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Persistent organic pollutants: towards a POPs-free future

Contents

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Images

Page 3: Agricultural machinery spraying the crops with pesticides in springtime, Belgium © istock / brytta
Page 6: Pesticide spraying in a fruit orchard © istock /wellphoto
Page 7: A polar bear eating in Greenland © istock / Photon-Photos
Page 8: House sparrow / Felix Broennimann CC0, pixabay
Page 10: Seafood on ice © istock / AlexRaths
Page 13: Maktaaq delicacy (bowhead whale blubber) by Ansgar Walk / CC-BY-2.5, Wikimedia Commons, 2002. https://commons.wikimedia.org/wiki/File:Maktaaq_2_2002-08-10.jpg
Page 17: Arctic shipping / 12019 CC0, pixabay
Page 19: Navigating the Antarctic / clara5656 CC0, pixabay
Page 21: Le Rhône entre Viviers et Donzère (Ardèche/Drôme, France) By Babsy (Own work) CC BY 3.0, via Wikimedia Commons
Page 25: North Sea shrimp net / Heibe CC0, pixabay
Page 26: Chemical hazard pictograms © istock / Antoine2K

This Future Brief is written and edited by the Science Communication Unit, University of the West of England (UWE), Bristol
Email: sfep.editorial@uwe.ac.uk

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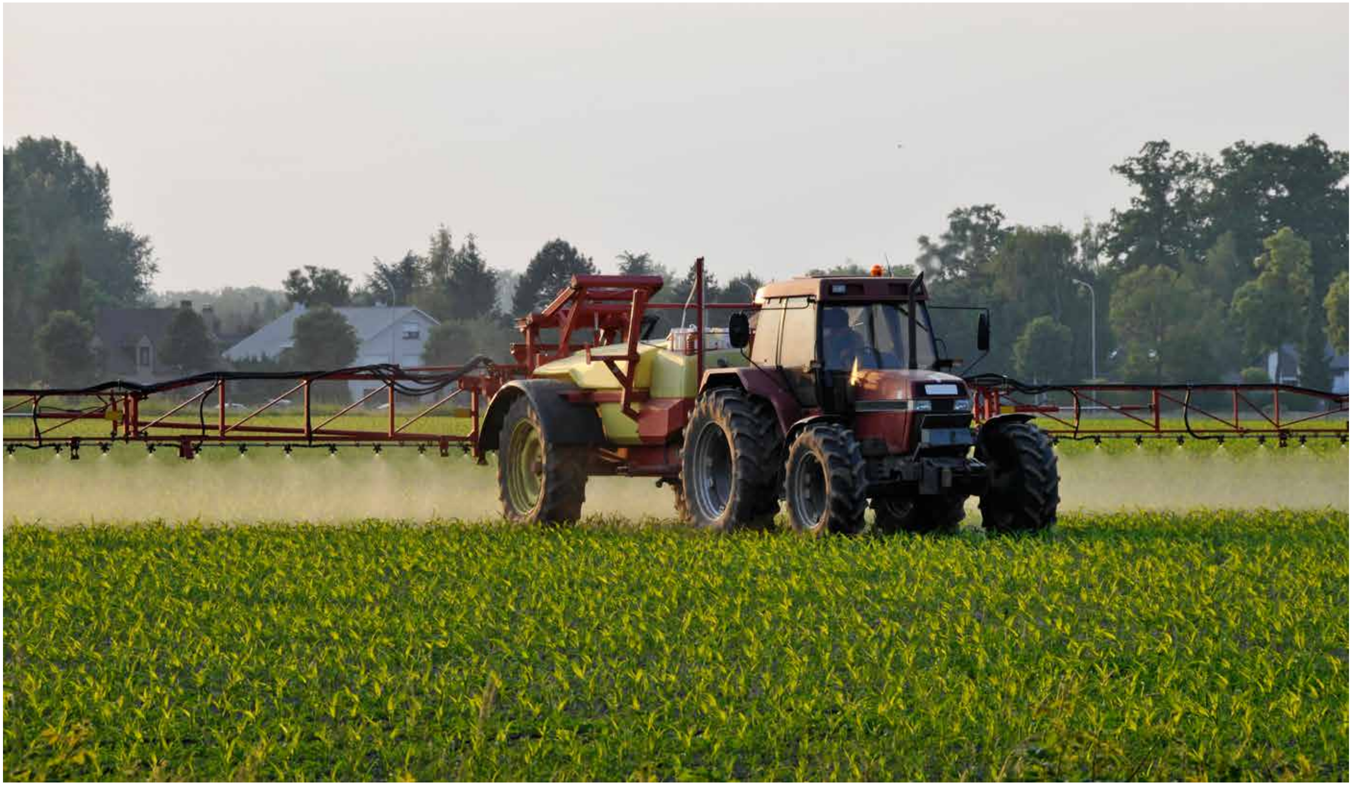
Introduction

POPs: a persistent problem, pole to pole

The majority of persistent organic pollutants (POPs) identified until now are banned or restricted around the world owing to concerns about their harm to ecosystems and human health. However, this is not the end of the story; even long-banned POPs still linger in the environment; others are still in use and are being directly emitted; and new POPs may be identified for which we have limited information. This Future Brief from Science for Environment Policy presents recent research into POPs' potential impacts, the levels and future outlook for POPs in the environment and humans, and how we can reduce our use of POPs.



Agricultural machinery spraying the crops with pesticides in springtime, Belgium.

As their name suggests, POPs are organic (carbon-based) compounds which do not break down easily in the environment. They accumulate through the food web, reaching peak concentrations in species at the top of the web, and pose a risk of harm to human health and wild animals. POPs are transported long distances through natural atmospheric and oceanic processes, across international boundaries and far from their original sources. They contaminate remote regions where they have never been used or produced, and thus present a threat the world over.

Many POPs were once commonly produced for a range of purposes including agriculture, disease control, manufacturing and industrial processes. However, even banned POPs remain in the environment for many years, mainly due to their persistence but also due to inappropriate management of decommissioned products, such as old electrical equipment containing polychlorinated biphenyls (PCBs). In addition, some POPs are still being directly emitted: polycyclic aromatic hydrocarbons (PAHs) are released into the atmosphere through combustion, from vehicle exhaust fumes, for example.

A number of POP substances are long-recognised as harmful and controlled under international regulation since at least 2003 (see **POPs regulation in the EU**, on page 5) — and much earlier in a number of cases. Perhaps some better-known examples include DDT, once used as an insecticide and now used only for disease vector control in some non-EU countries, and the class of industrial chemicals known as PCBs. These are often referred to as 'legacy' POPs, so-called because present-day contamination is mainly a 'legacy' of past emissions.

However, there are also newly recognised, emerging POPs, such as perfluoroalkylated substances (PFASs), a group of industrial chemicals, and polybrominated diphenyl ethers (PBDEs) (also known as brominated flame retardants), which have either been regulated more recently, are due to be regulated or are under consideration for regulation. There is a smaller evidence base for these newer pollutants than for legacy POPs, but is it is known that some (such as PFASs) tend to behave in a different way to older POPs, in that they bind mainly to protein and, to a lesser degree, fat, in humans and animals.

This Future Brief presents a selection of recent studies and analyses which highlight some of POPs' impacts on wild species and the human health effects which result from environmental contamination. It also presents data on changing levels of POPs in the environment — in air, water and animals — with some forecasts of future levels, and indications of the positive effects of regulation. Finally, it considers methods for how to substitute POPs in products and processes with safer alternatives.

The brief pays particular attention to levels and impacts of POPs in the Arctic. This is a highly vulnerable region, partly due to its delicate and unique ecosystem, but also due to the fact that POPs accumulate strongly there and in the species which live there. This is due to the ability of POPs to travel long distances on ocean and atmospheric currents, before being trapped by the cold climate. POPs thus pose a particular threat to local communities in the Arctic via the consumption of highly contaminated marine mammal species in traditional diets. The brief also gives special attention to some of the more newly identified POPs, to help build a better picture of their behaviour and effects.

POPs regulation in the EU

The EU is party to two major international agreements on POPs:

- The regional UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP), which addresses POPs through the Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Persistent Organic Pollutants (also known as the 1998 Aarhus POPs Protocol), which entered into force in 2003 and focuses on 16 substances.
- The global Stockholm Convention on POPs, which entered into force in 2004. This initially regulated 12 substances, known as the 'dirty dozen'; a further 16 have been added to the convention since 2009 and a further three are under consideration for listing.

Both instruments are implemented in the EU through the **Regulation on POPs**[1]. The Regulation complements earlier EU legislation on POPs and aligns it with Stockholm and Aarhus. Substances listed in Annex I of the EU Regulation are prohibited from manufacture or use within the EU, with some exemptions. The release of substances listed in Annex III should be reduced by Member States. See Tables 1 and 2 for substances listed under these two annexes, with examples of their main uses provided[2].

Some POPs also fall under other EU regulations; for example, Regulation (EC) No 1109/2007 and Regulation (EU) No 528/2012 contain provisions on POPs.

1. Regulation (EC) No 850/2004 of the European Parliament and of the Council of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC: http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32004R0850
2. Taken from the Stockholm Convention: http://chm.pops.int/TheConvention/ThePOPs/ListingofPOPs

TABLE 1: Complete list of Annex I substances listed under the EU POPs Regulation, subject to prohibitions in manufacture and use.

Substance	Main uses
Aldrin	Pesticide applied to soils to kill termites, grasshoppers, corn rootworm, and other insect pests.
Alkanes C10-C13, chloro (short-chain chlorinated paraffins) (SCCPs)	Used in fire retardants.
Chlordane	Used to control termites and as an insecticide on agricultural crops.
Chlordecone	Historically used as an agricultural pesticide.
DDT (1,1,1 -trichloro-2,2-bis(4-chlorophenyl) ethane)	Pesticide used to control insects that spread diseases, such as malaria, and also on crops, especially cotton.
Dieldrin	Used to control termites and textile pests.
Endosulfan	Used in agriculture to control insect pests on a broad range of crops.
Endrin	Insecticide used on the crops such as cotton and grains, also used to control rodents.
Heptabromodiphenyl ether	A main component of commercial flame retardants.
Heptachlor	Primarily used to control soil insects and termites.
Hexabromobiphenyl	Used as a flame retardant.
Hexabromocyclododecane	Used in expanded polystyrene and extruded polystyrene in buildings.
Hexabromodiphenyl ether	A main component of commercial flame retardants.
Hexachlorobenzene	Used to kill fungi that affect food crops.
Hexachlorobutadiene	Used as a solvent for other chlorine-containing compounds.
Hexachlorocyclohexanes (HCH), including lindane	Used as a broad-spectrum insecticide for seed, soil, plant and wood treatment and against parasites in both livestock and humans.
Mirex	Insecticide mainly used to control fire ants.
Pentabromodiphenyl ether	Used as an additive flame retardant.
Pentachlorobenzene	Used in PCB products, in dyestuff carriers, as a fungicide, a flame retardant and as a chemical intermediate. Also produced unintentionally during combustion, thermal and industrial processes and present as impurities in solvents or pesticides.
Perfluorooctane sulfonic acid and its derivatives (PFOS)	Used in electric and electronic parts, fire-fighting foam, photo imaging, hydraulic fluids and textiles.
Polychlorinated biphenyls (PCBs)	Range of compounds used in industry as heat exchange fluids, in electric transformers and capacitors, and as additives in paint, carbonless copy paper, and plastics.
Polychlorinated naphthalenes	Mainly used in cable insulation, wood preservation, engine oil additives and electroplating masking compounds.
Tetrabromodiphenyl ether	Used as an additive flame retardant.
Toxaphene	Insecticide used on cotton, cereal grains, fruits, nuts, and vegetables. Has also been used to control ticks and mites in livestock.

