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Review

Arctic Freshwater Environment Altered by the Accumulation of Commonly Determined and Potentially New POPs

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Abstract: Chemical composition of Arctic freshwater ecosystems depends on several factors. They include characteristics of the surrounding landscape, its lithology, geomorphology, vegetation, and hydrological features, as well as accumulation of anthropogenic pollution. In the Arctic, the problem of environmental contamination is widespread. That is why research on lakes and river catchments in terms of their chemical composition has enjoyed increasing interest among scientists worldwide. The freshwater reservoirs of the Arctic are fragile and particularly vulnerable to the uptake of pollutants that become trapped in the water and sediments for an extended period. This review summarises selected studies of freshwater bodies in the Arctic to highlight the problem of the accumulation of pollutants in these reservoirs. Moreover, it emphasises the possible negative impact of chemical pollutants on both animal and human health.

Keywords: POPs; freshwater ecosystem; Arctic; pollution; contamination; long-range transport; current-use contaminants

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1. Introduction

Polar regions are characterised by a great abundance and diversity of freshwater ecosystems, including streams, rivers, lakes, and wetlands which are among the most pristine ecosystems worldwide. They are particularly common due to low evaporation rates, widespread permafrost, and extensive meltwater from snowfields and glaciers. Glacial action and river water inflow shaped the basins of many lakes and distributed glaciogenic deposits of different sources and mineral composition across their catchments [1]. The unique and fragile Arctic water ecosystems, adapted to extreme environmental conditions of the area, are under pressure due to environmental changes resulting from warming of the polar regions more than twice as intensive as the global average temperature rise [2,3]. Furthermore, increasing air temperatures affect freshwater supply from shrinking glaciers and permafrost thaw [4,5], effectively modifying the hydrological regime of the Arctic lakes and rivers, and contributing to the formation of groundwater storages [6]. Moreover, climate instability leads to variability of water chemical composition, and for lakes located close to the sea—changes in their linkages with marine waters [1].

Arctic areas also contain major water resources that have been widely exploited. The dams and reservoirs constructed for hydropower engineering have impacted many Arctic rivers and lakes [7], often leading to changes in water inflow and temperature [8]. In the past, the construction of dams has also interrupted the river continuum and has been responsible for the decline of many migratory fish populations and other biota species [7]. Rivers in the Arctic have also been used for the transport of timber from forested inland regions, resulting in dam construction and canalisation. Flood protection measures have also been instigated in some Arctic freshwater systems where infrastructure was at risk [8].

In polar regions, especially in the Arctic, rivers and streams remain in a close and interactive relationship with their catchments. The significance of these interplays varies with changes in terrestrial vegetation and the extent of permanent snowfields and glaciers [9]. Freshwater reservoirs in the European Arctic considerably vary in size, from large, wide lakes and rivers of northern Russia, to the multitude of small and medium-sized aquatic ecosystems typical of northern Scandinavia. Lakes in the Svalbard Archipelago are typically medium-sized, and rivers are short, but may seasonally display high flows as a result of snow and ice melt [8]. Water temperatures in Arctic streams, rivers, and lakes are constantly low and fall with growing altitude and latitude. Low temperatures in rivers, combined with high sediment load and channel instability, place glacier-fed rivers amongst the most inclement habitats for aquatic biota. Snow and ice cover as particular features of Arctic rivers, however, indirectly provide unique environmental conditions that lead to the development of many adaptive mechanisms of the biota living in Arctic rivers, streams, and lakes, even if winter conditions inevitably cause high mortality in reaches susceptible to the formation of frazil and anchor ice. Lack of nutrients or their limiting concentrations, as well as long periods of darkness and ice cover, contribute to reducing species richness, biomass, and productivity [6,10–12]. Furthermore, the poor nutrient status of freshwater ecosystems in the Arctic makes them particularly vulnerable to the uptake of contaminants [8].

Apart from a slight influence of local pollution sources that occur on Svalbard, such as military installations, industrial outlets, relatively small settlements, power generators, and vehicle and ships exhausts, the primary reason for the presence of pollutants in the Arctic is their transport from remote areas [13]. The Svalbard archipelago stands out among polar regions by its location as the gateway to the Arctic. Its location, relatively close to the European mainland, makes this sensitive region exceptionally exposed to the influence of pollutants [1]. Climate change enhances moisture transport with air masses from lower latitudes towards the pole, contributing to an increase in precipitation in the Arctic, falling either as snow or rain, and transporting a variety of pollutants [14]. Those contaminants persist for long enough to be transported thousands of kilometres with air masses and ocean currents. Consequently, compounds that have not been produced over the past several decades still occur in the Arctic environment, including freshwater ecosystems.

The objective of the present review was to evaluate the chemical composition, including contamination, of freshwater ecosystems in the Arctic. Countries worldwide are striving to reduce emissions of potentially toxic chemicals that may negatively impact the functioning of freshwater ecosystems and human health [15]. They attempt to reduce pollutants concentration levels in all environmental media. Pollutants accumulation has the potential to affect lakes' evolution through a variety of processes, particularly in polar regions which are sensitive to even slight alterations in air temperatures and the chemical composition of water [16]. Therefore, studying the pollution of freshwater ecosystems in the Arctic is an important research task worth carrying out over a long period of time.

2. Transport of Pollutants into the Arctic

Pollution, as a topic concerning not only urban areas on Earth, but also extending to polar regions, has been discussed in environmental sciences for more than 50 years. Already in 1964, Rachel Carson pointed out the considerable adverse effects of pollutants in polar regions. Pollution in the Arctic atmosphere has been investigated since the 1970s when first atmospheric measurements revealed the presence of persistent organic pollutants (POPs) in the pristine polar environment [17,18]. Comprehensive research under the Arctic Monitoring and Assessment Programme (AMAP) showed that winds and ocean currents provide effective means of transport of contaminants to the Arctic. Therefore, the atmospheric POPs distribution and fate has been continuously surveyed since the early 1990's [19].

