mediated effects such as impaired fertility are predicted to affect whale populations near industrialized regions over the next 100 years, and those feeding at high trophic levels regardless of location, increasing the risk of population collapse (Desforges et al., 2018).

Other biotic and abiotic drivers

In general, concentrations of chemicals in biota depend on factors such as age, sex, season, nutritional condition and feeding ecology, climate and weather (Brown et al., 2018; Chételat et al., 2022; Hung et al., 2022; Ma et al., 2016; McKinney et al., 2022; Vorkamp et al., 2019). In this thesis, season, geographic origin and sex had no significant influence on total Hg levels in Arctic foxes and wolves. Usually, mercury burdens increase with the age of individuals, as shown in the Arctic foxes sampled in this work, in part due to longer exposure in older individuals (AMAP, 2011; Braune et al., 2015). This in turn can result in the accumulation of high mercury levels in hair and inner organs, as shown for the Arctic foxes in Chapter II. Since the Arctic wolf samples were collected opportunistically from different museums and information on age and other biological traits was not available, age related effects on Hg levels could not be analysed.

The large variance in the Hg exposure levels of Arctic foxes and wolves is assumed to relate to the interaction of several biotic and abiotic factors, since concentrations of total Hg and MeHg in Arctic biota are considered to be determined by complex interacting processes which act on one or both of the primary regulating factors, the size of the abiotic pool of Hg available for bioaccumulation and the transfer of MeHg through aquatic food webs (St. Louis et al., 2011; Kirk et al., 2012; Lehnherr, 2014; Loseto et al., 2008). Relationships between concentrations of Hg in high trophic level Arctic wildlife and atmospheric oscillation indices, sea ice conditions, temperatures and precipitation have been described for several Arctic predators in Canada and Greenland (Houde et al., 2020; Loseto et al., 2015; McKinney et al., 2017; Adam D. Morris et al., 2022; Rigét et al., 2012). For instance, increased warming and decreased sea ice cover are changing the fate and cycling of mercury, and instigate greater oceanic production of MeHg (AMAP, 2011), which could increase bioavailability and accumulation, particularly in high trophic level organisms. Sea ice coverage, precipitation and air temperature have all been related to Hg levels in marine mammals or seabirds, although the strength and direction of these relationships varies substantially (Foster et al., 2019; Houde et al., 2020; Rigét et al., 2012). Thus, in addition to the observed biomagnification and ecotype-dependent feeding habits as major drivers, other determinants of Hg exposure in Arctic wolves and foxes are most likely associated with differences in (local) geographical, ecological, biological and climatic conditions, global emission patterns and dietary differences of Arctic foxes and wolves in the study areas.

As also discussed in Chapter III, the high Hg levels detected in the Arctic foxes from Iceland and the Commander Islands may be partly related to local mercury releases from the numerous volcanoes and hot springs across Iceland. Active volcanoes and quiescent degassing are known to eject large quantities of volatile mercury into the stratosphere in sufficient quantities to influence the global and regional cycles of mercury for several years (Edwards et al., 2021a, Bagnato et al., 2015). One year before the sampling period of Arctic foxes on Iceland (2010), the volcano *Eyjafjallajökull* erupted, throwing volcanic ash several kilometres into the atmosphere, which likely affected the Hg loads of foxes. A recent study measuring Hg concentrations and fluxes from the *Fagradalsfjall* fissure eruption on Iceland in 2021 concluded that volcanoes were the dominant global Hg source to the atmosphere(Edwards et al., 2021b). Astonishingly, the eruption correlated with increased mercury concentrations detected in far-off regions of the northern hemisphere (Pankratov et al., 2018) and might thus have affected the Hg exposure of foxes on Commander Islands as well. All Aleutian Islands are volcanic in origin, with several active or potentially active volcanos (e.g. Kenney et al., 2012; Melekestsev, 2009). Accordingly, the Aleutian Islands experience an additional local source of Hg from regular volcanic activities (Kenney et al., 2012), which in turn likely affected the observed Hg levels in the Arctic foxes on Commander Island.

The results of this thesis may serve as a reference point for assessing future temporal trends in global Hg pollution in Arctic regions and their effects on terrestrial predator populations. To overcome knowledge gaps and to unravel the complex biogeochemical pathways of different Hg forms in Arctic predators and the relative importance of local sources such as contaminated sites or volcanic emissions, novel approaches for measuring Hg stable isotopic fractioning (Yin et al., 2014; Tsui et al., 2020) might become important.

