Persistent Organic Pollutants (POPs) and related chemicals in the global environment: some personal reflections

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What are POPs and why are they interesting and important?

In this article, I have been given the chance to 'tell the story' of POPs, through personal reflections on how the field has developed. I hope I can do that by conveying some of the sense of excitement, interest and importance for the environment and society that motivated me as a young research scientist and reflect on some of the ongoing needs and priorities for the environmental chemistry and chemicals management fields.

Persistent organic pollutants (POPs) and related chemicals are fascinating, because of their combination of physical-chemical properties and complex effects.[1] Most are man-made, but some also have natural origins. They are persistent in the environment - but can be broken down - variously by biodegradation, atmospheric reactions, and abiotic transformations. They can exist in the gas or particle phases – or both - in the atmosphere, and in the dissolved or particulate phases – or both - in water. These combinations mean that they may undergo long-range transport in the atmosphere or oceans, or they may stay close to sources. Hence, emissions from one country are frequently a source of contamination to another country. They are also usually lipophilic, so – combined with persistence – this means they can accumulate in organisms and bio-magnify through food chains. We all have a baseline of POPs residues in our tissues – even the unborn foetus via placental transfer and the newly born baby via mother's milk. POPs in biological systems occur in mixtures, so confirming effects caused by POPs on humans and other top predators is never straightforward. Depending on which papers you read, POPs may be relatively benign, or they could be responsible for key sub-chronic and chronic effects – on reproductive potential, on immune response, as carcinogens and on a range of behavioural and cognitive endpoints.[2-5] They could be a factor behind diseases and conditions which have been increasingly reported and studied in modern societies. In short, they are endlessly fascinating to scientists and a nightmare to regulators and policy makers.

Just now, I used the term 'tell the story of POPs'. To me, this really is a classic story of environmental science - a story of pollutants on a global scale, requiring multi-disciplinary teams to unravel their behaviour, with a powerful narrative for the public, industry and policy makers of 'unexpected consequences'. Where do they come from? Where will they go? How long will they be on the planet? What are they doing? There is also an unfinished and evolving story about whether we can prevent more and new POPs entering our planetary system. It is a great case study for school kids and undergraduates alike. It is also very real for many people in the world, who might be impacted by living close to manufacturing facilities, combustion sources, or waste dumps, or who have elevated dietary exposures, for example, or are worried about their baseline exposure from everyday products around them.

How the story started

For me, the POPs story began with the publication of Rachel Carson's classic book, *Silent Spring* in 1962.[6] As a child, I remember my parents - who were fascinated by the natural world - telling me about the book. The emotive issues raised by *Silent Spring* shaped me as I decided to study Environmental Science at London University in the late 1970s (a subject not taken so seriously at that time), rather than a more conventional subject. Manufacture of chemicals had been increasing dramatically through the 1930-60s. Before *Silent Spring*, I suspect the general view in a rapidly developing world was that the use of newly designed and manufactured chemicals – agrochemicals, industrial chemicals, chemicals of commerce – was a 'good thing' that was bringing a brave new world of greater food security, pest and disease control, improved standards of living and hygiene. Of course, we have seen many such benefits from the chemicals industry, but – with the publication of Carson's book and the ensuing discussions – society started to become aware that there are always trade-offs and risks.

New chemicals cannot always be regarded as ‘good’. As we know now, the case for chemical use/restrictions is usually not black and white.[7]

Two key issues also came together to shape the research and evaluation of POPs in the 1960/70s. One was the development of incredibly sensitive analytical methods and the other was the establishment of biological monitoring schemes/archives. Nowadays it is often taken for granted that we have access to sensitive, sophisticated and expensive instrumentation; we talk blithely about detecting parts-per-billion, even parts-per-trillion, and below in the environment. We can routinely measure chemicals that nobody knew were present in the environment in the past. However, we should not be motivated simply by making measurements. We need to give context, to justify *why* we are studying trace quantities of chemicals in the world around us, the significance (or otherwise) of levels detected and what they may be doing to the environment, ecosystems and humans.

James Lovelock is perhaps best known as the author of the Gaia hypothesis, which argues that the Earth can be viewed as a self-regulating ‘organism’ or system.[8] However, he also invented the electron capture detector (ECD) in the late 1950s, and by coupling it to gas chromatography was able to detect trace amounts of halogenated chemicals.[9] GC-ECD provided the ideal tool to measure chlorinated organic molecules with high sensitivity and selectivity at that time. Prior to this – there was little or no awareness that POPs were spreading around the planet and through food chains; we are only able to fill in the pre-1960/70 time trends by retrospective analysis of archived samples, or time resolved analysis of sediment cores. Now the ECD has largely been superseded by routine use of benchtop mass spectrometers (GC-MS) for most analysis of traditional POPs, showing how analytical developments have driven the science of research and monitoring of POPs.