Atmospheric long-range transport of pollutants to the Arctic is a complex process. Some indication for atmospheric long-range transport has been recorded much earlier—in the 1950s when aeroplane pilots reported smog-like phenomena over the surface of the Arctic region during their flights across the North Pole. This smog may still be seen during intercontinental flights. The smog phenomenon is mainly caused by sulphate-containing dust particles originating from industrial regions in the USA, Europe, and Asia, transported via the atmosphere, and deposited in the Arctic. This phenomenon is also known as 'Arctic haze' [20]. One of the most common processes in the atmospheric transport of POPs is global distillation. It forces contaminants out of warm reservoirs in temperate industrial areas into the cold environment in the Arctic, resulting in a complex POPs distribution cycle. Next to ocean currents and rivers, the atmosphere is considered one of the most important pathways for the transport of pollutants. Moreover, depending on the physicochemical properties of contaminants, water-soluble compounds may be transported in particle-bound or colloidal form in the atmosphere before deposition. Minor differences in volatility and solubility of pollutants can make a significant difference for the preferred transport pathway [20]. Therefore, small temperature changes will determine whether the substance will be associated with the gas phase or particulate/colloidal phase during atmospheric long-range transport (LRAT) events [21]. Furthermore, chemical compounds tend to vaporise at elevated ambient temperature and condense at lower air temperatures at higher latitudes. This phenomenon has been described as global distillation [22–24]. The Arctic is not considered a highly pristine environment anymore. Certain characteristic features, such as low temperatures, ice coverage, snow and rain precipitation, and extended periods of darkness during winter make the Arctic environment particularly vulnerable to the accumulation of globally transported contaminants, including POPs [12]. Figure 1 presents parameters favouring the special potential of the Arctic for the uptake of pollutants [20].

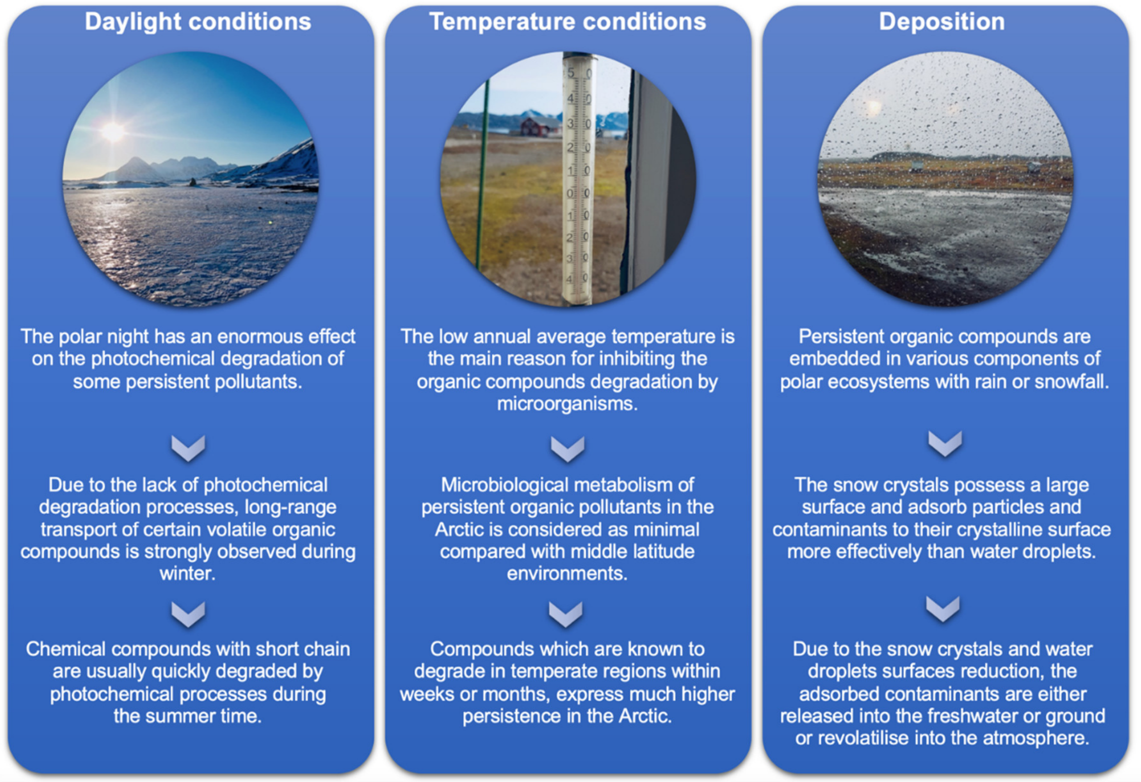

Figure 1. Parameters favouring the potential accumulation of pollutants in the Arctic [20]. All photographs appearing in the article come from the authors' private archive.

It is already common knowledge that POPs can migrate long distances from their original sources. Inputs of POPs from the atmosphere and surface waters, and releases from sediments and removal pathways such as volatilisation and sedimentation, can partly explain why countries that banned the use of certain persistent pollutants are experiencing less dramatic declines in environmental concentrations nearly a decade later [25].

3. Persistent Organic Pollutants in the Arctic Environment

Research conducted under the Stockholm Convention to identify POPs of potential global concern revealed nearly 40 substances in three categories [26]:

- Industrial chemicals;
- By-products;
- Pesticides.