9.2 FACTORS ASSOCIATED WITH THE DECLINE OF THE MEDNYI FOX POPULATION

What makes Mednyi Island unique is that it represents the only Arctic fox ecotype to have suffered a substantial decline, almost an extinction, in recent history. The reported clinical signs in the Mednyi Arctic foxes during the sudden population crash in 1970-1980 were unspecific, including low body weight, loss of fur, skin abnormalities and poor condition, but do resemble symptoms of chronic Hg poisoning in mammals (e.g. reviewed by Evers, 2017; Scheuhammer et al., 2015; Wren, 1986). Thus, Chapter II examines Hg as a possible cause of the population decline. It uses Hg levels from historical specimens from the Commander Islands from the population pre-crash period on Mednyi Island, as well as samples from contemporary Mednyi foxes and two unrelated and geographically remote ecotypes represented by inland and coastal Icelandic Arctic fox populations as control groups. Among all studied populations, Hg concentrations were indeed significantly highest, with a large proportion of individuals being at moderate risk, in the Mednyi population from the pre-crash period. This makes plausible the conclusion that chronic Hg exposure may have compromised overall health, e.g. through immune suppression (Dietz et al., 2019, 2013b; Sonne, 2010), making foxes more vulnerable to diseases or starvation. This is a tentative conclusion since the sample size of foxes was small (historical: n= 11; contemporary: n= 12), a fact that generally reduces the likelihood that a statistically significant result reflects a true effect (Button et al., 2013; Krzywinski and Altman, 2013). In any case, it is generally challenging to establish links between chemical exposure and health outcome in wildlife populations (Rodríguez-Estival and Mateo, 2019) given the numerous other natural and anthropogenic stressors that can also impact health endpoints.

We did not, for example, analyse the co-occurrence of other pollutants in the Mednyi population. Several studies documented that marine mammals and birds collected more recently from the Aleutian Archipelago were indeed exposed to high concentrations of Hg and OHCs and found that pollutant burden in biota increased along the island chain in a westward direction towards the Commander Islands (Anthony et al., 1999; Burger and Gochfeld, 2007; Correa et al., 2014; Kaler et al., 2014; Ricca et al., 2008; Rocque and Winker, 2004). This gradient increasing towards the western end of the archipelago has been attributed to storms that pull pollutants from Asia eastward along the archipelago (AMAP, 2002) and might partly explain why such high Hg levels were observed in the foxes on the Commander Islands. The Aleutians were also affected by military activities during World War II and continued until the end of the 1990s, which led to releases of a variety of anthropogenic pollutants including Hg (Kenney et al., 2012; US Navy, 1999).

Naturally occurring toxins might also be of relevance with a view to the health of the Mednyi foxes and other Arctic predators, and further research would be necessary to test this suggestion. Two of the most common harmful algal bloom toxins are domoic acid and saxitoxin which can be transferred to foetuses via the placenta, leading to reproductive failure or affecting the survival of successfully delivered offspring, as it is delivered to pups via lactation (Brodie et al., 2006; Lefebvre et al., 2016; Rust et al., 2014). These two compounds were detected in levels above toxicity thresholds in different whales, the walrus (*Odobenus rosmarus divergen*) and four seal species from the Bering Sea and the Aleutians for decades.

One of the strongest natural selection pressures on wildlife populations are pathogens, which challenge the immune system of individuals and can lead to diseases and death (Lehmann, 1993). Diseases caused by bacteria, parasites or viruses are a natural and widespread factor in population dynamics. In a small, endemic population such as the one on Mednyi Island, they may have dramatic consequences. It is possible that the sudden decline in Mednyi foxes was related to a lethal, infectious disease in the foxes. Therefore, we also screened for multiple plausible pathogens that could cause mass mortality in canids, such as canine parvovirus and canine distemper virus. However, retrospective examination is feasible only if sufficient and adequately treated and stored historical material is available. For most pathogens, acquiring historical DNA requires soft-tissue samples, faecal remains or samples preserved to medical archive standards (Devault et al., 2014), which were not available from Mednyi foxes from during or before the crash-period. Pathogen detection in ancient material remains a challenge because the presence, preservation and ultimate detection of ancient pathogen DNA or RNA depends on many complicating factors. These include the epidemiology of the disease, preservation techniques used, the biology of the pathogen and its load at time of death (Duchêne et al., 2020; Raxworthy and Smith, 2021; Tsangaras and Greenwood, 2012). The pathogen screening could therefore only be conducted on blood samples of contemporary Arctic foxes and revealed seronegative results for canine parvovirus, canine distemper virus and *Neospora caninum* and a prevalence of *Toxoplasma gondii* low compared to Icelandic foxes (personal communication by Gábor Á. Czirják). Even though these findings give certain insights into the health condition of the Mednyi foxes today, they allow no conclusion as to whether an infection of Arctic foxes is a possible explanation of the population decline.