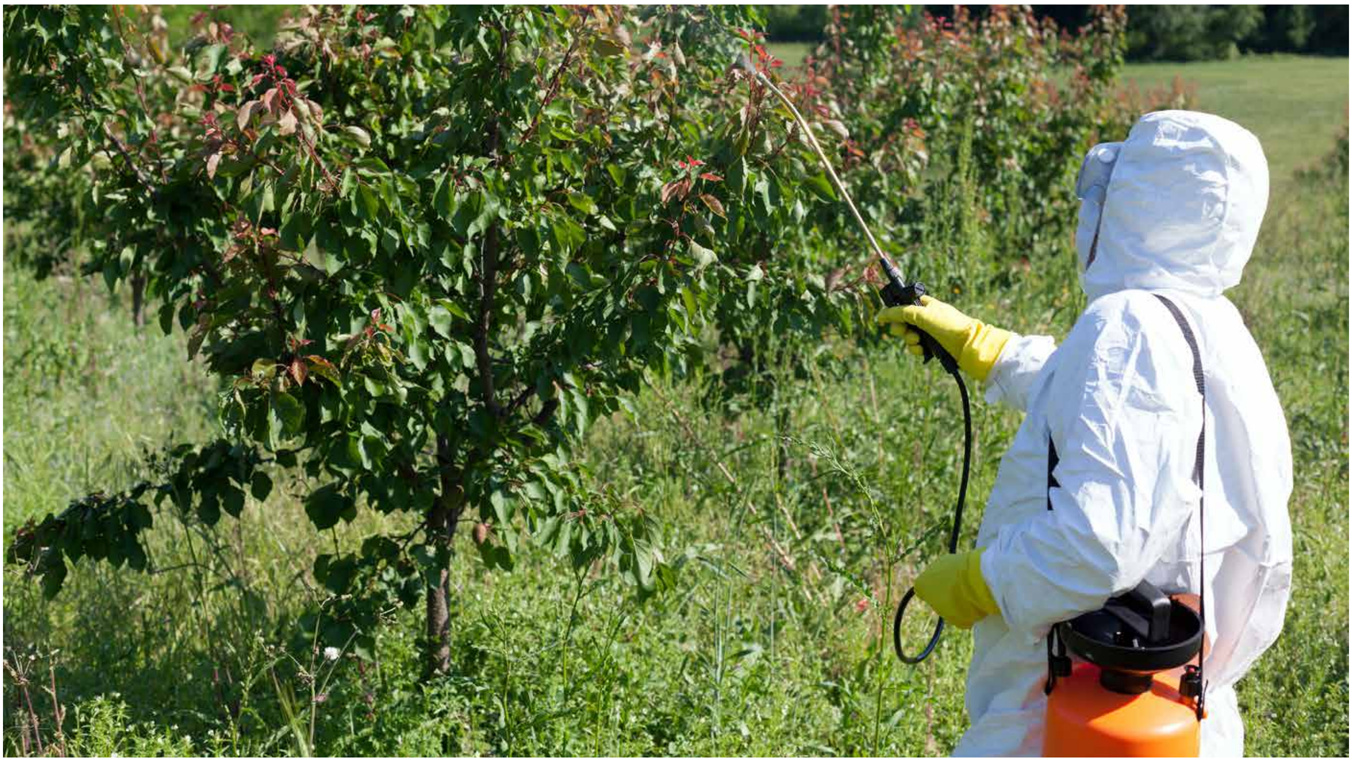
Table 1 substances are taken from Regulation (EC) No 850/2004. Example uses mainly taken from The Stockholm Convention on Persistent Organic Pollutants and the Aarhus Protocol, including 2009 amendments.

TABLE 2: Annex III substances listed under the EU POPs Regulation. These substances are subject to release-reduction provisions[3].

Substance	Main uses
Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDF)	PCDDs and PCDFs are produced unintentionally during the manufacture of pesticides, emitted through the burning of waste and in all incomplete combustion, e.g. in car engines.
Hexachlorobenzene (HCB)	Used to kill fungi that affect food crops.
Polychlorinated biphenyls (PCB)	Range of compounds used in industry as heat exchange fluids, in electric transformers and capacitors, and as additives in paint, carbonless copy paper, and plastics.
Polycyclic aromatic hydrocarbons (PAHs)[3]	Range of compounds released from the combustion of organic substances. For example, exhaust emissions, coal and industrial activities such as aluminium, iron and steel production and from burning tobacco.
Pentachlorobenzene	Used in PCB products, in dyestuff carriers, as a fungicide, a flame retardant and as a chemical intermediate e.g. for the production of quintozene. Also produced unintentionally during combustion, thermal and industrial processes and present as impurities in products such as solvents or pesticides.

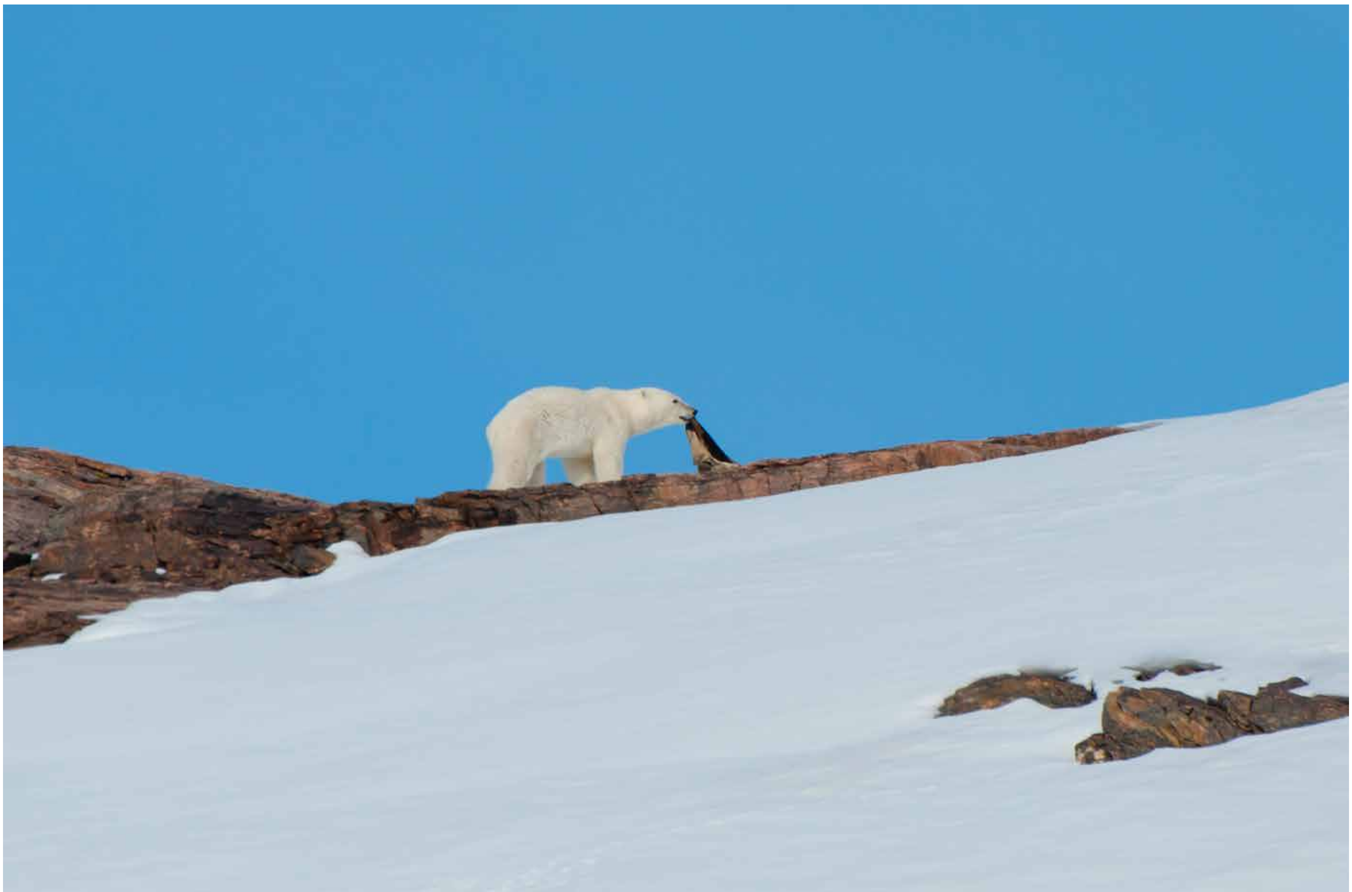
Table 2 substances are taken from Regulation (EU) No 757/2010 and Regulation (EC) No 850/2004.
Example uses taken from The Stockholm Convention on Persistent Organic Pollutants.

3. PAH details taken from Public Health England: https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/316535/benzoapyrene_BaP_polycyclic_aromatic_hydrocarbons_PAH_guidance.pdf



Pesticide spraying in a fruit orchard.

1. Ecological impacts



A polar bear, eating in Greenland.

POPs, and particularly PCBs (PCB-138, -153, -180), polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs), organochlorine pesticides (OCPs), PFOS and PFOA can be stored in fatty tissue. They are therefore often found in high levels in predatory species at the top of the food chain (via biomagnification), although they can accumulate in non-predatory species eating the predatory species.

Studies have suggested that POPs may have a wide range of effects on wild species, for example, smaller brain size and neurotoxicity, behavioural changes, hormonal disruption, carcinogenesis, cell and tissue damage and reproductive problems (Iwaniuk *et al.*, 2006; DeLeon *et al.*, 2013; Nossen *et al.*, 2016; Harmon, 2015; Da Cuña *et al.*, 2011; Barni *et al.*, 2016; Byer *et al.*, 2015).

This section focuses on two studies concerning POPS' ecological impacts on wildlife, which indicate the types of effects possible; the first considers hormonal effects of POPs on sparrows, and the second looks at bioaccumulation in tuna fish.

1.1 Endocrine disruption

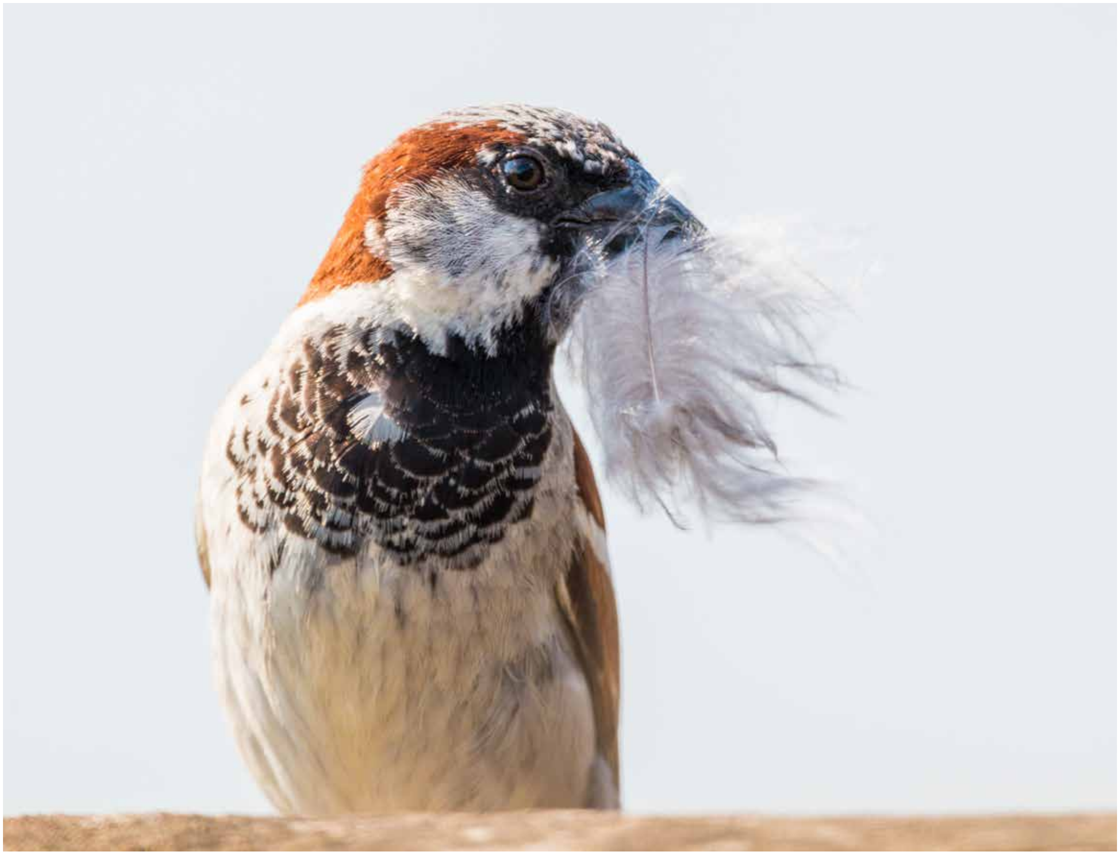
Some POPs are known to disrupt endocrine (hormonal) processes, including those that are involved in breeding and reproduction. Most research in this area has focused on species at the top of the food chain, such as polar bears (Erdmann *et al.*, 2013) and birds of prey, but new evidence indicates that these effects may also occur further down the chain. Nossen *et al.* (2016) found that POPs may affect house sparrows' (*Passer domesticus*) levels of oestrogen and testosterone, even at relatively low levels of contamination. This species is in the middle of the food chain.