By the mid-1960s, researchers applied these methods to screen biological samples from top predator species, such as birds of prey, otters and marine mammals. Swedish scientists reported high parts-per-million levels of polychlorinated biphenyls (PCBs) and pesticides in species and their potential for toxicological impacts.[10] This triggered one of the earliest examples of a ‘voluntary ban/restriction’ on chemical manufacture and use. Monsanto was the major manufacturer of the estimated 1.4 million tonnes of PCBs produced globally. [11] Markowitz and Rosner[12] recently published an interesting perspective. They conclude: Despite Monsanto's claim that it 'voluntarily' got out of the business (of PCB manufacture) in 1977, it was the concerted efforts of environmental activists, regulators, and the media that forced Monsanto finally to make this decision, because of its concern about the broad impact of the negative publicity on its image and ultimately its bottom line.' In 2018, a classic paper was published which argued that PCBs are probably responsible for major reproductive and immune system impairments to about half of the global population of killer whales (*Orcas*) [5] – even now - 50 years after PCB manufacture was curtailed.[11] Imagine if the early ban had not taken place; the situation for top predators could have been even more dire. With hindsight, the end result can be seen as a relatively rapid, very fortunate and far-sighted response. Now we have the Stockholm Convention, which is the international instrument to ban or restrict POP chemicals, but it was not in place until 2004 and the process to add chemicals to the list of restricted substances can take many years. **Table 1** summarises the compounds and current status of the Convention.

Co-ordinated collection of biological samples (biobanks, national archives) are an essential tool for early warning of chemical problems. This work started in earnest in the 1960s. Such collections have been invaluable for monitoring trends, providing definitive evidence of the changing chemical burdens in ecosystems, their response times following management interventions and providing clues about effects on key charismatic species.[13-15] Without them, there would not have been early warnings and restrictions on PCBs, DDT and other persistent and bio-accumulative chemicals.

POPs as multi-media chemicals and as tracers of processes

By the late 1970s/early 1980s, armed with the increasingly sophisticated analytical tools mentioned above, focus switched to understanding POPs on the regional scale and their multi-media transfer processes. Rather than simply studying direct 'visible' discharges of chemicals, attention started to focus on 'diffuse' atmospheric emissions and transport mechanisms. The North American Great Lakes provided a classic test system for the pioneering US and Canadian groups. Measurement, mechanistic understanding and modelling approaches were being brought together by researchers, such as Terry Bidleman, Steve Eisenreich, Ron Hites, Don Mackay, Derek Muir and Ross Norstrom. For the first time, these scientists and their teams shed light on the combined role of physical, chemical and biologically mediated processes on POPs. They brought clarity and quantitative understanding to: i. the potential for dynamic exchanges of POPs between air-water bodies and the water column and underlying sediments;[16-24] ii. mechanistic understanding to the processes of chemical bioconcentration, bioaccumulation and biomagnification; [25,26] iii. an appreciation of how effects of POPs on top predators can occur far from sources.[27-30]