Many substances belonging to these categories have been the subject of already existing research, and are comprehensively represented in the literature. They include polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dioxins/furans, and organochlorine pesticides (OCPs). Nonetheless, other classes of substances have not received the same level of investigation, or are only now being recognised as possible POPs [26]. Figure 2 shows examples of compounds that are not commonly included in chemical monitoring programmes, but appear in the Arctic environment, and are listed below [27]:

- Novel brominated and chlorinated flame retardants (NBFRs. NCFRs);
- Polyfluoroalkyl substances (PFASs);
- Pharmaceuticals and Personal care products (PPCPs);
- Current-use pesticides (CUPs).

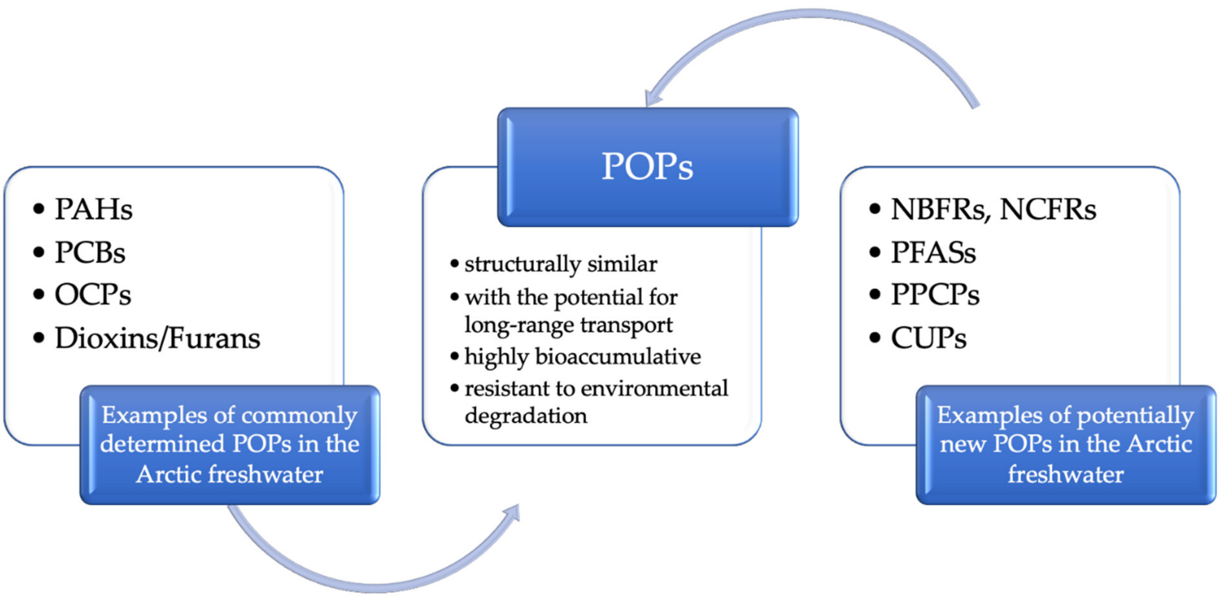

Figure 2. Examples of a wide variety of pollutants that may be considered as POPs.

3.1. Freshwater Bodies as Receivers of Commonly Determined POPs

Some of the processes occurring in freshwater environments have already been observed and provided the basis for the investigation of the fate and transport of POPs. As mentioned above, POPs have the potential to be distributed between particles, colloids, and the water phase. Deposition across the air-water interface is the primary input route for pollutants to the freshwater environment. Another way of supplying pollutants into freshwater is through melting snow and ice [28,29].

POPs may also sorb to particles and colloids, resulting in a decrease in freely dissolved concentrations, and therefore a reduction of their bioavailability to aquatic organisms [26]. Sorption of POPs to colloids, however, may enhance environmental transport due to the higher mobility of small colloids compared to larger ones. The extent of sorption to particles and colloids, and the distribution between various phases in freshwater reservoirs, have a significant impact on POPs migration in the Arctic environment, and their fate processes such as sedimentation, bioavailability, and degradation [30]. Table 1 shows selected freshwater bodies in the Arctic, including lakes, ponds, rivers, and streams, as well as lake sediments, where different organic pollutants have been detected.

Table 1. Examples of commonly detected organic pollutants in the freshwater environment of the Arctic.

Freshwater Receiver of POPs	Location/Area	Polycyclic Aromatic Hydrocarbons	Polychlorinated Biphenyls	Organochlorine Pesticides	Dioxins/ Furans	Other Organic Pollutants	Reference
			Svalbard				
lake water	Hornsund	+	+			+ (HCHO, ∑Phenols)	[1,11–13,23,29,31–33]
river/stream water	Hornsund	+	+			+ (HCHO, ∑Phenols)	[11–13,15,23,29,31–33]
lake water	Bellsund	+				+ (HCHO, ∑Phenols)	[34]
river/stream water	Bellsund	+				+ (HCHO, ∑Phenols)	[35–37]
lake sediments	Ny-Ålesund	+	+	+	+		[38]
river/stream water	Ny-Ålesund	+					[39]
river/stream water	Longyearbyen	+				+ (HCHO, ∑Phenols)	[40]
lake sediments	Bjørnøya		+	+	+		[41,42]
several lakes sediments	West coast of Svalbard	+	+				[43]
several lakes sediments	Coast of Wijdefjorden	+	+	+			[44]
			The other Arctic regions				
lake sediments	Canadian Arctic	+	+	+	+		[45–48]
lake water	Northern Sweden					+ (HCBDs)	[49]
lake sediments	Alaska	+	+	+			[50–52]
lake sediments	Northern Norway		+				[53]
lake sediments	Northern Russia (Siberia)		+				[53]

Many scientific papers regarding the accumulation of pollutants in freshwater at various locations in the Arctic have been included in the table above. This shows the extent of research and the importance that scientists attach to the topic of pollution of the polar environment. The authors of this review, who research POPs concentration levels in Svalbard, also contribute to the assessment of the state of the Arctic environment. Figure 3 shows the location of areas where research has been carried out by the authors of this review article over the last five years.