The study assessed levels of several groups of POPs in sparrows and examined impacts of these chemicals on steroid hormones, which include testosterone and oestrogen. It was conducted on the island of Leka in mid-Norway. The island contains 7542 acres of farmland, the majority of which is grassland used for livestock production. Fertilisers and pesticides that contain POPs are used to cultivate the grass.

The researchers caught over 90% of the estimated 137 house sparrows on the island and analysed 22 females and 25 males with a higher 'basal metabolic rate' (BMR) for levels of toxicants and steroid hormones. Birds with a higher BMR eat more food making them more likely to have a higher intake of POPs (i.e. they are likely to be the most contaminated birds in the population).

Liver samples for each bird were analysed for seven PCBs, six PBDEs, two OCPs and hexabromocyclododecane (HBCD). Blood samples were analysed for 11 types of steroid hormones. Body mass, beak size, wing length and the size of 'badges' on the chests of males were also measured. These black badges are used by females when selecting mates for breeding. The size of the badges determines how successful a male will be with females and may be influenced by testosterone levels.

PCBs were the most common contaminant found in the birds' livers, followed by pesticides and PBDEs. However, overall concentrations of these POPs were low, and a number of contaminants (five PBDEs and HBCD) were detected in less than 60% of birds. The highest concentrations were recorded for the pesticide p,p'-DDE (0.15-5.74 nanograms per gram (ng/g) of liver) and PCB-153 (0.04 – 32.2 ng/g). Both males and females exhibited high levels of testosterone, although levels of the hormone varied greatly between individuals.



House sparrow.

Higher levels of POPs in females were associated with lower levels of oestrogen. Higher levels of POPs in males were associated with higher testosterone levels as well as larger beaks and chest badges. Those results indicate that POPs may affect steroid hormone production particularly in reducing oestrogen levels in females. Although these enlarged features may make the males more attractive to females, and thus possibly improve reproductive success, the researchers comment that high testosterone levels can also have other effects in sparrows, potentially reducing the effectiveness of their immune response, for example.

This study shows the presence of PCBs and OCPs in house sparrows in Norway in a region far away from major sources of these contaminants, confirming that PCBs and OCPs can travel long distances on ocean and atmospheric currents and persist for a long time in the environment. However, the researchers say that surrounding agriculture could potentially be the source for pesticide-derived POPs and the local use of materials, such as plastic, paintings and electrical installations, may be the source of PBDEs and PCBs.

POPs were found at lower levels in this study than in studies which have looked at predators, such as birds of prey (Luzardo *et al.*, 2014). However, even these low levels may affect hormone production, in particular by reducing oestrogen levels in female sparrows.

House sparrows are declining globally and the adverse impact of pesticides may be one of the causes, alongside other factors such as habitat loss and pollution. This was a small-scale study and the researchers recommend that future research should look at sparrows from different rural and urban populations, as well as during the breeding season, to see if their observed impacts of POPs on hormones are found on a wider scale and at different points in the reproductive cycle.

1.2 POPs accumulation in wild species

POPs can persist in the marine environment for many years, contaminating a range of seafood and thus leading to potential health concerns for humans who eat these species. One study which illustrates how POPs accumulate in wild species is Chiesa *et al.* (2016), which measured PCBs, OCPs and PBDEs in bluefin tuna (*Thunnus thynnus*) from around the world. Tuna are top predators with a wide habitat range which feed on a variety of marine species. These factors make them particularly vulnerable to contamination by POPs.

The researchers analysed tissue samples from a total of 79 wild bluefin, provided by the Fish Market of Milan, Italy, for the presence of 29 different POPs: six PCBs, seven PBDEs and 16 OCPs. The fish came from four different fishing areas; 20 fish each came from the western central Pacific Ocean, eastern central Atlantic Ocean and western Indian Ocean, and 19 fish came from the Mediterranean Sea. It is known that levels of PCBs and OCPs are beginning to decline in the environment due to previous bans in the 1970s. However, there are also emerging POPs including PBDEs, also known as brominated flame retardants (BFRs), which the EU has asked Member States to monitor in food[4].

The PCBs investigated were found in all tuna samples, apart from one (PCB-153), which was only found in samples from the Indian and Atlantic Ocean. Only five OCPs were found in the samples. Strikingly, the tuna sampled from the Mediterranean had much higher levels of PCBs compared to tuna from the other three areas; Mediterranean levels ranged from 25 to 1650 nanograms per gram (ng/g) within fatty tissue, while in the other areas concentrations ranged from 5 to 36 ng/gram.

In addition, all PBDEs investigated were detected in the Mediterranean tuna. The researchers say this is probably because the Mediterranean is semi-

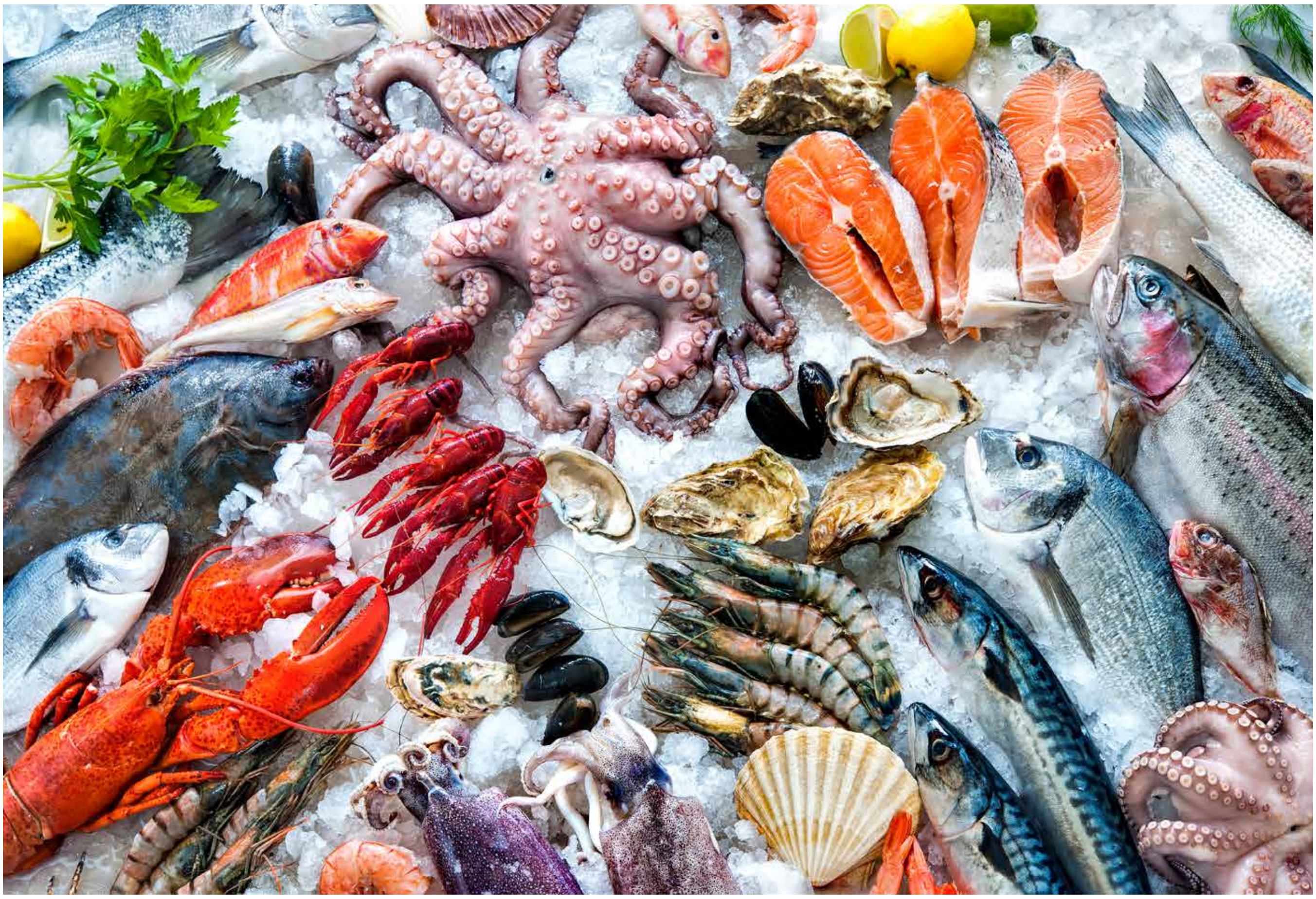
4. EU Recommendation No 118/2014 On the Monitoring of Traces of Brominated Flame Retardants in Food Text with EEA Relevance (PDF): http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2014:065:0039:0040:EN:PDF closed to the open ocean, which helps concentrate pollutants that are emitted from waste water and run-off from surrounding agriculture, industry, towns and cities. (For more information on sources of POPs to the Mediterranean, see Section 3.2).

However, the concentrations of PCBs found in this study were still below the maximum permissible levels (MPL) set by the EU[5], suggesting that the tuna sampled were legally suitable for human consumption.

The researchers say that their results demonstrate how the level of POPs present in fish reflect the pollution levels of the local environment (contamination of a top predator is an indicator of environmental contamination, due to bioaccumulation and biomagnification). The results also contribute to data on the levels of organic contaminants in seafood, particularly for PBDEs, where data are lacking. In addition, the researchers also say the method they used is a quick, cost-effective and accurate way to determine residues of POPs in fish. It could be used elsewhere to gain an overview of the presence of POPs in different regional seas and could contribute to assessing the risks to human health from contaminated seafood.

Monitoring contaminant levels in animals consumed by humans is an important part of protecting human health. For example, Carlsson *et al.* (2016) monitored POPs contamination in seafood from the Arctic waters of Norway, concluding that levels were within safe limits for human consumption. For further details, see Section 2 on human health impacts of POPs.

5. Decision No 2455/2001/EC establishing the list of priority substances in the field of water policy: http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32001D2455



Seafood on ice.

2. Health impacts OF POPs

Humans are exposed to POPs through various means: through pollution in air, water and dust, through contact with consumer products (e.g. textiles and packaging containing POPs) and also through diet, by eating POPs-contaminated food, such as certain types of wild animals and seafood which have accumulated POPs from their prey and environment. The state of the environment, therefore, has a strong role to play in the risk of POPs to human health.

Research has uncovered a number of potential links between this exposure to POPs and various health impacts, including hormone-dependent cancers, reproductive health issues, metabolic disorders (including type 2 diabetes), obesity and increased susceptibility to infectious diseases (IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2015; Bonefeld-Jorgensen *et al.*, 2011, 2014; Yu *et al.*, 2000; Ghosh *et al.*, 2014; Skakkebaek *et al.*, 2015; Weihe P *et al.*, 2016; Wielsøe *et al.*, 2017). A link between POPs and neurological effects, such as IQ effects or autism, is also an emerging concern (Stewart *et al.*, 2008; Mitchell *et al.*, 2012). It remains a major challenge to show definitive links between POP exposure and any health impacts, however, due to the long period between exposure and development of health problems and the influence of a range of other environmental factors (Antignac *et al.*, 2016).

This section presents research which appears to support a link between POPs and type 2 diabetes in particular. It also reports evidence of varying levels of POPs in humans. The complex influence of climate change on the health impacts of POPs is also briefly explored.