As a student, I remember reading how Beluga whales living in the St Lawrence Estuary – 1000s of km from sources to the Great Lakes themselves – had accumulated such high residues of POPs that their bodies were classified as 'hazardous waste' if they were washed up on the shore.[31] After graduating with my PhD in environmental chemistry in 1984, and starting a faculty position at Lancaster University in 1985, I attended the annual North American SETAC conference in Toronto in 1987. I listened to the session on POPs, and was overwhelmed by the quality of the science being discussed, and excited by the way in which specialists in the chemical, physical and biological sciences came together to present and discuss their findings. A major theme at that conference concerned POPs transferring to the Arctic and the native peoples living there, in ecosystems which many people had previously believed were pristine. The interplay between good science and the political and societal ramifications were emerging and fascinating (indeed, a big push for the Stockholm Convention came from the indigenous peoples of the circumpolar countries – see Downie and Fenge[32]). I resolved to work on POPs myself at that time. Interestingly, in Europe, a couple of related issues were emerging then too. One was the role of long-range atmospheric transport (LRAT) in carrying acid rain, heavy metals and radionuclides from the UK and other heavily industrialised countries to sensitive terrestrial and freshwater ecosystems in Scandinavia. The other was a focus on dioxins and related compounds from waste incineration and other combustion sources. Incineration was attracting much political attention, but there were questions about whether this was the most important contribution to the dioxin inventory.[32-34] This required ultra-sensitive analytical procedures with high resolution GC-MS, so this aspect of European POPs work was led by strong analytical chemistry groups in Germany and Sweden, such as the teams of Karl-Heinz Ballschmiter, Otto Hutzinger and Christoffer Rappe. This research helped pave the way for the step-change in analytical sensitivity which would be required to work on POPs in remote ‘receiving environments’ such as the Arctic, Antarctic and deep oceans, and to address questions around possible pre-industrial/natural sources of POPs. There were obvious parallels to the pioneering work of Claire Patterson at CalTech, who had introduced a new level of analytical rigour into studies on the sources and pre-industrial levels of lead. This was needed to resolve the highly politically charged questions around the addition of lead to vehicle fuels and lead solder in food cans, and the link to lead’s possible neurotoxicological effects in modern societies.[35] His work led to a total re-evaluation of the growth in industrial lead concentrations in the atmosphere and the human body. Patterson had used an ultraclean chamber, one of the first ‘clean rooms’ and paid scrupulous attention to sample collection and handling, so that his measurements of isotopic ratios were free of the contamination from sampling equipment and modern ambient air and dust that confounded the findings of other groups at that time. Inspired by such approaches, as our work on POPs started in Lancaster, we learned how important it was to avoid samples becoming contaminated with ambient POPs from the lab[36] and how all-pervasive POPs can be from diffusive sources, which can hamper measurements in supposedly background environments.[37] All this was a steep but necessary learning curve, before we could properly investigate the global scale movement of POPs and before research on possible natural/pre-industrial versus modern anthropogenic sources could really begin in earnest. This was an important topic for dioxins and furans (PCDD/Fs) in the 1980/90s, when there was a prevailing view that their presence in the environment was ‘recent’, linked to chloroaromatic chemical production and incineration of wastes containing chlorine, such as PVC plastic. However, with careful control of sample collection and handling, elimination of sample contamination and ultra-sensitive detection, analysis showed that: i. PCDD/Fs could be detected in samples that pre-dated the Cl industry; ii. inefficient (low temperature) combustion of coal and wood (e.g. for domestic heating) and even volcanic activity also led to PCDD/F formation; different sources have characteristic compound signatures, which can be used as a source apportionment tool to improve emissions inventories. This all helped to inform the public and policy debate and lead to scientifically based source reduction programmes. [38-41]

POPs on a global scale - the 'big idea'

Science works best when there are 'big ideas' and well-articulated hypotheses to test. Such an idea emerged for POPs through the 1970/80s (see [42]) and was further extended in the early 1990s, in classic papers by Frank Wania and Don Mackay.[43,44] The 'global redistribution' hypothesis proposed that certain POPs can undergo LRAT, then be deposited onto the earth's surface, and potentially be re-emitted to cycle (hop) again. Temperature is a principle control of this tendency to re-emit, such that – over time and with repeated hopping if the chemical was sufficiently persistent – POPs would ultimately 'condense' into colder environments. The Arctic, Antarctic and mountainous areas would therefore become important sinks for some POPs. **Figure 1** shows the ideas presented in the original Wania and Mackay papers – highlighting the key processes of *cold condensation or distillation*, *fractionation* and *grass-hopping*. Their paper visualised and clarified processes that had been discussed in a classic paper by Brynjulf Ottar entitled *'The Transfer of Airborne Pollutants to the Arctic Region'* some years earlier.[45] Ottar's far-sighted article discussed re-emission, long-term transfer of mercury and chlorinated hydrocarbons from warm to cold climates, the concept of equilibrium between air and surfaces, different times required to attain equilibrium and the transfers to biota from source – all key parts of the global re-distribution hypothesis. Interestingly, *before* the science was resolved, the hypothesis acted to drive policy discussions and international agreements to restrict transboundary air pollution, because it invokes several important ideas. For example: it focuses on atmospheric transport as the key transfer pathways and therefore draws attention to trans-boundary issues; the Arctic represents the key receiving system; it identifies humans and top predators in remote locations as potentially vulnerable; and temperature is seen as the key driver, at a time when climate change was also emerging as a global concern.