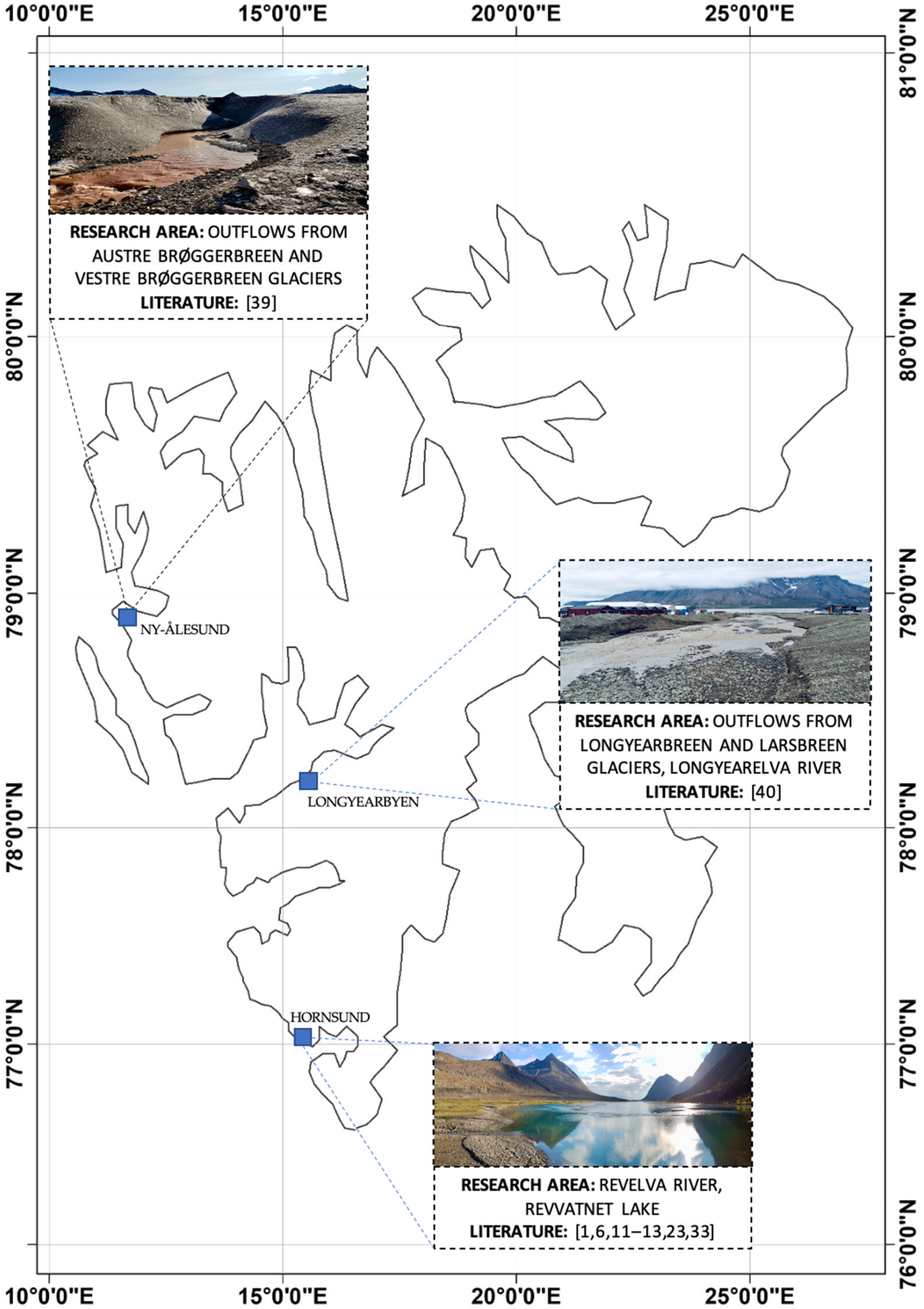

Figure 3. Location of areas where the authors of this review article have been conducting their research on POPs (mainly PAHs concentrations in freshwater). All photographs appearing in the article come from the authors' private archive.

Three areas in the Svalbard archipelago (Hornsund, Longyearbyen, and Ny-Ålesund) selected for research by the authors are exposed to both long-range transport contamination and local pollution. While exposure to external factors has been known for a long time, local pollution may be of as much importance there as long-range contamination. The selected locations are settlements of residents of Svalbard and scientists conducting research. In the case of Longyearbyen, the capital of Svalbard, mining, power production, transport, waste and sewage disposal all contribute to local pollution [29,40]. Nonetheless, knowledge gaps regarding the exact impact of the local human activities still exist, particularly with respect to their spatial coverage, chemical contamination profile, and temporal trends. The Hornsund and Ny-Ålesund research stations are also important points on the pollution map of Svalbard. The surroundings of all stations are subject to multiple chemical analyses (e.g., water, air, and soil research), and are regularly monitored by the station operators. Despite the consistent effort to limit contamination connected to the operation of the Arctic research stations, they are not zero-emission facilities. The chemical diversity introduced by all these factors requires continuous investigation [29].