Interestingly, on Bering Island and the Pribilof Islands (northeast of the Commander Islands), a northern fur seal population experienced decline owing to low pup production in the same period as did the Arctic fox population on Mednyi (1970 to 1980s, Fomin et al., 2019; Towell et al., 2006). While the causes remained unknown on the Pribilof Islands (Towell et al., 2006), a parasitic disease caused by hookworms of the genus *Uncinaria ssp.* was suggested as the reason on Bering Island (Starostin, 1973), with 95% of adult seals and 45% of cubs still infected today (Fomin et., 2019). No such data are available for Artic foxes from the Commander Islands.

Significant effects of POPs and Hg on humoral and cellular immunity for Arctic mammal species are well described (review by Desforges et al., 2016; Dietz et al., 2019), suggesting that pollutants may impair the ability of animals to respond to infectious pathogens as might have been the case in the Mednyi foxes. Parasites in turn can destabilise host populations (Anderson and May, 1978), decrease vital demographic parameters such as survival or fecundity (Anderson and May, 1978; Andreassen et al., 2017) and, when combined with environmental stressors, could have a fatal effect on populations as exemplified by the northern fur seals (Fomin et al., 2019). The relatively unspecific clinical signs and high cub fatalities observed in the Mednyi foxes during the period of population decline could also reflect symptoms described for other infectious diseases and zoonoses in Arctic foxes and other canids, such as sarcoptic mange (Pence and Ueckermann, 2002). With sarcoptic mange, mortality generally occurs within 3–4 months after infection in canid populations not previously exposed (Bornstein et al., 2001; Niedringhaus et al., 2019). Accordingly, rapid declines in both Arctic foxes (Mörner, 1992) and red foxes (Bornstein et al., 2001; Fuchs et al., 2016; Niedringhaus et al., 2019; Pence and Ueckermann, 2002; Pisano et al., 2019) following outbreaks of sarcoptic mange have been reported from several parts of the world. In the context of parasitosis, it has been suggested earlier that ear mites (*Otodectes cynotis*) were brought to Mednyi Island on the domestic dogs that accompany seafaring trappers, leading to otodectic mange and the decline of the fox population (Goltsman et al., 1996). However, unlike for *Sarcoptes scabiei*, infestation with *Otodectes cynotis* causes neither mortality in canids nor skin abnormalities of different parts of the body (Briceño et al., 2020; Saari et al., 2018) as observed in Mednyi foxes, which is why the proposed explanation seems unlikely. Furthermore, there was no systematic parasitological investigation on the parasite burden and clinical signs in Mednyi foxes, only oral reports from trappers, and thus no causal link with the fox decline can be conclusively drawn.

Another relevant aspect is the potential loss of diversity in the gene pool of the Mednyi fox population. It is well documented that isolation and/or large demographic declines affect the ability of a population to maintain genetic diversity over time due to genetic drift (Frankham, 1997; Keller and Waller, 2002). Genetic studies have shown that the current Mednyi population indeed displays low variability (Ploshnitsa et al., 2012; Prôa and Nanova, 2020), a probable consequence of the bottleneck in the 1970s (Ploshnitsa et al., 2012) or of being isolated for such a long period. In this context, a reduction in the diversity of the major histocompatibility complex (MHC), an important genetic system for combatting infections in vertebrates (e.g. Klein, 2013), may have led to reduced fitness and an increased susceptibility to infection (e.g. O'Brien and Evermann, 1988). Accordingly, Ploshnitsa et al. (2012) investigated the functionality of the MHC polymorphism in Mednyi Arctic foxes by comparing museum samples collected before the population crash with contemporary Arctic foxes, and suggested that the observed reduction in MHC variation might explain pathogen susceptibility and the small population size after the decline. However, several studies on other species also report limited MHC variation in species that (i) are supposed to have undergone population bottlenecks but demonstrate compensatory mechanisms in terms of a strong innate immunity (Heinrich et al., 2017), or (ii) have undergone population bottlenecks or small had small founder populations and expanded in number (beaver [*Castor* ssp.] e.g. Babik et al., 2005; Ellegren et al., 1993). This implies that low variation in MHC loci cannot be assumed to be responsible for serious susceptibility to pathogens or population declines but requires the demonstration of a specific mechanism by which low MHC variation is linked to the operation of a specific pathogen or a group of pathogens.