However, in the late 1980s/early 1990s, systematic evidence *specifically* supporting cold condensation and global fractionation of POPs, and indeed evidence for large-scale global re-distribution of POPs *generally,* was very limited. Certainly POPs were being detected far from sources,[45] but that could simply be due to dispersion via LRAT, rather than repeated hopping. An early study of carbon tetrachloride in seawater had found higher concentrations in polar waters than in temperate or tropical ones,[46] but carbon tetrachloride is not a POP and more akin to the chlorofluorocarbons (CFCs) which were being linked to ozone depletion over the poles by that time.[47] Calamari et al [48] published an important early study, showing hexachlorobenzene (HCB) at higher concentrations in Arctic vegetation than temperate or tropical vegetation, but again – by itself – this was not definitive evidence, as there was an important confounding factor. Because there is no 'standard plant' growing everywhere, they had to compare long-living lichens, mosses and pine needles from cold places with short-lived tropical tree leaves. If plants have a high capacity to store airborne POPs in their waxy cuticles, they could continue to take up POPs over months or years,[49] so exposure times differed for the species tested. Later, this notion of a 'standard sampler' of air which could be deployed on networks and transects inspired the development and widespread application of passive air samplers (see below).

Against this back-drop, a series of scientific questions arose about the global re-distribution hypothesis: Is global re-distribution really occurring for a range of POPs and conditions?; What is the solid evidence for fractionation, cold condensation and hopping?; Are there really higher loadings and doses of POPs in remote cold places than in warmer source areas?; Is temperature the/a 'dominant driver' or are there confounding factors and other important processes controlling where POPs are and where they might go?; Is the Arctic really a major sink? Will POPs approach a 'global equilibrium' over time and – if so – how long will this take?; What will ultimately control the clearance of a POP from the environment, once it has been banned? Such questions led to a focus on studying 'background environments', away from the direct impact of sources.

These are challenging questions to address, because there are often important confounding factors. Proximity to/distance from sources, ongoing diffusive sources following a ban and varying source strengths over time can all confound the picture, whilst environmental gradients of factors other than temperature (e.g. precipitation, carbon stocks, ecosystem types) can impact what is deposited and retained in the environment.

Comments on the source term

A new chemical can spread into the environment from fresh 'primary' sources and subsequently be released from stocks, stores and environmental reservoirs, such as in-use products, waste dumps, soils and sediments, waters – so-called 'secondary sources'.[50] For most traditional POPs (e.g. PCBs, the pesticide dichlorodiphenyltrichloroethane (DDT) and other organochlorine (OC) pesticides, HCB – see Table 1) there has been a 'pulse' of chemical introduced into the environment, following increasing manufacture over several decades (e.g. the 1940-1960/70s), then a ban and decline in primary sources (e.g. 1980s-present). The input source term will therefore have been changing throughout this period, during which air-surface exchange moves the POP towards steady state between environmental compartments. Simultaneously - removal from surface compartments to deeper horizons of soils, water bodies and sediment, as well as degradation processes, have also been acting to remove the POP from the 'recyclable pool' (**see Figure 2**). Given that POPs are inherently persistent and the processes just mentioned act concurrently, measuring or deriving estimates of their loss/removal is challenging.[51-55] Meanwhile, for some industrial POPs such as PCBs and polybrominated biphenyl ether (PBDE) flame retardants, there could be substantial reservoirs of the chemical still in use, in products or in wastes. Products such as capacitors, electronic goods and furnishings, for example, that may contain industrial POPs can have life times of decades and they may themselves have contaminated buildings and infrastructures close to points of manufacture and use, such that they will slowly outgas or release POPs themselves over decades too. There is a parallel here with pesticides. Pesticide stocks continued to be used in some regions of the world long after they were banned. Unravelling the significance of different sources, reservoirs and sinks is a scientific challenge, but also crucial to inform policymakers, faced with the question 'is there more we can do to reduce POPs, or have we taken most practical measures already?' This, of course, is behind the Stockholm Convention's requirement for countries to conduct source inventories and monitoring of POPs.[1]

Gathering evidence to test the global re-distribution hypothesis

Unravelling these various processes has required a combination of research approaches and tools. These can be broadly grouped as follows:

Global scale source and emissions inventories: This is critical information, but often difficult to obtain reliably and rigorously. It may be difficult to know how much of a POP was manufactured or released, where and when. Tracking its use patterns can be very difficult too, and deriving estimates of emissions from products in use, or from environmental reservoirs is subject to large errors. Early work led by Knut Breivik on the global inventory for PCBs highlighted these uncertainties.[12,57,58] Later improvements became possible, as the early inventories helped identify the key uncertainties, so that an iterative process of measurement/modelling and refinement helped close the gaps between emission/release estimates and environmental measurements.