Characteristic Features and Sources of POPs in the Arctic Freshwater Ecosystem

In many water ecosystems around the world (including the Arctic), climate change and environmental contamination affect water quality and water-related ecosystems. Persistent organic pollutants represent one of the most intensively studied classes of environmental contaminants due to their well-known cancerogenic and mutagenic effects on organisms. They are generally ubiquitous, lipophilic, and accumulate in lipid tissues and through food chains [54]. Commonly detected POPs in the Arctic environment include:

- Polycyclic aromatic hydrocarbons (PAHs) belonging to the group of organic compounds consisting of 2 to 13 aromatic rings. PAHs are weakly volatile, and dissolve in water, with solubility decreasing with an increase in the number of aromatic rings. They are chemically inactive and have low vapour pressures, therefore bonding to particulate matter. They are also highly thermo- and photosensitive when adsorbed on the surface of dust [26]. The Integrated Risk Information System (IRIS) of the U.S. Environment Protection Agency (EPA) contains an assessment of over 540 individual chemicals with potential human health effects. Among them, 17 unsubstituted PAHs have been identified by EPA as priority pollutants [55,56]. PAHs transported to the Arctic (via the atmosphere and ocean currents) are generated as a result of incomplete combustion of materials containing carbon and hydrogen, which includes coal, crude oil, fuel, gas, wood, and organic materials, as well as combustion of polypropylene and polystyrene, communal and industrial waste, and used tires [57]. Emission of PAHs from natural sources includes volcanoes, forest fires, or industrial sources such as stack emissions and combustion [26]. Releases of these compounds to freshwater include industrial and wastewater treatment plant discharges, precipitation of industrial and natural dust particles, and urban runoff [58,59];
- Polychlorinated biphenyls (PCBs) are a class of chlorinated derivatives of aromatic organic compounds with 1–10 chlorine atoms attached to biphenyl which is a molecule composed of 2 benzene rings. They are described by various trade names such as Aroclor, Askarel, Phenoclor, Clophen, Kanechlor, and Therminol [10]. The empirical formula of PCBs is $C_{12}H_{10}$-xCl_x. It comprises mixtures of 209 possible synthetic organic chemical congeners, ranging from oily liquids to waxy solids [10]. PCBs transported to the Arctic and accumulated in freshwater mainly originate from their industrial application as flame retardants in paints, additives for insulation purposes, and dielectric fluid in capacitors and transformers. Between 50 and 100 congeners are generally released into the environment during the destruction and decommissioning of electrical equipment and buildings. PCB congeners show a very wide range of physicochemical properties that dictate their transport pathways and environmental fate. They are considered Arctic indicator contaminants for trend and risk assessments [20,54];
- Organochlorine pesticides (OCPs) are organic compounds attached to 5 or more chlorine atoms. They represent one of the first categories of pesticides ever synthesised, and are used all over the world. OCPs belong to the group of chlorinated hydrocarbon derivatives with a high variety of applications in the chemical industry

and agriculture. These compounds are known for their slow degradation, high toxicity, and bioaccumulation. Even though many of the compounds belonging to OCPs have been banned in developed countries, the use of these agents has been on the rise. This particularly concerns abuse of these chemicals across the continents. Although pesticides have been developed to target organism toxicity, the non-target species are often negatively affected by their application [60]. All pesticides may also be transported over long distances, and can be trapped in cold Arctic water reservoirs [10];

- Dioxins/furans are a group of chemical compounds that share chemical similarities and mode-of-action (biological) characteristics. A total of 30 of these dioxin-like compounds belong to closely related families: polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and certain PCBs. PCDDs and PCDFs are generated as unwanted by-products of chemical syntheses, but may also be inadvertently produced in nature. Other sources of these xenobiotics include combustion, chlorine bleaching of pulp and paper, and many other industrial processes [55]. Like other organic pollutants, dioxins and furans are transported to the Arctic from distant regions.

3.2. Occurrence of Potentially New Classes of POPs in the Arctic Environment

Several environmental studies have already approached the identification of potentially new contaminants occurring in the Arctic using mathematic modelling of the probability of long-range transport, taking into account the physicochemical properties of various compounds and their production volumes [27]. One of the developments in the assessment of potential new classes of POPs in the Arctic has been the screening of chemicals in commerce by means of models that indicate that many current-use organic compounds have chemical characteristics that make them similar to commonly known POPs. They, therefore, have the potential to be transported to the Arctic [61]. The study by Brown and Wania, 2008 [61] employed a data set of more than 100 000 distinct industrial chemicals subjected to screening system models, and identified 120 high production volume chemicals that are structurally similar to already known Arctic contaminants and have partitioning properties allowing for considering them potential Arctic pollutants. Muir and Howard, 2006 [62], using 11,000 organic chemicals from the Canadian Domestic Substances List, also identified 28 chemicals with long-range transport potential based on predicted atmospheric oxidation half-lives (>2 days). Among these 28 compounds, only 8 were thought to have been measured in environmental samples [27]. Combining modelling for long-range atmospheric transport or oceanic transport and food web bioaccumulation seems to be the best approach for screening thousands of chemicals in commerce [63]. This information is crucial to the ongoing consideration of including new pollutants in the existing national, regional, and global agreements aimed at the regulation of emissions of POPs. Moreover, model predictions always require confirmation through targeted environmental measurement campaigns. Compounds that are not commonly covered by chemical monitoring programmes, but occur in the Arctic environment, include:

- Novel brominated and chlorinated flame retardants (NBFRs. NCFRs) constitute approximately 25% of all commercially used flame retardants. They are a large group of chemicals used in different materials to delay or prevent flaming. Since the ban of commonly used flame retardants, novel brominated and chlorinated flame retardants have emerged [64]. NBFRs and NCFRs are emerging contaminants that occur in these materials and can reach the distant polar environment. They are relatively new on the market and show many different properties. They are therefore applied in a wide range of products, such as furniture, plastic, textiles, foams, and electronic devices. The European Food Safety Authority (EFSA) published a report on emerging and novel brominated flame retardants in food, but could not perform the risk assessment due to limited data on their toxicity [65]. The European Commission (Directive, 2014/118/EU) has issued recommendations regarding monitoring traces of BFRs in food, including several NBFRs (such as 2-ethylhexyl 2,3,4,5-tetrabromobenzoate) [64]. The majority of countries have no laws to monitor the use of NBFRs or NCFRs. The production of these compounds in the United States is registered and reported by Chemical Data Reporting (CDA), part of the U.S. Environmental Protection Agency (EPA). Data on the production volumes of NFRs, however, are often non-existent. The European Chemicals Agency (ECHA) registers and monitors data on chemicals used in the EU [66], and EFSA has recommended monitoring of NFRs in food [64];