In conclusion, there is a paucity of data due to both challenges in pathogen screening methodologies and a lack of suitable historical material from before the 1990s, which currently prevents any firm conclusions on the true causes of the Mednyi fox decline. Since the Mednyi foxes solely depended on marine food sources, as the pollutant gradient increased towards the western Aleutians islands and POPs peaked in Arctic marine food webs in the 1970s to 1980s (Brown et al., 2018; Muir et al., 1999; Rigét et al., 2019), it is plausible that Mednyi foxes were exposed to several hazardous chemicals including Hg, which in combination contributed to health and/or reproductive impairment, and possibly made them more vulnerable to canine pathogens. Most likely the complex interplay of pollutant exposure with other factors, such as possibly pathogen-born infections, was responsible. To better understand the exact causes, further analyses with larger sample sizes for ancient material would be needed, ideally including both a wide-scope target and non-target contaminant screening, and novel techniques such as next-generation sequencing, to allow detection of a broader spectrum of pollutants and pathogens.

9.3 UTILITY OF FUR FOR MONITORING SOFT TISSUE MERCURY LEVELS IN ARCTIC CANIDS

Fur samples have been repeatedly demonstrated to be promising for minimally-invasive biomonitoring of Arctic pollution using polar bears as a sentinel species (Bechshoft et al., 2019, 2016; Dietz et al., 2006a, 2009). The physicochemical and biochemical stability of hair (Crewther et al., 1965) allow both sequestered chemicals such as Hg (Appelquist et al., 1984) as well as biomolecules to remain stable over time. Hair has been widely studied as an indicator of blood Hg levels and dietary exposure in humans (FAO/WHO, 2003). Similar relationships were studied between soft tissues such as muscles, kidney, liver and brain, and non-lethally, minimally-invasively collected samples such as blood and fur in both captive and free-living apex species like red foxes (*Vulpes vulpes,* Binkowski et al., 2016; Dainowski et al., 2015), sledge dogs (*Canis lupus familiaris*, Lieske et al., 2011), polar bears (Bechshoft et al., 2019; Dietz et al., 2022), and various seal species (Peterson et al., 2016 a,b). Though the Arctic fox is recognized as a good model for understanding the toxicokinetic dynamics of pollutants in the Arctic, because of its circumpolar distribution (Harley et al., 2016; Sonne, 2010), no such data were available for this species. Hence, Chapter III used fur from Arctic foxes to extrapolate Hg concentrations in soft tissues and provide predictive regression models applicable for cross-tissue comparison. The results are consistent with the predictions from our hypothesis that the strong and significant correlations between fur and organ concentrations can be used as a minimally-invasive matrix to determine Hg liver and kidney levels, which will be useful for future biomonitoring studies of Arctic canids. Since liver is usually the target organ in ecotoxicological studies, the transformation equations will ultimately help with interpreting the exposure risks of canine species at the individual and the population level.

Our findings are also consistent with results from Alaskan red foxes (Dainowski et al., 2015) which demonstrated that total Hg (THg) concentration in hair was correlated with the concentration in organ tissues and these correlations were strong enough to predict THg concentrations in soft tissues, specifically liver and kidney. Similar strong correlations were also found between blood, feathers and internal organs in birds (Bianchi et al., 2008; Eagles-Smith et al., 2008). There is a slight caveat in that the study described in Chapter III was conducted on free-living arctic foxes and not under controlled conditions. Hence there are additional factors which may have contributed to the variation in tissue concentrations, as no information was available on previous exposure levels to Hg, feeding behaviour, home range size or periods of food restriction which might influence Hg levels and the links between hair and soft tissue concentrations.

Furthermore, the sample size of 35 Arctic foxes was relatively small and it could be argued that this limits the predictive power of the study. Is this sample size sufficient to provide a predictive regression equation with a good confidence interval? In previous publications, statements varied as to what should be the minimum sample size, or better the number of subjects per variable (SPV), which are required for an accurate estimation of a linear regression model or estimated regression coefficients and standard errors. Harrell (2015) suggested that 10 SPV was the minimum required sample size for linear regression models to ensure accurate prediction in subsequent subjects, Green (1991) suggested that 20 SPV would be preferable and that the minimum required SPV should be five. As a minimum SPV of 10 is required to test the assumptions of normality for the residuals and of homoscedasticity of variance, Harrell's minimum of 10 appears to be the lowest possible threshold (Harrell, 2015). And as the relationship between the power of a regression model and the sample size in terms of SPV is a curve with diminishing returns (Harrell, 2015), an SPV of 35 in the models applied seems adequate to ensure accurate prediction of Hg in tissues from fur levels in Arctic foxes.