2.1 Type 2 diabetes

The obesity 'epidemic' alone cannot explain the huge rise in type 2 diabetes rates in recent decades. Many studies strongly suggest that POPs also contribute to this global health challenge. However, this is a complex and challenging topic to research and, consequently, studies disagree on which POPs are linked with type 2 diabetes, and there are also a few studies which have found no link at all.

However, the theory that there is a potential link between POPs and type 2 diabetes is supported by a recently published study of women in Norway (Rylander *et al.*, 2015). This study found that type 2 diabetes patients are more likely to have high levels of POPs in their blood than the rest of the population.

Although the study does not prove that POPs cause type 2 diabetes, its results add to the body of evidence produced by the majority of studies on POPs and diabetes, which suggest that POPs may be partly responsible for the onset of type 2 diabetes. It points to PCBs and OCPs, in particular, as possible culprits.

The researchers analysed blood samples taken from 108 women with the condition for three groups of POPs: PCBs, OCPs and perfluoroalkyl acids (PFAAs). They compared the samples of these patients with blood from 108 women of a similar age who did not have type 2 diabetes.

Concentrations of all the PCBs and OCPs, except PCB-180, were significantly higher in the blood of diabetes patients than in their diabetes-free counterparts. For example, mean average concentrations of the OCP beta-hexachlorocyclohexane (β-HCH) in diabetes patients was 20.3 nanograms per gram (ng/g) of lipid (fatty substances in the blood in which POPs accumulate), compared with 10 ng/g lipid in the women without diabetes.

There was no difference in PFAA lipid levels between the two groups of women, with the exception of branched PFOS, which was found at slightly higher levels in the diabetes patients (average of 8.4 ng/g lipid, compared with 7.4 ng/g lipid for the non-diabetes patients). Even when the researchers accounted for the effects of the women's weight and smoking habits on type 2 diabetes risk, they still concluded that these harmful contaminants have a significant association with the disease.

This study is an example of a cross-sectional study, in which a one-off test looks for an association between a hazard (such as a pollutant) and a health outcome in a population. However, as the researchers themselves admit, cross-sectional studies cannot show that POPs cause type 2 diabetes. Future research may be able to find clues to a causal relationship through long-term study which explores the diagnosis of disease in relation to changing exposure and levels of POPs over a lifetime. This could identify sensitive life-stages (e.g. pre-birth in the womb, or in early life, as discussed by Boekelheide *et al.*, 2012) when exposure to stressors is more closely associated with onset of the disease in later life, or certain exposure thresholds.

This study did also attempt to explore such associations by modelling the life-histories of 79 of the diabetes patients and 79 non-diabetes patients, taking account of factors which affect POPs accumulation, such as historical emissions, year of birth and diet. Unfortunately, this modelling exercise did not find any links between exposure to PCB-153 (used as an indicator for all POPs) and type 2 diabetes. This may be due to the low number of people studied as well as factors which complicate results, such as weight change in the subjects throughout their life. The researchers recommend that future studies which take a similar modelling approach should include additional POPs and explore different measures of exposure to help overcome some of the problems they faced.

One literature review of the epidemiological evidence for the association of dioxin and other persistent organic pollutants (POPs) with diabetes (De Tata, 2014) finds that exposure to the POP dioxin may cause dysfunction in pancreatic beta cells. The reviewer notes that this shows a possibility that a new, non-genomic pathway is contributing to the worldwide rising prevalence of type 2 diabetes; he indicates the need for additional animal and *in vitro* mechanistic studies to clarify the role of POPs in metabolic disease development.

2.2 POPs levels in humans

As discussed in Section 1, POPs accumulate in animals, and levels are particularly high in animals at the top of the food chain, as predators eat the contaminated tissue of other species. Concern has therefore been raised about the potential health effects of human diets which are high in contaminated species.

Dietary exposure

Long *et al.* (2015) demonstrate how diet affects POPs levels in their Greenlandic study of Inuit women in Greenland. They found that higher pollutant levels in the blood of pregnant Inuit women are related to higher consumption of marine mammals.

The levels of many POPs are falling in the Arctic. Many Arctic communities have also lowered their consumption of traditional food from marine mammals such as whales, walrus and seals, which also reduces exposure to POPs.

Maktaaq delicacy (bowhead whale blubber) by Ansgar Walk

However, certain POPs, such as perfluoroalkylated substances (PFASs) and PBDEs, are still used widely and bioaccumulate in humans and marine mammals. Regulations on PFASs have been made, with PFOS added to the Stockholm Convention and restricted in Europe[6]. The EU restricts the use of PFOA through Regulation (EU) 2017/1000, which regulates PFOA, its salts and certain related substances as a new entry (68) to Annex XVII of REACH[7]. However, although a decrease has been found in the European population, new data suggest that PFOS and PFOA are being substituted with other PFASs; for example, the use of perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), and perfluoroundecanoicacid (PFUnA) has increased since 2002 (Bjerregaard-Olesen *et al.*, 2017; Bjerregaard-Olsen *et al.*, 2016).

Long *et al.* (2015) analysed blood samples from 207 pregnant Inuit women across Greenland for 30 different 'lipophilic' POPs, which dissolve in fat, comprising 14 PCBs, 5 PBDEs and 11 OCPs. They also analysed 15 PFASs, which are also POPs but, unlike lipophilic POPs they bind to blood proteins and are stored mainly in the liver, kidneys and bile system. The researchers focused on pregnant women specifically because foetal development is the period most sensitive in life to stressors including POPs (Weihe *et al.*, 2016); and environmental exposure to pollutants during pregnancy is an important part of understanding the health of the next generation. Separate research has confirmed that unborn children can be significantly exposed to PFOS and PFOA, for example, because they can cross the placenta from the mother (ArcRisk, 2013).

The researchers also used questionnaires to assess various lifestyle characteristics, including diet and whether the women smoked. Specific questions were asked regarding how often the women consumed approximately 40 traditional Greenlandic foods and 23 imported foods. In addition, plasma fatty acids within blood were measured, as the ratio between omega 3 polyunsaturated fatty acids and omega 6 fatty acids is a good indicator of the relative consumption of marine traditional food compared to imported food.

Most of the 30 lipophilic POPs sampled were detected in blood samples. The highest concentrations recorded were for three PCBs (PCB-138, -153, -180). Pregnant women living in the east and north regions had higher levels of PCBs and OCPs compared with other regions of Greenland. For example, women in the east had a median level of 220 micrograms of PCB-153 per kilogram (µg/kg) of plasma lipid (blood fat) compared to 51 µg/kg in the west. PFASs also showed regional differences, with higher levels of PFASs found in northern and eastern Inuit women compared to women from the southern and western regions.

In the northern and eastern regions, women appeared to eat a more traditional diet, indicated by higher consumption levels of marine mammals, such as polar bear and walrus, and (particularly in the east) more local fish, such as trout and cod, and

6. Directive 2006/122/EC: http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32006L0122
7. http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32017R1000&from=EN

land-based species, such as caribou and musk oxen. The ratio of omega 3 to omega 6 fatty acids in blood samples from the north and east also indicated that the women here ate a more traditional marine diet.

To put some of this study's results into context, levels of PCB-153 and p,p′ -DDE (an OCP substance) in the women in this study (conducted 2010–2013) were higher than the levels of non-Inuit pregnant women living in Northern Arctic Norway and pregnant American women (Hansen *et al.*, 2010; Veyhe *et al.*, 2015; Woodruff, Zota & Schwartz, 2011). They had slightly higher levels of PFOS and PFOA than pregnant women from Northern Norway during 2007–2009 and Canadian non-Inuit pregnant women during 2007–2008 for PFOS (Berg *et al.*, 2014; Webster *et al.*, 2014).

However, for some POPs (such as PCB-153 and p,p′ -DDE) this study showed that levels were lower than those found in earlier research for Greenlandic Inuit in 2002–2004 and 1999–2005 (Toft *et al.*, 2010; Wojtyniak *et al.*, 2010; Bjerregaard *et al.*, 2007), which suggests that regulatory action has limited POPs in the Arctic; but the human level of lipophilic POP in Greenland is still much higher than in other parts of the globe, for example more than ten times higher than in Dansh pregnant women.

The researchers say further studies into the health effects of exposure to these chemicals in the womb is needed. They intend to follow the development of children, born to the women in this study, up until the children are 4–5 years old.

Local influences on POP levels in humans

Numerous studies have examined POPs in breast milk, demonstrating how POP levels in mothers are highly variable between individuals and populations of different countries. One such recent study was conducted by Antignac *et al.* (2016). This study compared concentrations of POPs within the breast milk of French, Danish and Finnish women, and found that levels in milk from Danish women were around 1.5 to 2 times higher than in milk from French women.

Newborn babies may be particularly vulnerable to the negative health impacts of POPs and monitoring breast milk of mothers is a good way to assess exposure of babies following birth, as levels within milk reflect the chemical exposure of the mother.

This study assessed a range of POPs (dioxins (PCDD/Fs), PCBs, PBDEs and OCPs) in breast milk samples from 96 French, 438 Danish and 22 Finnish women. The three groups of women were similar in terms of age and body mass, which are two main factors in determining bodily levels of POPs stored within fatty tissue. Breast milk samples were taken up to two months following birth.

The researchers found that French women had lower proportions of PCDD/Fs and their overall milk levels of PCBs were significantly lower compared to Danish and Finnish women. In addition, levels of PCDD/Fs and PCBs were higher in Danish women than in Finnish women. The results are expressed as the total toxic equivalency (TEQ) for each POP, which is a measure developed by the World Health Organisation to quantify the burden of toxins. For PCDD/Fs, French women had TEQ levels of 1.3 to 14.6 picograms per gram (pg/g) of lipid; Finnish women had TEQ levels from 3.5 to 17.8 pg/g; and Danish women had higher TEQ levels of 2.8-61.8 pg/g. PBDEs levels were also similar for Danish and Finnish women (median TEQ of 4.9 nanograms per gram (ng/g) and 5.2 ng/g respectively). For PCBs, French women had a median TEQ of 4.3pg/g compared to 4.6pg/g for Finnish and 6.6 pg/g for Danish women.

The researchers say the differences are likely related to differences in exposure to POPs, such as from industrial sources, which may be affected by each country's own regulations in relation to chemical pollutants. Lifestyle and dietary differences may also explain the variation, for instance, if certain populations eat fish that contain higher levels of POPs. The study points out that French and Danish women eat similar levels of seafood, for example (the French eating slightly more), but that some of the patterns of POP concentrations in Danish breast milk reflected those found in fish from the Baltic Sea. As the Danish (and Finnish) populations are more likely to eat Baltic seafood, this could be a potential, and partial, explanation (which needs further investigation) for the differences observed in POPs levels.

Lower levels of POPs detected in French women may also be partly explained by a decrease in exposure levels to POPs over the last ten years in the EU; the French samples were taken during 2011-2014, which is later than for the samples from the Danish and Finnish women (taken 1997–2002).

Some PFAA levels in decline

A decline in POPs in humans has been shown by Bjerregaard-Olesen *et al.* (2016). This research found that perfluorinatedalkyl acid (PFAA) levels in the blood of pregnant Danish women decreased between 2008 and 2013, which suggests that measures to ban or restrict the use of several PFAAs are proving to be successful.

PFAAs have been manufactured since the 1940s and their water-, oil- and dirt-repellent properties have made them useful in a wide range of industrial processes and consumer products. PFAAs are found, for example, in fire-fighting foams and cleaning products, and are used to waterproof textiles and upholstery, as well as in coatings for food containers and non-stick baking papers.

PFAAs are associated with toxicity to the immune, nervous and reproductive systems, among other health issues. People are typically exposed to PFAAs through food, drinking water, dust and air. In pregnant women, PFAAs can pass through the placenta to the foetus and babies can be exposed through breast milk.

Concerns over the health and environmental impact of these chemicals, particularly long-chain PFAAs (with six or more carbon atoms), which are especially bioaccumulative, have led to regulatory action. The Stockholm Convention has classified one PFAA, PFOS and its derivatives, as POPs and restricted their production and use in 2009. In Europe, PFOS and perfluorooctanoic acid (PFOA), another PFAA, are classed as persistent, bioaccumulative and toxic and EU-wide restrictions on the use of PFOS have been in place since 2006[8]. Restrictions for the manufacture, marketing and use of PFOA in Europe have recently been adopted. Moreover, one of the largest manufacturer of PFAAs globally phased out use of long-chain PFAAs by 2002 and seven other manufacturers agreed to similar actions by 2015.