- Polyfluoroalkyl substances (PFASs) whose occurrence and fate in the aquatic environment is recognised as an important emerging contaminant issue. They are persistent and bioaccumulative chemical compounds [26]. They have been widely used in numerous industrial and commercial applications since 1950 [67]. The carbon-fluorine bond is strong and stable, and the chemical and thermal stability of PFASs provides for highly useful and enduring properties. As a consequence of their widespread use, PFASs have been detected in the environment, wildlife, and humans. The global regulatory community is interested in 'long-chain' perfluoroalkyl acids and perfluoroalkyl carboxylic acids, and their corresponding anions [26]. PFOS and PFOA are the two 'long-chain' perfluoroalkyl acids most often reported in the scientific literature [26,67]. An important research topic, directly related to environmental fate and transport, is the question of how fast PFOS and PFOA themselves and their homologs and precursors are transported away from their emission sources over long distances in air and water [68,69];
- Pharmaceuticals and Personal care products (PPCPs)—A diverse collection of thousands of chemical substances, including prescription and over-the-counter therapeutic drugs, veterinary drugs, fragrances, sunscreens, detergents, and cosmetics. Among PPCPs, some compounds are capable of disrupting the endocrine system of animals, including humans, wildlife, and fish. These substances are termed endocrine-disrupting chemicals (EDCs). PPCPs suspected to have EDC properties are considered to be an emerging class of contaminants, and may behave similarly to POPs. Even though PPCPs are not formally listed as persistent organic pollutants under the Stockholm Convention, and there is an active debate regarding whether PPCPs fall into the category of POPs, they have become emerging contaminants of concern because of their potential to affect drinking water supplies and their uncertain consequences of chronic low-level exposures of wildlife [26]. Municipal wastewater, attributed to the widespread use of PPCPs both in health care and personal care facilities and at home, is the primary pathway by which chemicals from prescription and over-the-counter products find their way into the aquatic environment, and may be transported to polar regions [70];
- Current-use pesticides (CUPs) continuously discovered in remote regions. They are sufficiently persistent to undergo long-range transport, and like other contaminants commonly known as POPs, they may represent an environmental concern. Current-use pesticides reported in the Arctic span diverse structural classes, although they share several general characteristics. Many of them exhibit a moderate to low solubility in water, and relatively low air-water partitioning (log K_{AW} values ranging from −3.5 to −6.0), allowing them to reach the Arctic, primarily from the atmosphere with some possible contribution via ocean currents [71]. According to the most recent report from the Food and Agriculture Organisation (FAO), 4.1 million tonnes of pesticides were used globally in 2016, with herbicides representing their largest share [72]. In the northern hemisphere, pesticide usage has increased by 27% since 1996, reaching 3.2 million tonnes in 2016. Reports regarding the use of individual chemicals within countries are scarce, making it difficult to identify the source areas and global trends of their production and use. Nevertheless, most CUPs of Arctic

concern have been recognised as high production volume (HPV) chemicals produced or imported in amounts greater than 1000 tonnes per year [73]. Current-use pesticides detected in the Arctic fall under varying levels of regulation [74–76]. Most of them are still approved for use, whereas the use of some (e.g., chlorpyrifos and pentachloronitrobenzene) is subject to restrictions, and others (e.g., endosulfan and dicofol) are beginning to be subject to domestic and international regulations. In 2011, endosulfan was added to the Stockholm Convention List of POPs. In 2017, the POP Review Committee (POPRC) recommended listing dicofol in the Conference of the Parties to the Stockholm Convention [77,78]. In 2010, endosulfan and trifluralin were under review for inclusion in the UNECE Long-Range Transboundary Air Pollution (LRTAP) Convention [79], but no further action has been taken [71].

Due to the increasing amount of research on potentially new classes of POPs in the Arctic, including freshwater research, Table 2 shows examples of current-use compounds that have been detected in selected freshwater bodies and sediments.

Table 2. Examples of potentially new classes of POPs detected in the freshwater environment of the Arctic.

Freshwater	Location/Area	Potentially New POPs	Reference
lake water	Canadian Arctic	CUPs: Chlorpyrifos, Chlorothalonil, Dacthal, α-Endosulfan, PCNB, Trifluralin	[80–83]
		PFASs: PFOA, PFNA, PFDA, PFOS, PFBS	[84]
		PPCPs: Naproxen, Ramipril, Azithromycin, Ciprofloxacin, Sulfamethoxazole, Lincomycin, Trimethoprim, Anhydro erythromycin A, Ceftiofur, Sulfamethazine, Carbamazepine, Oxcarbazepine, Venlafaxine, Desvenlafaxine, Diphenhydramine, Fexofenadine, Ibuprofen, Salicylic acid, Acetaminophen, Docusate, Atorvastatin, Levothyroxine, Amlodipine	[83]
		NBFRs. NCFRs	[85]
	Greenland	CUPs: Chlorpyrifos, α-Endosulfan, β-Endosulfan, Endosulfan sulfate, Dicofol, Methoxychlor	[86]
	Northern Russia (Siberia)	CUPs: Lindane, α-Endosulfan	[87–89]
	Svalbard	PFASs: PFBA, PFOA, PFNA, PFDA, PFOS	[90]
river/stream water	Svalbard		
lake sediments	Canadian Arctic	CUPs: Endosulfan sulfate	[91,92]
		PFASs: PFOA, PFDA, PFBS, PFOS	[93,94]
		PFASs: PFOA, PFNA, PFDA, PFOS, PFBS	[84]
		NBFRs. NCFRs	[85]
	Greenland	PPCPs: Salicylic acid, Ibuprofen, Diclofenac, Naproxen, Lidocaine, Paracetamol, Metformin, Metoprolol, Atenolol, Furosemide, Amiloride, Dipyridamole, Citalopram, Venlafaxine	[95]
wetland	Canadian Arctic	PPCPs: Atenolol, Carbamazepine, Sulfamethoxazole, Trimethoprim	[96]