While numerous studies have documented high concentrations of total Hg , which includes all forms of Hg in a sample including both inorganic Hg and MeHg in fur of Arctic predator, it is MeHg that is of primary interest as it is the Hg form that readily biomagnifies as it moves up trophic levels, and is a strong neurotoxin (Cardona-Marek et al., 2009; Chételat et al., 2020) and endocrine disruptor (Tan et al., 2009; Zhu et al., 2000). However, due to the high costs of the analyses, MeHg concentrations have rarely been quantified in tissue samples (Bechshoft et al., 2019). While the Hg ingested by top predators is predicted to be predominantly in the form of MeHg species-specific as well as individual capacity for detoxification may lead to a wide variation of Hg forms in different organs and tissues of mammals (Evans et al., 2016; Gamberg et al., 2015; Wang et al., 2014). Because MeHg is the toxic and biologically available form of Hg, it is imperative that additional studies are done to validate the methods and concentrations among tissues before hair can be reliably used as a matrix for assessing MeHg loads in Arctic carnivores (Bechshoft et al., 2020, 2019). Because a few early studies inferred that almost all Hg measured in fur was MeHg, e.g. in polar bears (Dietz et al., 2011; Eaton and Farant, 1982), it has been assumed that THg concentrations in hair accurately reflect THg and MeHg concentrations in other tissues. However, MeHg concentrations have rarely been quantified in Arctic predators, despite the above-mentioned studies in polar bears, and thus the distribution of different Hg forms in tissues and fur remains still unknown.

Most research assumes that THg is uniformly distributed across the fur in a pelt, but there are few studies which tested this assumption. A recent study showed an unequal distribution of both THg and MeHg across the pelts of four river otters (*Lontra canadensis*), with significant THg clusters in undercoat fur and to a lesser extent in topcoat fur (Eccles et al., 2019). Furthermore, Eccles et al. (2019) demonstrated that the error rate for predicting internal THg is lowest in the forebody region of the topcoat, at least in otters, thus making this the optimal region to sample for biomonitoring. Yet, hardly any data are available on the distribution in the pelts or typical proportions of mercuric forms in the different organs and fur of other Arctic predators, and no transformation predictive regression models are available for most Arctic top predators. Thus, further systematic investigations are warranted in order to improve the assessment of the distribution and proportion of inorganic and organic mercury forms in the soft tissues and fur of Arctic predators in different parts of the body. This would help to refine the toxicity models and risk assessment of particular Hg forms in Arctic predator species in the future.

9.4 STATE OF KNOWLEDGE ON THE BIOLOGICAL EFFECTS OF HG AND OHCS ON KEY ARCTIC WILDLIFE SPECIES

The increasing production and release of chemicals ensures that wildlife, people and ecosystems are continuously exposed to chemicals. Although high levels of legacy compounds are still reported, there has been little effort to quantify population level effects of pollutant exposure in Arctic wildlife despite the multiple health effects reported from field studies (Dietz et al., 2019, 2022). Accordingly, comparably little data are available on population level effects of mercury and OHCs in Arctic species. The weight of evidence provided by correlations of OHC and Hg exposure with various physiological and biochemical endpoints may identify certain contaminant hotspots, including East Greenland and Svalbard (Dietz et al., 2019). However, comprehensive reviews of OHC exposure and effects in Arctic wildlife showed there are virtually no data demonstrating a direct OHC- or Hg mediated cause-effect (Letcher et al., 2010; Sonne, 2010).