To see how effective such measures have been, this study analysed the blood of pregnant Danish women for 16 different PFAAs. In all, 1 533 women, pregnant with their first child, who gave birth between 2008 and 2013, were randomly selected from participants in the Aarhus Birth Cohort[9] in Denmark. The blood of between 228 and 288 women was tested per year.

Of the 16 PFAAs, seven long-chain PFAAs were found in over 50% of the blood samples. These were: perfluorohexane sulfonate (PFHxS) (found in 100% of the samples), PFOS (100%), PFOA (100%), PFNA (99%), PFDA (99%), PFUnA (87%) and PFHpS (76%). Short-chain PFAAs, which have been used as substitutes for their long-chain counterparts, were either detected in less than 10% of the samples or not detected at all, although PFHpA was detected in 26% of the samples.

The researchers then examined how the blood serum concentrations of the seven PFAAs differed for each year, for every year between 2008 and 2013. They found that all seven had decreased over this time period. The average decreases were: PFHxS 7.0%, PFOS 9.3%, PFOA 9.1%, PFNA 6.2%, PFDA 6.3%, PFUnA 7.1% and PFHpS 14.8%, each year.

Compared with average blood levels of 35.3 nanograms per millilitre (ng/mL) of serum for PFOS and 5.6 ng/mL for PFOA found in pregnant Danish women from an earlier (1996–2002) period of study, the average levels of 6.4 ng/mL PFOS and 1.6 ng/mL PFOA found in 2013 in this study reveal just how much the blood levels of

8. Directive 2006/122/EC of the European Parliament and of the council of 12 December 2006
http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2006:372:0032:0034:en:PDF
9. http://www.ab-biobank.dk/ (in Danish).

these PFAAs have fallen. However, as mentioned above (Bjerregaard-Olsen *et al.*, 2016), since 1996-2002, the levels of PFNA, PFDA and PFUnA have increased in Danish pregnant women, which might indicate that some of the regulated PFASs are being substituted with other PFASs.

Other studies in Germany, Norway, Sweden, the US, Australia and Japan have also found a decreasing trend for PFOS blood levels among pregnant women and the wider population (Fei *et al.*, 2007; Schröter-Kermani *et al.*, 2013; Haug, Thomsen & Becher, 2009; Glynn *et al.*, 2012; Kato *et al.*, 2011; Toms *et al.*, 2014; Okada *et al.*, 2013). This is most likely a result of measures to ban or phase out PFOS production in these countries.

However, in China, PFOA serum levels increased dramatically between 1987 and 2002 (Jin *et al.*, 2007), suggesting that the production of PFOA, used for the manufacture of products such as Teflon, may have moved from the western world to the east, as reported by Land *et al.* (2015).

2.3 Could climate change increase the risks to health from POPs?

As POPs are able to travel long distances, they are found to collect in remote polar regions, where they are subsequently trapped by the cold climate, even though the Arctic and Antarctic are far from the sources of pollution. They reach the poles through the atmosphere, rivers and ocean currents — which are all influenced by climate-related factors, such as temperature, winds and snow and ice cover. The very cold temperatures at the poles mean that the air and water currents do not always have the kinetic energy to move the POPs out of the cold zone and closer to the equator – hence their higher concentrations in the Arctic and Antarctic. It is reasonable to suppose, therefore, that climate change will affect the transport of these pollutants to polar regions.

Even though pollutant levels are relatively low in the Arctic, the health of Arctic populations may be particularly at risk due to the region's unique food web, which includes species that store large amounts of fat for energy and warmth (where high concentrations of POPs can accumulate), such as polar bears and seals. These species also form an important part of the traditional marine local diet. As a result, some Arctic populations are more exposed to certain contaminants than people anywhere else on earth.

The EU project ArcRisk[10] explored how climate change may influence POPs, and the subsequent health effects on people living in the Arctic. The project provides a wide range of information which can be used by policymakers and built upon in future studies. Much of this information is relevant to communities all over the world. Some of the project's main results are presented by Pacyna *et al.* (2015).

For instance, the project explored how climate change will affect marine ecosystems and, therefore, sources of seafood. Together with a potential increase in contaminants, this has serious implications for Arctic communities. However, the health effects of these changes remain unclear and depend on many factors including contaminant content, consumption levels and type of seafood consumed.

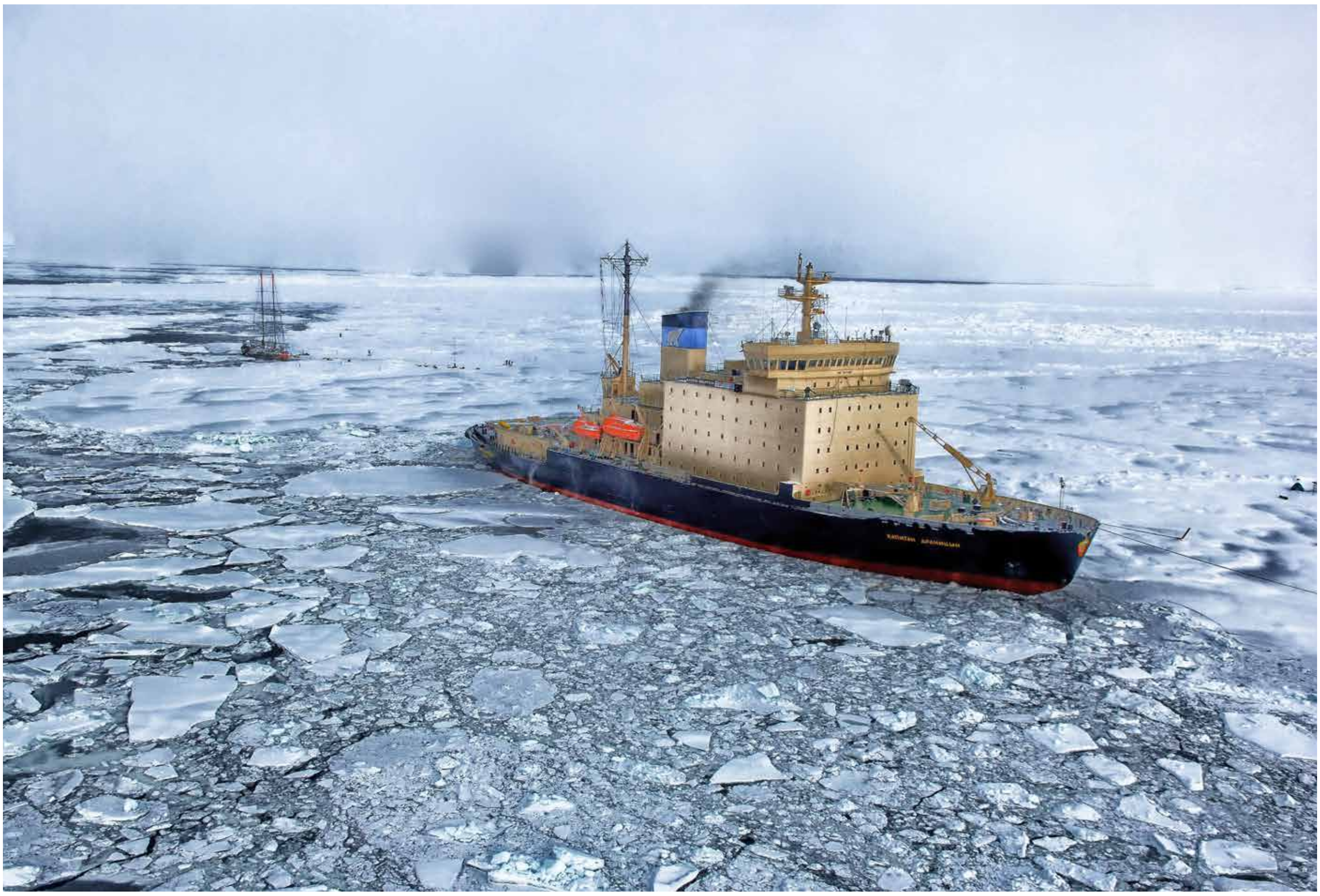
Several findings of the ArcRisk project suggest that climate change will increase emissions of POPs, which has potential health implications. For example, ArcRisk research suggests that a global temperature rise of 1 °C will lead to an increase of 10–15% in the volatility (ability to

10. ArcRisk was supported by the European Commission under the Seventh Framework Programme. www.arcrisk.eu evaporate) of semi-volatile POPs, such as PCBs and PBDEs. They are, therefore, more likely to be released from products such as paint, sealants and flame retardants. In this respect, climate change could counteract Stockholm Convention efforts to reduce POP releases.

Other increased sources of POPs under climate change include melting ice and snow and thawing soil in the Arctic, which otherwise lock away pollutants. Melting Arctic ice is also opening up new shipping routes to the north; these ships bring a new source of POP emissions to the region. Furthermore, an increase in extreme weather events, such as storms, could lead to the re-release of POPs from waste dumps and uncontrolled landfill sites. Waste management policy therefore plays an important role in controlling POPs emissions, especially as burning of POP-containing waste is also a major source of emissions.

ArcRisk also considered the beneficial effects of policies on climate change and energy. For example, replacing fossil fuels with renewable sources of energy will reduce emissions of PAHs, which are generated by burning of some fuels.

In summary, the project team highlight the complicated relationship between climate change and health, especially in the Arctic; there is evidence that a changing climate can both reduce and increase the effects of contaminants. Much more research is needed, they say. However, they have provided tools and knowledge that will help scientists conduct further research, and policymakers to mitigate risk, for example, through the management of waste or the development of renewable energy.



Arctic shipping.

3. POPs in the environment: detection and trends

Detecting and measuring POPs in the environment is essential to understanding the long-range transport and sources of these harmful contaminants. Furthermore, long-term studies which identify trends in levels can be used to help assess the effectiveness of international control measures. This section reports on a selection of recent studies concerning the detection and monitoring of POPs pollution.

3.1 The Stockholm Convention's Global Monitoring Plan

Monitoring is a particularly important element of the Stockholm Convention, which encourages Parties — including the EU — to contribute to its global monitoring activities in order to collect comparable data. Global monitoring is important for building a picture of POPs around the world, how their concentrations have changed over time, how they are moving in the environment, and — critically — how effectively the Stockholm Convention is being implemented. Magulova and Priceputu (2016) discuss the implementation of the Stockholm Convention's Global Monitoring Plan (GMP). The Plan provides a harmonised, global framework to generate and share knowledge about levels of POPs in three 'core media': ambient air, human samples (blood or milk) and water.

The GMP was established in 2007, three years after the Convention itself, as a way for Parties to comply with Article 16 of the Convention. This requires periodic review of the effectiveness of the Convention, which involves obtaining monitoring data on the environmental presence of the 23 substances listed in its annexes. Regional groups have been set up to implement the GMP in the five United Nations Regions (Africa; Asia Pacific; Central and Eastern Europe; Latin America and the Caribbean; and Western Europe and others), as well as a global coordination group that standardises these regional activities.

Several capacity-building projects and strategic partnerships have also been set up to close data gaps and improve global coverage under the GMP. In air monitoring, for example, there are three major strategic partners: the Global Atmospheric Passive Air Sampling Programme (GAPS), the MONnitoringNETwork (MONET) programme, and the South East Asian POPs Monitoring Programme.

Thanks to activities such as these, the availability and coverage of data has significantly increased since the first phase of the GMP, with long-term monitoring programmes expanding the scope of their activities to cover chemicals that are new to the Convention. A major focus of the second phase of the programme has been to provide support to regional groups for the collection, processing, storage and presentation of their data. The GMP data warehouse[11], for example, offers an online tool to store and visualise global data on concentrations of POPs and helps regional groups to produce monitoring reports.