Long-term time trend data at air monitoring stations: This is a key source of information. It allows changing emissions to be inferred, and the role of environmental factors (temperature, wind speed and direction etc) to be understood. Such stations were first set up in the late 1980s/early 1990s around the North American Great Lakes (the Integrated Atmospheric Deposition Network)[59] and the Arctic and in the UK.[60,61] These networks are still running today and are a valuable source of information, from which the rates of POPs decline following bans and restrictions can be measured directly[62,63] and where the presence and increases of new compound classes can be detected. The paper by Hites in this Special Issue showcases what is possible.[64] Unfortunately reliable air measurements and time trends were not made directly pre-1990, because the analytical methods, sampling equipment, resources and political will were not robust enough, so the pre-ban trends have to be inferred by other means (see below).

Passive air sampling techniques: For the first time, passive air sampling allowed time-integrated measurements to be made in many locations around the world *simultaneously*. This was a critical requirement to enable source areas, gradients and remote areas to be identified, based on differences in concentrations. Latitudinal transects were established specifically to look for evidence of fractionation and cold condensation. A UK-Norway transect started in the early 1990s [65,66] and is still being maintained today. By the early 2000s, surveys were being undertaken at the continental scale in Europe, North America and Asia [67-69], while Tom Harner and colleagues established GAPS [70] - a global air monitoring network for POPs - which has yielded a wealth of key data. Passive air sampling is a tool which has been viewed with some scepticism, but is quick, convenient and low cost and has become widely used and accepted as a key asset in the Global Monitoring Programme (GMP) operated under the Stockholm Convention.[1]

Time trends from biological monitoring programmes: As discussed earlier, systematic biological monitoring schemes have proved invaluable in unravelling the changes in POPs over time. Programmes established in the 1960/70s have been maintained for a few areas (i.e. the Great Lakes, Sweden and the UK),[13,15,71-73] so there is a limited geographical/latitudinal coverage, but well designed and maintained biobanks are becoming a major resource for exposure monitoring and for research on possible effects.[3,4]

Retrospective analysis of archived samples and dateable sediment cores: Time trends have been obtained for decades/centuries from sediment and peat cores, enabling the rise and fall of certain POPs to be inferred,[19,74] while analysis of carefully preserved stored samples can give clues about historical levels.[38-41,75] Again, these studies are mainly for temperate industrialised countries in the northern hemisphere, so it is still difficult to build up a global picture of changing trends.

Ship-based transects of air and seawater: These can give snapshots of source-remote regions, and important information on the dynamic exchange or equilibrium status of water bodies with the atmosphere. They have helped identify important source areas and highlighted the close coupling of air-water-phytoplankton systems in the open oceans, showing that biota can have a key role in exchange and removal of POPs.[51,76-80]

Chemical marker techniques as indicators of fresh or weathered signals: Primary emission signals will be unaltered by the environment when they are first released. Chiral signatures and breakdown products can provide *definitive* evidence that a compound has spent time in soil or water bodies, so if they are detected in air they show that re-emissions must have occurred. Terry Bidleman has championed this approach[87,88] and there is a wonderful example in this Special Issue.[89]

Physico-chemically based multi-media fate models: These are the remaining key part of the POPs scientists' 'toolkit'. They can draw together estimates of emissions, transport and degradation/losses, to help make sense of the whole story. They can be used to highlight the main areas of uncertainty and to make forward projections. Their champion is Don Mackay and a succession of scientists from his research group.[25,26] There are many classic examples of their models being used as research tools to elucidate global scale processing of POPs.[84-94]

Primary (fresh) or secondary (re-emission) source dominated worlds

Before examining some of the evidence for the global re-distribution of POPs, it is helpful to imagine 2 hypothetical scenarios and what environmental measurements would reveal in each case.

A world still dominated by ongoing primary (fresh) sources of POPs and/or emission from products/stocks: Under this scenario, air concentrations would be highest close to sources/source regions and lowest in remote locations. The *rate of change in air concentration* would match the changing emission term and would presumably be similar near to the source and far away, assuming the air masses travelled from the source area to the remote area. Concentrations in systems receiving deposition would reflect the fact that the net flux is from air to the surface. There should be largely 'fresh and unweathered' chemical signatures in the air (minus any changes due to photodecomposition or atmospheric reactions), with little evidence of biological alteration of the signature.