As discussed above, one reason is that understanding and predicting the real biological effects of chemicals and complex contaminant mixtures within a multi-stressor framework is one of the greatest challenges in ecotoxicology (Dietz et al., 2019). This is because many different agents, receptors, routes to exposure, endpoints and scales need to be considered (Townhill et al., 2022), which makes the risk assessment complex and complicated. To this end, single field studies often remain of limited explanatory power to identify adverse cause-and-effect mechanisms. However, examining correlative relationships between biological endpoint measurements and contaminant levels, using a large cross-species data set, offers a more powerful possibility to unravel causal links between exposure and biological effect. Such information is extremely important for the management and conservation of wildlife populations, and provides evidence to support the regulation of contaminant emissions. Thus, Chapter IV strived to update knowledge and highlight knowledge gaps in understanding the biological effects of legacy pollutants and chemicals of emerging concern on Arctic marine and terrestrial mammal, bird and fish species and populations which may serve as basis for conservation management. Previous assessments (e.g. AMAP, 2011; Dietz et al., 2013b) reported contaminant levels in Arctic species in order to compare them with levels known to elicit detrimental effects or in relation to toxicity thresholds. Such toxicity thresholds and endpoints for Arctic predators are currently grossly lacking in literature for most pollutants and CECs. One way to bridge this gap previously, is to estimate these endpoints and toxicity thresholds for untested wildlife species based on known ecotoxicity data (Forbes et al., 2016; Pavlova et al., 2016; Raimondo et al., 2007), from semi-field studies or observations of affected animals in the wild. In Chapter IV we went one step further by using risk quotient calculations, which allow the summing of the cumulative effects of OHC and Hg for which critical body burdens can be estimated for Arctic predators (AMAP/UN Environment, 2019; Pedersen and Petersen, 1996). The five risk categories proposed in Chapter IV reflect effects on reproduction in various Arctic marine and terrestrial mammals, fish and birds. These categories can now be applied in future studies reporting on Hg and OHC tissue or fur levels in Arctic marine and terrestrial mammals, fish and birds, in order to enhance our understanding of the cumulative effects of contaminants and their mixtures.

Chapter IV concludes that for certain species and populations, some OHCs and Hg tissue contaminant burdens reported post-2000 were observed to be high enough to exceed putative risk threshold levels that have been previously estimated for non-target species or populations outside the Arctic. While most marine mammal species are shown to be at no or low risk for health effects mediated by Hg or OHC exposure, for some species at high marine trophic levels, such as the polar bear, killer whale, pilot whale, narwhal and beluga as well as hooded seal, a proportion of the population is at high or severe risk for health effects mediated by Hg or OHC exposure. Several populations of raptors such as white-tailed eagle (*Haliaeetus albicilla*), gyrfalcon (*Falco rusticolus*) and peregrine falcon (*Falco peregrinus*) were shown to be at risk of PCB-mediated biological health effects, and thus should be further monitored in future studies. We recently published an update on the risk assessment of the health risk associated with Hg concentrations in 3500 individuals of 13 marine and terrestrial mammal species across the Arctic, showing that Hg exposure was low in most populations (Dietz et al., 2022). Exceptions were subpopulations of polar bears, pilot whales, narwhals, beluga and hooded seals which are still or increasingly exposed in geographic hotspots, raising concern about Hg-induced toxicological effects. Reasons for the high levels of banned OHC and Hg found in some Arctic wildlife may relate to reemissions from depositional sinks in the Arctic such as snow and ice (reviews by Brown et al., 2018; Lehnherr, 2014), or marine sediments, largely due to the settling of organic matter-bound chemicals through the biological pump operating in oceanic surface waters (Galbán-Malagón et al., 2012).

As discussed in Chapter IV, the developed methods for assessing OHC and Hg mediated risks in Arctic wildlife species were based on laboratory toxicity tests in rodents and were extrapolated to Arctic predator species. Therefore, substantial uncertainties remain. Small sample sizes, different methodologies, extrapolation from tissue to whole body levels, and the use of tissue-to-fur transformation equations across species preclude firm conclusions on the assessment of toxicity and health effects. Finally, additional information on the proportions of MeHg (as the most toxic form of mercury) and inorganic Hg in different tissues would be needed in order to increase the level of confidence in ecotoxicological assessments – these should be considered in future work.

Knowledge gaps with regard to the pollution status of terrestrial predators were identified, particularly in the Russian Arctic. Data gaps include the establishment of concentration thresholds for individual compounds as well as for realistic cocktail mixtures that in fact indicate biologically relevant, and not just statistically determined, health effects for species and subpopulations. Risks to wildlife populations are often based on oversimplified scenarios where predicted impacts are derived from exposure to a single chemical or stressor (AMAP, 2021b). Thus, the drivers of observed temporal changes, the complex interplay of synergistic, combined causes and effects and the ultimate consequences for the survival of Arctic wildlife populations remain unclear in most cases. Therefore, in future it will be necessary to assess exposure levels and their temporal changes and effects, including those for emerging chemicals, in key Arctic species. The endpoints assessed should be relevant to population viability, such as survival, reproductive success and population density (Nuijten et al., 2016), in order to increase the level of confidence and to correctly evaluate adverse effects on Arctic predator populations. Such efforts require a harmonisation of pan-Arctic studies in relation to target species, sampling frequency, season, and various methods for the measurement of chemicals and associated biomarkers and biological endpoints that are applicable to effect assessment.