Information is also available to the broader academic community, enabling further research. As a result of the GMP, there are now 11 years of harmonised, comparable and global monitoring data available on chemicals listed under the Stockholm Convention. Its implementation shows how a global treaty can organise and coordinate existing monitoring effects and promote the development of an international monitoring network. The activities carried out within the GMP have contributed to wider agendas on chemicals and waste.

11. Global Monitoring Plan on Persistent Organic Pollutants: http://www.pops-gmp.org/

3.2 Sources of POPs: Antarctic and Mediterranean case studies

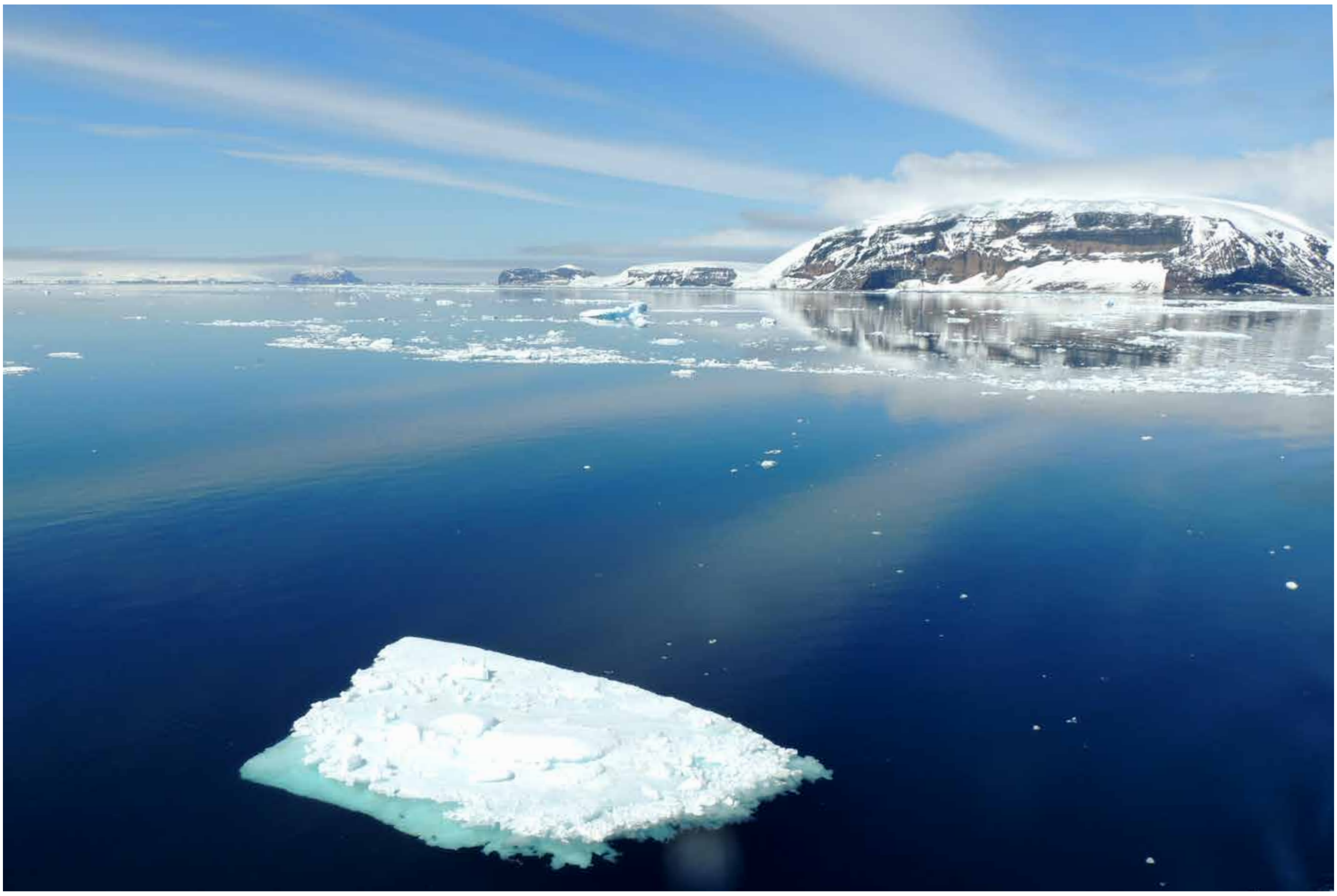
Antarctic

Various studies have analysed POPs in the environment to help build a picture of where they come from. For example, Khairy *et al*. (2016) conducted chemical analyses of POPs found in a region of the Antarctic. Their findings confirm that melting glaciers are an important secondary source of POPs, which originate from distant sources. PBDEs detected in the study seemed to have a local source, possibly a scientific research station.

Snow is able to trap large amounts of POPs released by human activity far away; long-range transport in the atmosphere is responsible for most POPs in the polar regions. Over time, melting snow and ice can re-release these pollutants to the air as secondary emissions as well as take up more pollution from the atmosphere. A better understanding of how POPs exchange between snow and air is needed to better understand their fate in polar regions. Many studies have researched POPs in the Arctic, but Khairy *et al.* (2016) is one of very few to focus on the Antarctic.

The research team explored: 29 types of PCB, 27 OCPs, including DDT, and 12 types of PBDE. They first analysed 12 samples of air and seven samples of snow for concentrations of these pollutants. The samples were taken from various locations near a US research station on the western Antarctic Peninsula in 2010.

Results show that concentrations of POPs were highest in samples of melted snow from a glacier. For instance, hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCBs), and DDTs (all OCPs, see Table 1 for more detail on HCBs and DDTs), PCBs and PBDEs were 1.4 to 23 times greater than at the other locations. This confirmed the researchers'



Navigating the Antarctic.

suspicions (based on earlier studies) that glaciers are an important secondary source of persistent pollutants in the western Antarctic Peninsula.

Of the OCPs, DDTs had the highest snowmelt concentrations of 30 to 412 picograms per (pg/L), depending on location (a picogram is one trillionth of a gram). Snowmelt concentrations of PCBs ranged between 300 pg/L and 770 pg/L, while PBDE concentrations were 20-161 pg/L.

For the atmospheric samples, HCBz had significantly higher levels than all other OCPs at 34 picograms per cubic metre of air (pg/m^3) on average. It was also the most abundant of all the POPs. All PCBs combined had an average atmospheric concentration of 12 pg/m^3. PBDEs showed an average concentration of 4.2 pg/m^3.

Chemical analyses suggested that the PBDEs had a local source, as they showed little sign of degradation by light, which would normally be expected of chemicals from long-distance sources. The researchers, therefore, believe that the research station itself could be the main source of local PBDEs.

The snow and air appeared to be influenced by different sources. For instance, the researchers suggest that OCPs found in the snow but not in the air are the result of old contamination, as the detected OCPs were banned more than 30 years ago. In addition, calculations of exchanges of POPs between snow and air suggested that PBDEs and two types of OCP — chlordanes and alpha-endosulfan — are mainly deposited onto snow from air. All the PCBs, HCBz, heptachlor (an OCP) and DDTs, on the other hand, appeared to have been mainly re-released by snow as secondary emissions to the air. The researchers hope their data can help scientists and policymakers to develop a better picture of the relative importance of long-range vs. local primary and secondary sources of POP emissions in the Antarctic.

Mediterranean Sea

In a separate piece of research, Brumovský *et al.* (2016) considered sources of PFASs in the western Mediterranean Sea. Their analysis suggests that the main sources are probably the Atlantic Ocean and rivers including the Rhône and Ebro.

PFASs are manmade chemicals that have been used for over 60 years for a range of purposes, including as emulsifiers, lubricants, components of fire-fighting foams and stain repellents. However, they have become widespread pollutants. Several PFASs are restricted by law; most uses of PFOS, for example, are banned in the EU under Regulation (EC) No 850/2004 as it is a POP. PFOS is also listed under the Stockholm Convention, where it was recently recommended that another toxic PFAS, PFOA, also be listed[12].

This study measured PFAS concentrations in the open Mediterranean Sea for the first time. In 2014, the research team analysed samples of seawater taken from 16 locations in the centre of the western basin. All water was taken from the surface, apart from one sample which was taken from a depth of 1390 m near Sardinia. The researchers' overall aim was to understand more about how PFASs enter the Mediterranean.

Fifteen PFASs were detected altogether. In combination, concentrations ranged between 246 and 515 picograms per litre of water (pg/L) in surface water samples, with an average concentration of 357 pg/L. The most abundant PFASs included PFOA and PFOS, with concentrations of 19–86 pg/L and 18–60 pg/L, respectively. The other most abundant substances were PFHxA, PFHpA and PFHxS, with respective concentrations of 20–189 pg/L, 21–131 pg/L and 5–41 pg/L.

Total PFAS concentrations in the deep-water sample were 141 pg/L, which is a relatively small difference from the average surface concentrations. The researchers suggest that PFASs reached these lower

12. *http://chm.pops.int/TheConvention/POPsReviewCommittee/POPRCRecommendations/tabid/243/ctl/Download/mid/14049/Default.aspx?id=5&ObjID=20843*

Le Rhône entre Viviers et Donzère (Ardèche/Drôme, France).

depths as a result of downwards movement of water over time, as part of local circulation patterns.

The researchers ruled out the atmosphere as a likely major source of these chemicals in the Sea. A high PFHpA/PFOA ratio in the water can indicate that rainwater has deposited large amounts from the atmosphere. However, the ratios were low in this study (with the exception of one sample which was collected during a period of heavy rainfall). The researchers estimate that the atmosphere is responsible for depositing 0.8 tonnes of PFOA and 0.4 tonnes of PFOS to the western Mediterranean every year, based on average rainwater concentrations of PFASs.

Instead of the atmosphere, the researchers suggest that the main sources to the Sea are the Atlantic Ocean and discharges from rivers in the western Mediterranean. Nearly all water entering the western Mediterranean comes from the Atlantic, and PFAS concentrations in this study were similar to concentrations found in the Atlantic near the Strait of Gibraltar, where it enters the Mediterranean Sea. The total amount of water from rivers entering the Sea represents just 0.5% of Atlantic inflow. However, some of these rivers, especially the Rhône, which flows through Switzerland and France, and the Ebro, which flows through Spain, are highly contaminated because they drain some of the most populated and industrialised regions in Europe.

Previous research in this region has only considered a limited number of PFASs in the Western Mediterranean Sea and only included waters up to 100m from the coast. However, the results of these studies are broadly in line with the results of Brumovský *et al.* (2016). For example, the levels of PFBS, PFHxS, PFOS, PFOA and PFNA investigated by Sánchez-Avila, Meyer and Lacorte (2010) ranged generally from several tens of pg/L (i.e. similar to Brumovský *et al.* (2016)) to concentrations that were 1-2 orders of magnitude higher in areas close to major estuaries. These earlier results, therefore, also suggest that rivers are an important source of PFASs to the Western Mediterranean Sea. PFASs observed by Brumovský *et al.* (2016) in Atlantic water that enters the Mediterranean through the Gibraltar strait are similar to previous data from the Atlantic Ocean measured between 2007 and 2010 in proximity to the Gibraltar strait and the southwest of Portugal (Ahrens *et al.*, 2009; Ahrens, Xie & Ebinghaus, 2010; Benskin *et al.*, 2012; González-Gaya *et al.*, 2014).

3.3 POPs in decline: trends in atmospheric POPs

Two recent studies are presented here which demonstrate a decline in atmospheric levels of most POPs in northern regions. However, there is evidence in some locations that levels are increasing for some substances.

The Arctic

The Arctic environment is traditionally considered a pristine area, but investigations in the 1980s revealed unexpectedly high levels of POPs in the region. Soon after, the Arctic Monitoring and Assessment Programme (AMAP)[13] was set up to monitor a number of priority 'pollution issues of concern'. AMAP provides air monitoring data that can be used to assess trends and connect individual, national monitoring efforts with the Stockholm Convention's Global Monitoring Plan (see Section 3.1 for more information on the Global Monitoring Plan (GMP)). Data collected under AMAP on the persistence and long-range transport of chemicals were important for the negotiations leading to the Aarhus Protocol on POPs and, shortly after, the Stockholm Convention. Since then, AMAP has played an important role in the continued development of the Convention.