A world approaching steady state and/or dominated by secondary sources (re-mobilisation from environmental reservoirs): Under this scenario, concentrations in the air would be closely coupled to their underlying surfaces. Air concentrations could be expected to fluctuate with surface temperatures as POPs are re-emitted. The loadings to/from surface compartments (soils, water bodies, vegetation) would be a function of their *storage capacity*,[95,96] balanced with losses (e.g. biodegradation). Because traditional ('legacy') POPs are lipophilic, this will reflect the organic matter (OM) loading of the surface compartment. In soils, OM represents a large store, much of which will have accumulated prior to the manufacture of POPs, but the surface layers will likely reflect several decades of POP production/accumulation and may therefore broadly reflect the balance between cumulative POPs deposition and any degradation. In lakes and oceans, the surface water's storage capacity for POPs may be strongly influenced by water temperature in oligotrophic systems which have a low loading of carbon, and by the biological (e.g. phytoplankton) carbon store in productive systems.[76]

What does the evidence reveal?

So, what have the environmental datasets shown us? Which scenario(s) are supported by the field data for different POPs?

PCBs: It is appropriate to use PCBs as a classic case, because we have the most complete information for the global emissions inventory and environmental measurements. They are also a family of chemicals with a gradation of physico-chemical properties, so they are ideal for looking for evidence of fractionation and variations in approach to equilibrium. In summary, this what the PCB story shows:

i. Global production and use peaked in the 1960/70s, with temperate industrialised countries in the Northern Hemisphere the main production and use areas.[12,57,58]

ii. In those areas of the world, air concentrations appear to have declined steadily since then, broadly matching the estimated trends for the atmospheric source inventories.[62,63,66,73]

iii. Air concentrations still follow an urban > rural > remote gradient[67] and the rates of decline are similar in different places on latitudinal transects. [62,66,97]

iv. There is fractionation of different PCB congeners along the urban, rural, remote gradient, in a way that is consistent with relative travel distances from primary source areas.[63,66,85] This means more volatile, lower molecular weight, photo-stable compounds travel further.

v. Time trends in biota tissue concentrations are also similar on latitudinal gradients.[15]

vi. Chiral PCBs in latitudinal air transects are still racemic, although in underlying soils they are not.[98] Preferential biodegradation of one stereoisomer over another in the soil occurs, but that pattern is not reflected in the overlying air, indicating that fresh inputs of PCBs still dominate the inputs to ambient air.

Essentially all these observations (i-vi) indicate that *the underlying trends in ambient PCBs have been/are still broadly controlled by fresh outgassing from sources or products from the past - even now - 50-60 years after voluntary bans on PCB production and use came into force worldwide*.

So, does this mean there is no evidence to support the global re-distribution hypothesis for PCBs? No - there is also evidence that PCBs have undergone re-cycling and dynamic exchange between the air and underlying surfaces:

vii. A number of studies have shown air concentrations undergo diurnal and seasonal cycling, with higher concentrations when surface temperatures are higher. This phenomenon has been observed above vegetated surfaces in rural areas, above coastal water bodies, over parts of the remote ocean and in urban areas.[77,99-103]

viii. Close coupling and near-equilibrium conditions have been reported between air and the surface of large water bodies.[20] Removal rates to deeper waters are comparatively slow,[51,53] and surface layers of oceans and lakes can buffer the atmosphere.[79,80]

ix. Terrestrial vegetation (grasslands, forests) has a large surface area covered in a thin layer of wax, which is subject to a large diurnal temperature variation. This has a high capacity to store and then exchange POPs with the atmosphere[101,104-106], so plant biomass is important in the re-cycling story.

x. There is a strong correlation between PCB concentrations and the OM content of global background soils. The correlation is strongest for lighter congeners, which can hop more readily than heavier congeners.[107] When the same locations were sampled in 1998 and 2008, the correlation became stronger with time,[98] indicating an approach to air-SOM equilibrium partitioning over periods of years/decades.[108] Soils with high OM status (which mainly occur in northern hemisphere temperate and boreal latitudes), have a very high storage capacity for POPs, probably making them a more important global sink than the ice-sheets and cold waters of the Arctic and Antarctic. Indeed, they may have served to 'protect' the Arctic from receiving such high loadings of POPs.

In short, the data for PCBs shows a complex interplay of many sources and many factors governing their cycling. Now, in some parts of the world, lighter PCBs in the atmosphere may be beginning to transition from being controlled by ongoing diffusive releases of fresh PCBs to reflecting a secondary source controlled world,[62,109] but it has taken many decades to reach this point.