4. POPs as Environmental Risk Factors in Remote High-Altitude Freshwater Ecosystems

Continuous and comparable monitoring programmes regarding the transport and occurrence of persistent organic pollutants in remote regions are essential for a better understanding of the global movement of these chemicals, and the evaluation of the environmental risk factors of POPs. Although the presence and altitude-dependent increase in POPs levels in the Arctic ecosystems (including freshwater) are confirmed by many international studies, their ecotoxicological consequences still remain largely undiscovered [97]. Arctic lakes are fragile ecosystems with significant potential for pollutants accumulation. They are considered among the most pristine waters of the Arctic. Therefore, even small amounts of contaminants greatly alter the chemistry of freshwater and sediment characteristics [1,98]. Freshwater ecosystems are identified as crucial 'early warning' sites with respect to global and medium-range transport, as well as the distribution processes for POPs. Moreover, the presence of anthropogenic contaminants in these vulnerable ecosystems may disturb their sensitive freshwater ecological balance [97].

The role of high-altitude lake systems as indicators of ecotoxicological risk has already been described in the context of monitoring and assessment of freshwater acidification and trace metal contamination. For POPs, high-altitude freshwater fish species are considered valuable indicators for ecotoxicological risk assessment of the Arctic [99,100].

Freshwater sediments and fish species usually act as sinks for atmospherically transported POPs that are deposited with rain and snow precipitation, or introduced with snowpack melt to freshwater bodies [101,102]. Particularly freshwater salmonid fish species have shown to respond very sensitively to elevated POPs concentrations [97,103]. A preliminary attempt at an ecotoxicological risk assessment has been made for selected POPs in freshwater fish based on various data comparisons. The research revealed that freshwater fish from the Arctic environment are almost equally contaminated with POPs as fish from mountain lakes and aqueous systems in other parts of the world [97,99,100,103].

According to the current scientific knowledge on the environmental behaviour of POPs in remote pristine environments, a great variety of effects caused by POPs exposure may be expected in high-alpine ecosystems, should highly elevated pollutants be found in 'hot spot' environments. Indeed, ecosystem-specific structures (e.g., geological structures, vulnerable food webs, or temperature adaptations) can reduce the effect threshold for organisms adapted to high altitudes, as comprehensively evidenced for the Arctic ecosystem [3]. Monitoring of the biological effects should cover immune system suppression and/or endocrine disruption for species at high trophic levels of remote high-latitude food webs. Although the original damage caused by chemical contamination such as POPs is often found at the molecular level (e.g., immune system-related effects, endocrine mimicking, induction of mixed functional oxidases, neurotoxic effects), emergent effects are also expected at the population level, such as the loss of genetic diversity [97].

5. Environmental Factors Determining Changes in POPs Occurrence in the Arctic over Time

Persistent anthropogenic pollutants are transported via the atmosphere, ocean currents, and rivers into the Arctic. After entering the polar environment, the chemicals are immediately redistributed within the regions by the same pathways. They can also be incorporated into biological systems through accumulation in the food webs. Every step along these transport pathways to and within the Arctic ecosystems is influenced by the currently observed climate change through reactivity, adsorption, and temperature-dependent processes. At the same time, the composition and geophysical properties of transport media are expected to change. Sea and land ice are already changing, and so is the extent and composition of potential intermediate storage compartments for pollutants, such as freshwater reservoirs and terrestrial elements [20]. To sum up, changes in global climate and the associated environment transformation in the Arctic are expected to have significant consequences for contaminant pathways [104].

The cryosphere of the polar regions, both Arctic and Antarctic, are currently undergoing comprehensive climate change-related transformations. They are therefore considered early warning regions for the scientific elucidation of environmental consequences related to climate change. This also includes the elucidation of the fate of atmospheric long-range transported POPs. It is not surprising that many research-based findings on POPs and climate change reported in the current scientific literature are based on study results from polar regions [104]. Figure 4 shows examples of potential effects of climate change on pollutants fate and distribution.