10. RECOMMENDATIONS AND FUTURE PERSPECTIVES

10.1 IMPLICATIONS FOR FUTURE RISK ASSESSMENT

This thesis underscores that although established chemicals found to be hazardous to human and environmental health are being phased out or internationally regulated, concentrations remain high in Arctic biota of concern in many cases. Problems arise particularly from persistent chemicals, as unpredictably high levels may accumulate in humans, wildlife and the environment over extensive periods of time, which is why long-term effects are difficult to assess (Moermond et al., 2012). Hundreds to thousands of legacy and new chemicals are now being commercially used globally that are predicted to have persistent properties (AMAP, 2018) and may thus affect Arctic wildlife and people. Chemical pollution, in aggregate, now poses a threat to the integrity of ecosystems worldwide, including the Arctic, as pollution has recently been shown to surpass safe limits for humanity in terms of exceeding planetary boundaries (Diamond et al., 2015; Persson et al., 2022). The impacts of chemicals as multiple pollution pressures on pristine Arctic ecosystems are, astonishingly, not part of any working group's mandate and there are few experimental and observational datasets that include contaminants as multiple pollution pressures (Townhill et al., 2022). As shown for Hg and OHCs in Chapter IV, knowledge gaps on the toxicological effects of chemicals often stem from a lack of toxicity data under the different regulatory frameworks, which make the overall risks they pose difficult to assess. One reason is that acute or chronic toxicity data either do not exist as shown for instance for industrial chemicals (Springer, 2015) or are not publicly accessible due to reasons of confidentiality in case of pharmaceuticals (Oelkers, 2021). Chapter II and IV emphasise the critical role of environmental biomonitoring for recording chemical exposure and identifying unknown and unexpected effects on wildlife populations, which should prompt a timelier risk management and foster the development of an early warning system for the Arctic.

The findings of the thesis, and lessons learnt from the regulatory history of many POPs, call for a systematic consideration of ecotoxicological field and biomonitoring data on Arctic predators in regulatory risk assessments under different legislations. For instance, under the Stockholm Convention, there are four screening criteria that have to be met to establish that chemicals be recommended for listing under the annexes to the convention: they need to be persistent, bioaccumulative, have potential for environmental long-range transport and display adverse effects (UN, 2001). Evidence supporting the criteria of potential for long-range environmental transport includes observations that the chemicals are found at locations '*distant from sources*' or '*where monitoring data show that long-range environmental transport of the chemical … may have occurred*'

(UN, 2001). Monitoring data of chemicals in biota have become an important indicator for assessing long-range transport, persistence and bioaccumulation of chemicals under global and European chemical legislation (de Wit et al., 2019). Under REACH, for instance, all available data, including contaminant data from wildlife, can be used in a "weight of evidence" approach to decide whether a chemical meets bioaccumulative properties. Thus, academic studies and monitoring programs reporting the concentration of the respective chemical in top predator tissues are in principle a rich data repository for chemical risk management. Unfortunately, there is currently limited guidance on how to make use of tissue or organ concentrations reported in wildlife studies in a regulatory context and how tissue levels can be converted to whole body concentration values (EA, 2022; Treu et al., 2022b), which should be addressed by the regulatory community in future.

New analytical tools such as non-target screening approaches should come into play in order to identify, quantify and prioritise the CECs (Badry et al., 2022; Chiaia-Hernández et al., 2020; Hollender et al., 2019; Schymanski et al., 2015). These tools now allow biomonitoring to move beyond detecting single substances, so we can ultimately identify the effects of chemical mixture cocktails on wildlife species. A pan-Arctic list of priority CECs and their typical environmental mixtures detected in biota, people and environmental compartments would help to identify and prioritise chemicals for further regulatory scrutiny. Furthermore, ecological contexts and mixture toxicities should be more frequently considered in regulatory risk assessments to trigger risk management measures before adverse effects in individuals or populations start to manifest. To reduce pollutant exposure and impacts to protect the integrity of (Arctic) ecosystems in the long term, strict reduction of anthropogenic emissions of mercury, OHCs and other pollutants through a more rigorous legislation and a switch to sustainable and less hazardous chemical alternatives are inevitable.