Hung *et al.* (2016) reviewed the results of AMAP and describe declines for most POPs. Their study discusses the measurements taken at the four longest-running stations under AMAP: Alert, Canada; Stórhöfði, Iceland; Zeppelin, Svalbard, Norway; and Pallas, Finland. Air samples (one of the core media under the GMP) have been collected at these four stations using high-volume air samplers. Although the frequency of sampling differs between sites, all stations follow strict quality-assurance protocols to ensure that data are comparable.

Air monitoring for legacy POPs, including OCPs and PCBs, began at the four AMAP stations in the 1990s, and provides some of the longest time trends of POPs in the world. There are also several datasets that pre-date the Stockholm Convention, such as those for endosulfan (a toxic and endocrine-disrupting insecticide) and PBDEs, which contributed to an essential risk assessment that led to the decision to classify them as POPs.

The results of AMAP monitoring show that most Stockholm Convention POPs, including PCBs, DDT and chlordane (an organochlorine pesticide), are declining in the Arctic due to reduced primary emissions. However, reductions have been slow due to the persistence and slow degradation of these chemicals. Indeed, several of these chemicals can be still detected in countries where they have been banned for decades.

More surprisingly, some chemicals have been shown to increase in certain locations, such as lighter PCBs (52 and 101) in Iceland. The researchers suggest this may be due to continued emissions in some areas and to climate warming. At Stórhöfði, a coastal site close to the Icelandic ice caps, melting could cause the re-emission of previously deposited PCBs from oceans and ice.

13. Arctic Monitoring and Assessment Programme: http://www.amap.no/

Another important element of AMAP is its ongoing assessment of emerging contaminants. To nominate a chemical to be added to the Stockholm Convention, risk profiles must be analysed to determine *"whether the chemical is likely, as a result of its long-range environmental transport, to lead to significant adverse human health and/or environmental effects, such that global action is warranted."* For future risk assessments, the researchers recommend increasing the coverage of air-sampling sites (including remotely and within communities) combined with surveillance measurements above northern oceans, which are less influenced by local sources. They say this would enable assessment of the relative importance of long-range transport and local emissions for emerging compounds and ultimately Stockholm Convention classification.

Finland and Sweden

Another recently published study that shows a fall in levels of atmospheric POPs was conducted by Anttila *et al.* (2016). The research from Finland and Sweden indicates that international bans on POPs have successfully reduced concentrations of these damaging substances in the Scandinavian atmosphere, including a part of the Arctic. It also predicts when different POPs will eventually disappear from the air in this region.

This study monitored 27 different POPs between 1994 and 2011 at two air-quality monitoring stations to understand the status of POPs in the Scandinavian atmosphere. The first station, Pallas, is in a remote part of northern Finland, 170 km north of the Arctic Circle. The second, Råö, is in a rural part of southern Sweden, 1000 km further south of Pallas.

Råö's more southern, less remote location means it is more exposed to ongoing primary emissions than Pallas. Pallas is potentially more influenced by re-mobilised secondary emissions from reservoirs of POPs that have accumulated in the Arctic. These stores of POPs have gathered through the 'grasshopper effect', whereby POPs — particularly those that are more volatile — are repeatedly exchanged between the atmosphere and the Earth's surface due to varying temperatures, moving gradually towards colder regions. Once they reach cold regions, such as the Arctic, low temperatures lead to POPs being deposited and trapped.

The monitoring data showed that atmospheric POP levels were generally higher in Råö than in Pallas, which can be explained by Råö's proximity to sources of primary emissions. For instance, PAH concentrations were typically 3-9 times higher at Råö than Pallas. However, levels of low-chlorinated PCBs and chlordanes were around the same at the two sites, and alpha-hexachlorocyclohexane (α-HCH) was higher in Pallas. This is because these substances are more volatile and thus more prone to the grasshopper effect. In addition, α-HCH is particularly persistent in cold, northern waters, such as those near Pallas.

Overall, atmospheric concentrations of most POPs declined over the study period. PCBs fell by 2–4% per year, HCHs by 6–7%, chlordanes by 3–4% and DDTs by 2–5%. This suggests that legislation has successfully reduced atmospheric concentrations of POPs over Scandinavia, the study's authors say. These results also reflect findings from other studies; declining PCB trends have also been seen in research from other Arctic and European areas, as well as North America (Hung *et al.*, 2016; Kong *et al.*, 2014). However, most PAH substances did not show any overall decline, which can be largely explained by their ongoing primary emission through combustion.

Based on these figures, the researchers predicted how long it would take for the different POPs to disappear from the Scandinavian atmosphere. For example, higher chlorinated PCBs will be depleted by the 2020s (with PCB-180 being eliminated first, by the year 2020), although low-chlorinated PCBs will not be depleted until the 2030s at the earliest, they predict. HCHs and the most degradable chlordane, trans-chlordane, will be depleted by around 2020, and cis-chlordane and trans-nonachlor should be eliminated a few years later, during the 2020s. Chlordanes will be depleted from the southern Scandinavian atmosphere first, given that they move northwards over time.

It is expected that DDT will be removed from the atmosphere in the late 2010s at Råö. However, the most long-lived compounds, such as p,p'-DDE (a breakdown product of DDT), may persist in the atmosphere at Råö until the early 2030s, the researchers suggest; and they will not be removed from the atmosphere at Pallas until ten years later.

The monitoring of POPs by this study confirms that the atmosphere is an important route for these contaminants into the Nordic environment, even in more remote areas. It also confirms that there are variations in behaviour between substances, which differ in the south and north of Scandinavia.

3.4 Detecting POPs in Arctic seafood

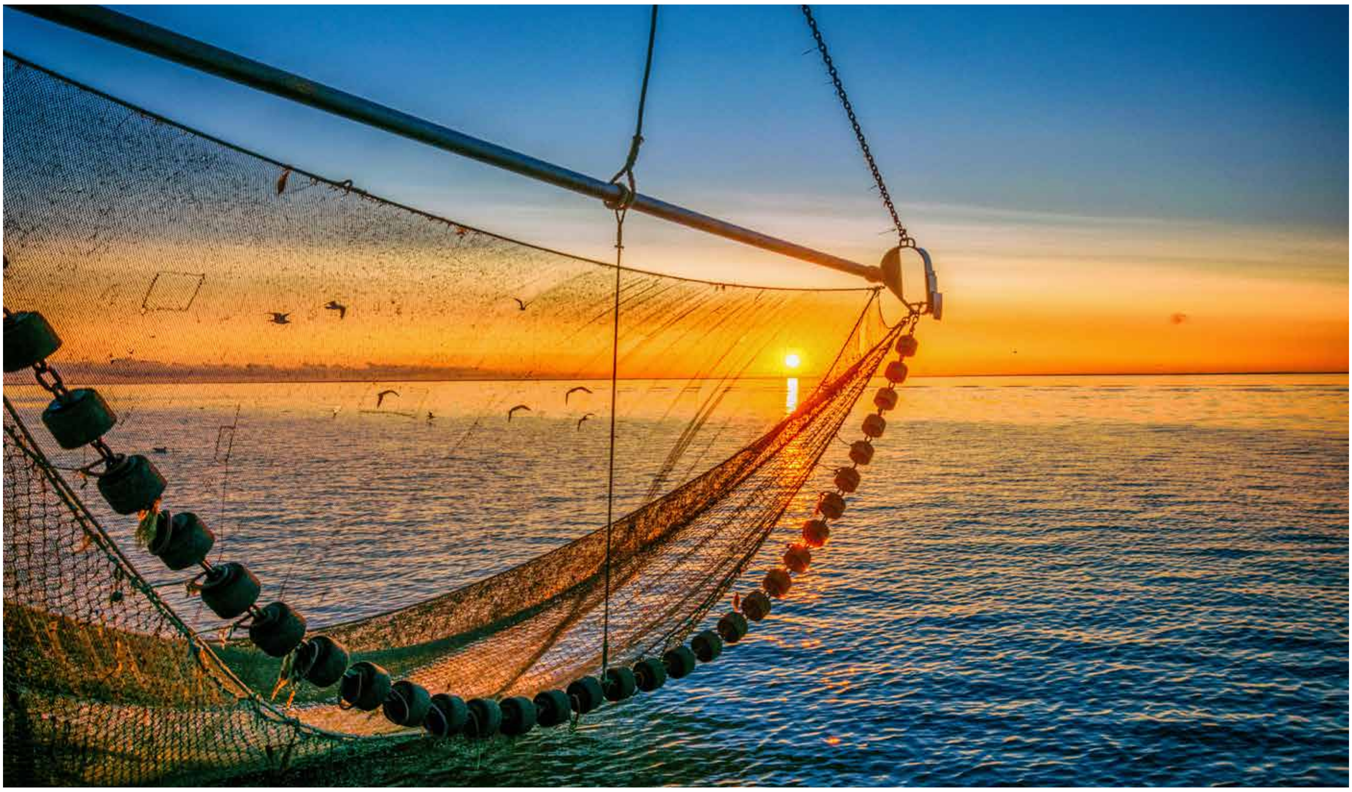
As discussed, POPs can contaminate wild species which may carry potential risks for human health if these species are consumed as food. Monitoring POPs contamination levels in such species is therefore an important public health endeavour. One study to carry out such an assessment is Carlsson *et al.* (2016), which measured POPs in two commercially important marine species in Arctic Norway — halibut (*Hippoglossus hippoglossus*) and shrimp (*Pandalus borealis*). The results show that levels in these species comply with EU guidelines, but that exposure risk varies between legacy and emerging contaminants, which should be reflected in future risk assessments. Furthermore, although contamination levels were judged to be safe, it is also important to note that these two species are clearly not the only sources of POPs exposure for humans, and the risk of cumulative exposure from all sources needs to be considered in future research.

This study, which was conducted as part of the Arctic Monitoring and Assessment Programme (AMAP) (see Section 3.3), investigated the levels of contaminants within halibut and shrimp in the coastal waters of Arctic northern Norway. These are popular and commercially important food products in Norway, where around 5000 tonnes of coastal shrimps and 1400 tonnes of halibut are caught each year. Although marine food products are regularly scanned for pollutants in Norway, very few surveys have looked at the levels of POPs in these species. Source of POPs in the region may include local activities, such as shipping, as well as long-range transport from other parts of the globe.

For the study, the researchers purchased halibut fillets from local fishermen that were caught from coastal waters between 2008 and 2012. Shrimps were provided by a local supplier in 2012. The samples were analysed for a range of PCBs, PBDEs and OCPs. These are legacy POPs that have been around for many years and are strictly regulated. The researchers also looked at levels of PFASs, a group of emerging POPs that have been more recently produced and regulated.

Encouragingly, contaminant levels were all within tolerable levels. For PCBs, median levels in halibut were 4.9 nanograms per gram (ng/g) of fish and 2.5 ng/g in shrimp, which are both well below the EU guideline[14] of 75 ng/g. Levels of PFOS (the most abundant PFAS) — 0.9 ng/g for halibut and 2.7 ng/g for shrimp — were also well below EU guidelines for safe daily intake.

14. Commission Regulation (EU) No 1259/2011 of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32011R1259



North Sea shrimp net.

The researchers say it is unlikely that consumption of other, non-seafood products would increase PFAS exposure enough to exceed the EU guidelines. However, they say indoor air and consumer products are also sources of PFASs and recommend including exposure from air, dust, food packaging and cooking devices in future risk assessments.

There are currently no tolerable daily intake levels for PBDEs in the EU, but concentrations were far below the benchmark doses (estimated safe levels). According to the European Food Safety Authority, only BDE-99 could be of potential health concern, and this was present at very low levels in the samples.

The results also showed important differences between the contaminant composition of the two samples. The halibut fillets mainly contained PCBs, which contributed half of their overall POPs load, while shrimps, which have a higher protein content than halibut, mainly contained PFASs (74%). While older, 'legacy' POPs, such as PCBs, are associated with organisms at high levels in the food chain (such as large and old fish), this is not necessarily the case for emerging contaminants like PFASs. The researchers say this reflects the different pathways of contamination for newer contaminants, which tend to accumulate in protein, compared to older POPs, which bind to fat. They therefore recommend that future discussions of PFAS levels consider the protein content of food items, in a similar way to how discussions about legacy POPs consider fat content.