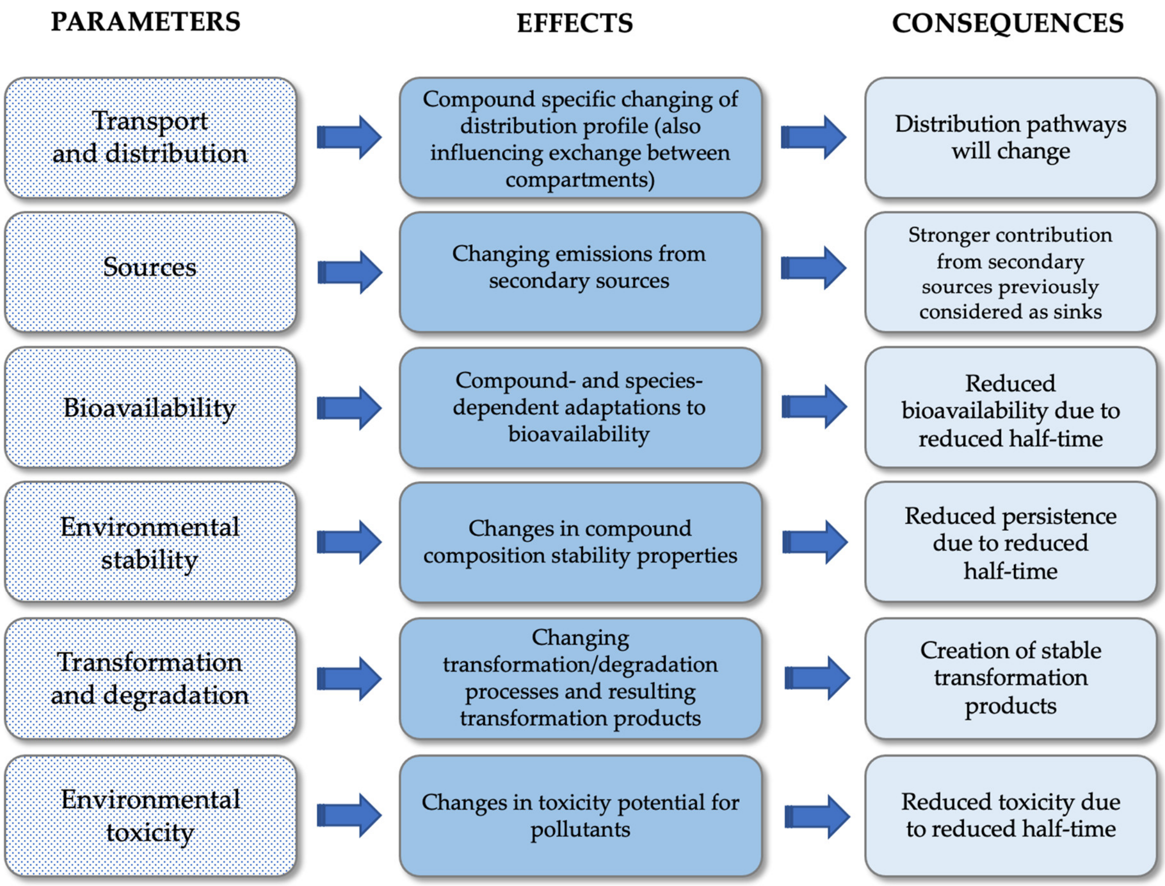

Figure 4. Expected effects of climate change on environmental processes related to transport and distribution of POPs [20,104].

Over the next decades, seasonal sea ice coverage is expected to disappear during the Arctic summer as a consequence of the overall observed ambient temperature increase. Therefore, remobilisation of previously-stored contaminants (secondary sources) is expected to significantly intensify in the coming years, leading to increased future exposure of local indigenous human populations and wildlife over certain time periods. Climate change is also expected to have a significant impact on complex global contaminants pathways, distribution processes into and within the Arctic, and storage [20,97].

6. Conclusions and Perspectives

Arctic freshwater ecosystems are quite varied in terms of their type, physicochemical characteristics, including proximity to the ocean and geology, their associated biota, and type of terrestrial vegetation in the surrounding catchment, as well as anthropogenic pollution [1]. Due to this, the impact of climate change, accumulation of persistent organic pollutants, and increased UV radiation levels will be variable and highly specific to particular freshwater reservoirs. These water bodies are particularly sensitive to many climatic alterations due to numerous hydro-ecological processes. The occurring processes may gradually adjust to changes in climate, or abruptly as environmental or ecosystem thresholds are exceeded. This is particularly the case for cryospheric components that significantly affect the water cycle of lakes, rivers, and ponds, the habitat characteristics of these freshwater systems, and the flora and fauna that occupy them. Prior to considering the specific effects of climate change on the Arctic freshwater systems, it is useful to place the climate projections generated by the five ACIA-designated atmosphere-ocean general circulation models (AOGCMs) for the Arctic as a whole into a more suitable freshwater context. For the most part, this requires focusing on model projections for the major Arctic terrestrial landscapes, including freshwater systems [104].

Freshwater reservoirs, including lakes, rivers, streams, ponds, wetlands etc., contain a wide range of POPs, as described in this review article. These pollutants, although under considerable restrictions and even banned for decades, still appear at significant concentration levels in polar regions. Monitoring of POPs originating from long-range atmospheric transport should be considered a versatile tool for the evaluation and assessment of the compound-specific distribution and transport processes and recognised by the international scientific community as well as by regulatory authorities [20]. Furthermore, along with the continuous development of new technologies and techniques for ultra-trace quantitative analysis of POPs, a lot of new chemicals are classified as compounds of emerging concern and introduced into the list of priority substances to be evaluated as potentially persistent, toxic, and bioaccumulative. For this reason, the research focus is slowly moving from the 'legacy POPs' towards 'new POPs' of emerging concern in polar freshwater ecosystems. Moreover, the impact of climate change on the transformation patterns of POPs in the Arctic is developing into a priority research for polar environmental scientists. Local investigations on POPs distribution patterns reveal expected changes in source strength, distribution, and deposition pathways in Northern environments [104]. Moreover, while emphasising the ecotoxicological risk of POPs the Arctic is continuously exposed to, it should be mentioned that international research efforts and funding priorities are urgently needed today to explicate this risk [97]. Arctic studies on persistent organic pollutants can also be expected to provide a new scientific insight that can be extended to understanding climate change impacts on contaminants in lower-latitude regions [104].

Even after more than 50 years of research, monitoring of POPs and compounds of emerging concern is still a crucial research topic for scientists worldwide. Long-term monitoring data sets from national and international Arctic monitoring programmes will continue to allow for in-depth interdisciplinary research on contaminants pattern profiles, temporal and spatial trend studies, environmental distribution and fate modelling, as well as an understanding of regional transport pathways in the future [20]. Many environmental scientists are hopefully encouraged to contribute their knowledge and expertise to the persisting substantial scientific gaps identified for this research area.

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