10.2 ARCTIC POLLUTION IN FACE OF CLIMATE CHANGE

Climate change is reshaping the way in which pollutants and contaminants move through the global environment, to a large extent by changing the chemistry of the oceans and affecting the physiology, health and feeding ecology of marine biota (Alava et al., 2017). Recent mathematical modelling of food-web structures on a global scale suggests that global warming could also directly reduce the number of species, the proportion of basal species and the number of interactions while it indirectly increases omnivore levels, connectance and trophic level through its direct effects on the fraction and number of species (Gibert, 2019). There is also abundant evidence from observations and mathematical modelling to show that climate variation has an effect on POPs and Hg levels in biotic and abiotic environments and enhances the mobilisation of pollutants to marine and freshwater ecosystems (reviewed by Hung et al., 2022; Ma et al., 2016). All these climate induced changes will have wide ecological impacts on the Arctic and consequences for the extent of pollutant exposure as a response to altered release rates and biomagnification processes. The findings of this thesis complement the overall knowledge base linking dietary aspects, accumulation patterns and pollutant induced effects in Arctic predators. However, we still do not have sufficient information to fully appreciate the effects of pollutant exposure and climate change, which makes it difficult to predict, mitigate or adapt to such impacts. Thus, future efforts at the science and policy levels are urgently needed to improve our understanding of the connection between climate change and pollution. These efforts will form the cornerstone for a pro-active approach towards protecting Arctic ecosystems in the face of current and future environmental change.

11. GENERAL CONCLUSION

The ability of animals to respond to changes in their environment is critical to their survival, reproduction and their population persistence. In the Arctic, climate change and pollutant exposure are two important environmental threats to top predators. This thesis investigated Hg and OHC levels and their potential health effects in key Arctic predator species. It suggests absolute exposure to pollutants may be less important than the feeding ecology and feeding opportunities in the two model species. This may in turn influence their population health and stability. We found evidence that the decline of the Arctic fox population on Mednyi Island was associated with high Hg exposure levels related to the consumption of marine prey in coastal ecotypes. Still, additional drivers of the decline were not systematically investigated, mainly due to the lack of suitable historical study material. Overall, the results suggest conservation management of Arctic canids should consider dietary aspects. The combined use of multiple isotopes and novel techniques for chemical analysis are recommended for future studies in order to derive a refined estimate of dietary sources and pathways of pollutants in Arctic top predators. The thesis also confirms that fur is a suitable, minimally-invasive sample matrix for Hg assessment in Arctic canids helpful to conservation contexts. The derived regression model equations for the extrapolation of Hg tissue levels from fur allowed a direct comparison among studies and an evaluation of the exceedance of toxicity benchmarks. We suggest to increase sample size and refine the analysis of the proportion of inorganic and organic mercury forms in soft tissues and fur of Arctic predators in future research. This would allow to link effects to specific Hg forms, increase the power of the applied models and improve risk assessments. The review of literature post 2000 revealed that most Arctic marine and terrestrial mammal species are at no or low risk for reproductive or other health effects mediated by Hg and OHC exposure, though for some species at high marine trophic levels, a proportion of the population exceeded putative risk thresholds. Although Arctic wildlife is exposed to a broad range of OHCs and Hg and their mixtures, our understanding of their risks and possible chronic impacts, particularly in terrestrial Arctic predators, is far from complete. Endpoints assessed in future assessments should be relevant to populations – survival, reproductive success and population density. The thesis demonstrates that the existing chemical regulations of Hg and OHCs tend to be ineffective, as exposure still poses a risk to certain Arctic wildlife populations and the integrity of ecosystems. Particularly in view of climate change, emerging pathogens and recent exceedance of planetary boundaries by pollution, further investigations are needed to better understand the mechanisms and interplay of pollutant drivers and their effects on individuals and populations. Finally, to overcome the challenges facing the Arctic, we require interdisciplinary research approaches, fundamental advancements in risk assessment methods and, ultimately, stricter and faster chemical regulations. This requires efforts on all sides, the science, conservation and policy level.

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13. ANNEXES – SUPPLEMENTARY INFORMATION CHAPTERS I – IV

Chapter I: https://doi.org/10.1016/j.scitotenv.2022.156171

Chapter II: https://doi.org/10.1371/journal.pone.0060879

Chapter III: https://doi.org/10.1016/j.scitotenv.2017.10.143

Chapter IV: https://doi.org/10.1016/j.scitotenv.2019.133792