Finally, the results suggest that local sources are not contributing significantly to the contaminant burden, as concentrations of legacy POPs and PFAS were similar to other studies in the Arctic. This study provides important insight into contaminant levels in coastal fisheries and the uptake of POPs in two commercially important marine species. The results suggest that consumption of these organisms do not pose a significant risk to human health, and provide data that could improve human exposure assessments for POPs.

4. Reducing and replacing POPs

Strategies are needed to reduce our use of POPs — strategies which identify safer alternatives to these contaminants. This section presents two analyses of how harmful substances can be substituted; these approaches could be applied to POPs as well as other toxic chemicals.

4.1 Functional substitution

In an analysis of sustainable chemicals management, chemical-safety experts argue that chemists and policymakers should focus on a chemical's function, and not just its risks, to identify safe alternatives to toxic substances (Tickner *et al.*, 2015). This 'functional substitution' approach could lead to a range of alternative solutions to the use of toxic substances, which may not be chemicals, but different and more sustainable methods of achieving the same goal.

Traditional approaches to chemicals management gather information on chemical hazards, uses, exposure and risks, and develop risk-management measures, such as restrictions on use or other ways of reducing exposure. These approaches provide an important basis for decision-making and can lead to the production of safer chemicals. However, these measures are time-consuming and can also mean that a broader range of solutions, such as alternatives to chemical use, are not considered.

The functional substitution approach switches the main focus from risk to function in order to identify safe, alternative solutions. For instance, this approach can identify different options for achieving flame-retardant properties — which may include chemical as well as non-chemical solutions — rather than managing the risks for a single flame retardant, or similar chemicals. Tickner *et al.* (2015) define 'function', in this context, in three ways. Each needs to be considered simultaneously.

Chemical function. Chemists design chemicals to have specific properties that will help them to perform their intended functions (e.g. release colour, in the case of dyes). This focus allows synthetic chemists to find chemical replacements to toxic chemicals, which have properties that enable the same function but without toxic side effects.

End-use function. This relates to the specific purpose of a chemical and allows alternative materials or design processes to be identified. For example, if the aim is to protect food, instead of using PVC film that contains phthalates (which may have adverse health effects), a different material that does not contain such plasticiser additives could be used, such as low-density polyethylene.

Function as service. This relates to the broad service that a chemical provides in a material, product or process, such as microbial resistance or flame retardancy. The necessity that the product provides this service, or even the necessity of the service itself, can be questioned. For example, anti-microbial substances in handsoap may be regarded as unnecessary as plain soap and water provide the same service.

The researchers illustrate the approach with specific examples, including methylene chloride, a solvent used to degrease metal parts and which is restricted in the US as it is a suspected carcinogen. To replace

Chemical hazard pictograms.

it, one could take the conventional approach and consider a chemical replacement (i.e. look at the chemical's function), such as a chlorinated solvent degreaser. However, this risks introducing other toxic elements. Alternatively, one could consider the end-use function, degreasing, and other means of achieving this. A redesign of the process, such as introducing water-based cleaning, could work here. Finally, when considering the service, one could question whether degreasing is necessary at all — and identify different means of achieving the same purpose (providing clean metal parts), such as different metal-cutting methods which do not require degreasing.

The functional substitution approach, therefore, provides a wider lens for finding safe alternatives, the researchers say. It leads to a greater range of options for replacing hazardous chemicals and reduces the risk of 'regrettable situations', in which one toxic chemical is simply substituted with another.

4.2 Alternative assessment frameworks

Various alternative assessment frameworks have been developed to help ensure that substitutes are not more dangerous than the original substance, from both environmental and human health perspectives. These frameworks identify, compare and eventually select safer alternatives.

A recent study (Jacobs *et al.*, 2015) compared 20 of these frameworks to identify areas for improvement. It concludes that the frameworks need to be more consistent and should be refined. For example, more attention should to be paid to understanding the environmental risks of substitute chemicals.

The researchers focused on frameworks with a multi-step process for evaluating chemicals and alternatives. They evaluated them according to six components.

Hazard assessment

The most common types of hazardous property (or 'end points') assessed by the frameworks were physicochemical properties (e.g. flammability, explosiveness), human toxicity, environmental toxicity and additional workplace hazards. The most commonly assessed environmental properties were aquatic toxicity, persistence and bioaccumulation, which are covered by 13 of the frameworks. The researchers say that a much wider range of ecotoxicity end points need to be included in frameworks, not just aquatic ecotoxicity. Only one framework, the National Academy of Sciences (NAS) framework[15], considered terrestrial ecotoxicity (to plants and animals).

Technical assessment

Technical assessment considers whether a substitute can perform the same function (e.g. solvency) as effectively as the original. It may also look at whether it is feasible from legal, labour and supply-chain perspectives. The majority of the studied frameworks lack specific guidance on how to assess technical feasibility. The frameworks that provide more detailed guidance draw information from stakeholders, scientific studies and pilot tests, among other sources.

Economic assessment

Frameworks judge economic viability using general categories, such as commercial availability, direct costs (e.g. for manufacturing), indirect costs (e.g. emissions controls for regulatory compliance) and long-term costs (e.g. long-term profitability). However, consistency on which costs are included is lacking between frameworks.

Exposure characterisation

Eighteen of the 20 frameworks evaluate exposure (although in different ways) — whether to workers, the public or the environment. The majority evaluate exposure using indirect measures, such as a chemical's potential to disperse, rather than actual exposure data. The researchers therefore suggest that methods for assessing exposure should be refined, for example, to enable rapid categorisation of different types of exposure.

Life-cycle analysis and thinking

These elements aim to avoid choosing substitutes that simply shift risk from one stage of a product's lifecycle to

15. National Academy of Sciences Framework: https://www.nap.edu/catalog/18872/a-framework-to-guide-selection-of-chemical-alternatives another. Life-cycle analysis is more thorough than life-cycle thinking, as it has a well-defined method, rather than a more general consideration of impacts at different life stages. However, it is expensive and time-consuming and so it is included by only four of the frameworks assessed. Other limitations to this component include a lack of data on chemical toxicity and chemical release.

Decision-making

Frameworks that include decision-making help identify trade-offs between substitutes, helping to ensure, for example, that a chemical with a health risk is not replaced by a chemical with an environmental risk. Thirteen of the frameworks consider decision-making, although they have different approaches for doing so and many provide very little guidance. It is not yet clear what the 'best' decision-making process is, or how the frameworks lead to different choices.

Two of the most comprehensive frameworks assessed in this study, in terms of levels of assessment and complexity, include the European Commission's framework, Minimising chemical risk to workers' health and safety through substitution, from the Directorate General for Employment, Social Affairs and Inclusion[16], and the USA Interstate Chemicals Clearinghouse's framework[17]. Overall, however, the researchers say frameworks should be more consistent with each other, and recommend more scientific collaboration across fields to refine substitution methods.

5. Summary

A number of issues arise from the research on POPs' complexities, as illustrated in this Future Brief. For example, studies presented here have implications for ecological risk assessment of POPs. Research indicates that we may need to move away from a pure focus on a species' positioning in the food chain as an indicator of contamination levels and of POPs' effects. It is not necessarily only species at the top of food chains — or the most contaminated species — that are most affected. For some newer POPs, the protein content of species, rather than their fat content, or position in a food chain, may be a better indicator of contamination levels.

Research also confirms that POPs are clearly still in the environment, animals and people, and that they are having damaging effects; reproductive effects (on mother, foetus and child), endocrine disruption and type 2 diabetes are just some examples of possible effects explored in this report. However, existing regulations to curb emissions have been successful, monitoring studies suggest. These show encouraging declines in the atmosphere and in humans. However, research also issues a warning: climate change could undo some of these positive impacts of regulatory work by increasing emissions of some POPs. Impacts, emissions and mitigation measures need to be carefully researched and considered for more newly identified POPs, for which there are fewer data.

To reduce our usage of POPs we need practical ways of finding substitutes. These alternatives may well be other chemicals, but we must also take care not to replace one toxic problem with another, as the analyses above argue. A broad range of non-chemical alternatives should also be considered, as well as considering the methods for identifying alternatives themselves.

While the studies presented provide many reasons to be vigilant about POPs, they also offer clear hope that we can reduce and even eliminate many of these harmful substances in the environment through concerted research, regulatory and implementation efforts.

16. https://osha.europa.eu/en/legislation/guidelines/minimising-chemical-risk-to-workers2019-health-and-safety-through-substitution
17. www.theic2.org/aa_library

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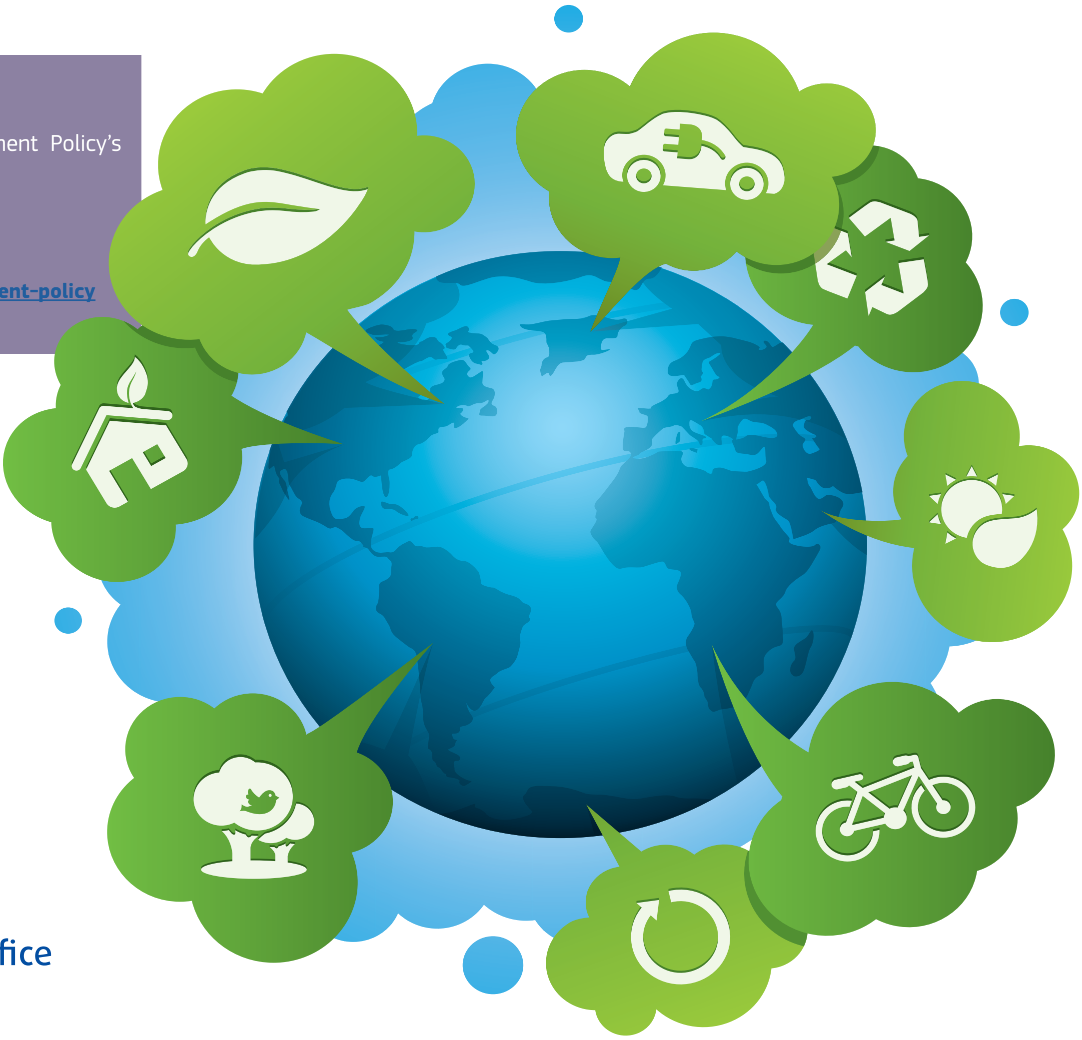

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