However, the story of PCBs doesn't end there. Another important set of observations have been made in parts of Africa and Asia over the last 10-20 years. Despite the fact that PCBs were never widely used in these regions, some of the highest levels of PCBs have been recorded in these parts of the world. This might initially seem in total contradiction to the ideas of the cold condensation theory. However, the explanation is that there has been widespread bulk movement of PCBs (and PBDEs and other POPs) in waste materials exported from the areas of former higher use – primarily in Europe and North America.[110,111] In part, the declines in 'the industrialised west' may reflect the exports of such stocks.[112] Often the wastes are poorly dumped, sorted, or burnt on open fires, generating very high emission factors. PCBs, DDTs and other POPs are also continuously sweating out of soils which have been treated or contaminated in the past.[82,113,124] In a sobering calculation, Kurt-Karakus et al[115] estimated that soil treated with DDT could continue to emit it to the atmosphere for centuries to come. It has been estimated that a single informal waste dump in China emitted ca 1 tonne PCB/year to the atmosphere in 2006, more than the estimated emission for the whole of the UK in that year.[116]

There are probably hundreds of such sites across China, India, Pakistan and west Africa, for example. Imports and open burning of such wastes is now heavily controlled in China, but this has probably shifted the exporting (often illegally) of such wastes to other countries. For China, the legacy from handling such wastes will remain an important part of their emissions inventory and baseline human exposure well into the future. [117,118]

We are also still learning about previously unknown sources. Unintentional production of PCBs via combustion processes (e.g. metallurgical industries) and some manufacturing processes and re-mobilisation during forest fires and natural burning events have also only recently been recognised as unexpected contributors to the inventory.[119,120]

Brief comments on other 'legacy POPs' and related compounds - HCB, PBDEs, DDT, PCDD/Fs

Hexachlorobenzene (HCB) has lower air-surface partition coefficient values and longer atmospheric stability/residence times than PCBs, and it is therefore probably the POP that is furthest along in the transition to 'global equilibrium'.[121] It is reasonably well mixed in the atmosphere now and there is evidence for its cold condensation. However, it has a range of industrial, agricultural and combustion-derived ongoing sources, some of which are 'unintentional' as defined by the Stockholm Convention,[1] which complicate the picture.

In broad terms, the rise and fall of the flame retardant chemicals polybrominated diphenyl ethers (PBDEs) and their primary versus secondary global source areas, latitudinal fractionation and the regional re-distribution through transport in wastes mirrors that of PCBs.[98] However, the input/decline 'pulse' is delayed by 20-30 years, because this class of compounds was manufactured and restricted more recently.[122]

The insecticide DDT was used in large quantities as an agricultural chemical in North America and Europe in past decades, but its use phased out in the 1970s in the USA. There has been widespread ongoing use in the tropical zones, because of its use as an insecticide in treating malaria.[123] However, like many POPs, there is a surprisingly complex story of unexpected and ongoing sources.[130] Degradation forms an important part of its changing global mass balance and the trends in biota bioaccumulation.[15] Polychlorinated dibenzo-p-dioxins and –furans (PCDD/Fs) and polynuclear aromatic hydrocarbons (PAHs) are 'unintentionally produced' [1] primarily through different combustion sources to the environment, both natural and anthropogenic. PCDD/Fs were also impurities in a range of industrial chemicals. The trends for both groups have generally been declining in many industrialised countries, as combustion and industrial sources have been controlled. Diffusive sources, which are more difficult to control, probably now dominate fresh releases. However, concentrations may be rising in some developing countries, linked to greater industrialisation, combustion and waste generation.[131]

Comments on the Stockholm Convention and its effectiveness

In 2004, the Stockholm Convention listed 12 substances as POPs for internationally agreed control (PCBs, HCB, PCDD/Fs, HCB, DDT and other organochlorine pesticides) (Table 1). The Convention provides a mechanism for compounds and chemical classes to be proposed as POPs, thereby making them the subject of international bans/restrictions. That process has brought forward other compound/classes which have very similar properties to these original 'legacy dirty dozen POPs' (see Table 1). However, in recent years some perfluorinated (PF) compounds, which have higher aqueous solubilities and complex reaction chemistries, have also been proposed and added to the list. For these substances, the issues of persistence are similar, but the primary modes of transport through the environment can be via aquatic discharges, riverine flow, ocean currents and sea spray[70,99,132-135] (see also Sha et al.[136] in this issue). There are also interesting issues to address, because the compounds targeted (PFOS, PFOA) are breakdown products of other PFs which are themselves chemicals of commerce. Research on these compounds has therefore also focussed on comparing modes of transport (water versus air) and seen the development of passive *water* samplers,[137,138] so that regional and global mapping of PFs is becoming possible, in the same way that passive air samplers have been widely deployed for traditional POPs.

The Stockholm Convention drives a huge effort through the Global Monitoring Programme, which is generating co-ordinated datasets which can be used to assess the effectiveness of the Convention.

This has really gathered momentum over the last decade and shows the power of mature international co-operation. Jana Klanova and her team in Brno, Czech Republic have had a great part to play, [139] and their article in this Special Issue gives recent updates and perspectives.[68] As the earlier example of exporting/importing of hazardous wastes illustrated, the work to support the SC also needs close co-operation with the Basel and Rotterdam Conventions, which address international movement, handling and disposal of wastes.[140,141] The European Commission also recently decided that by 2022, persistent, mobile and toxic/very persistent very mobile (PMT/vPvM) substances should be included as a new category of substances of very high concern (SVHC) under European REACH legislation.

What have we learned from POPs?

If we take a step back from the details, what have we learned from POPs that can help us improve our management of chemicals in future? Perhaps we could summarise this simply as:

POPs get everywhere, often via practices, pathways and processes which were not originally envisaged. Chemicals management therefore needs to address the whole life cycle of use/products, and envisage unexpected consequences of poorly managed use; POPs take a very long time to disappear and the current methods of international control are slow and cumbersome; in general, the world is a better place without them. All of this implies chemicals management needs to be inherently pre-cautionary and risk-averse. This is especially true, given the toxicological/ecotoxicological and public health concerns surrounding POPs; these chemicals should not be allowed into our environment.

The bigger societal, science and policy picture of chemicals management

As just described, slowly and step-by-step, there are international mechanisms working to rid the world of troublesome persistent chemicals. This is great news, but it is – of course – addressing a ‘downstream’ problem. Addition of chemicals to the Stockholm Convention list takes a long time and – meanwhile – the scale of chemical use and the rate at which new chemicals are being designed and manufactured is staggering. For example, although I mentioned earlier that the global of production of PCBs was a large number – 1.4 million tonnes altogether – that is completely dwarfed by the scale of *total* chemical release, which has been estimated at *200 billion tonnes per annum*.[142,143] This figure includes the full array of chemicals of commerce, most of which are more benign than POPs, but it represents a huge total loading into the planetary system. There is also an ever-increasing 'cocktail' of chemicals reaching our ecosystems. Over 235,000 chemicals are in use by society around the globe[144] and new synthetic chemicals are being developed, at a rate of 2000-3,000 per annum in recent years.[145] In 2011, world sales of chemicals were estimated at $3,500 billion, equivalent to ~$500 per year for every person on the planet.[146] Economists may consider this a 'good thing', but to me it is signalling an ever increasing and unsustainable dependency on chemicals. There is a danger we can get drawn into a cycle of chemical invention, marketing/manufacturing, use, ban/restriction, replacement/substitution.[145,147,148] So, a big future challenge is: how we can take our knowledge of chemical fate, behaviour and effect, to stop new potentially troublesome chemicals from being approved and registered in the first place?

At a time when the European Union has introduced greater transparency and accountability to the chemicals registration process through its REACH legislation,[149,150] we have also witnessed a shift in where chemicals are made and wastes are handled globally, with China and India becoming the 'factories of the world' and current projections suggesting production could double in just the next 10-15 years.[146] China is now the largest chemicals producer in the world, contributing 36% of global chemical sales in 2018.[146] China is doing much to introduce its own chemicals and management process,[151-153] and this is obviously a critically important opportunity to try and ensure best practice for the benefit of future global chemicals management.[154]

Regardless of the precise chemical species, these figures point to an enormous consumption of the Earth's resources, and huge demand on its 'carrying capacity' and ability to cleanse itself. Of course, POPs and other forms of chemical pollution are just one of the many pressures and stressors on our planetary system. This is occurring in concert with land use change, biodiversity losses, increased urbanization, and climate change.[155] Indeed, chemical pressures interact with, and can accelerate, some of those planetary changes too (e.g. CFCs and climate change; pesticides and biodiversity).

Meanwhile, in humans, the global burden of cognitive, reproductive and developmental disorders and diseases linked to environmental pollution is rising in recent decades, with ~5 million deaths attributed to environmental exposure and management of selected chemicals each year.[156] The European Environment Agency[157] has highlighted environmental impacts from chemicals as an area of major concern and predicted that these impacts will increase in the future due to global megatrends, such as increasing urbanisation and climate change.

So, nearly 100 years after the first POPs were invented, where are we in the grand scheme of things? Where are we now and what will the next decades bring?

Properly regulated chemicals provide societal benefits. However, there are many gaps in our understanding of how chemicals impact the environment and how to accurately assess, predict and manage these risks. My sense is that, right now, our scientific community is overwhelmed with the scale of the chemicals management challenges we are being asked to address. The numbers of chemicals are so large, the demand for tools and resources to manage them is enormous, and the topic is hugely complex. Our response, as a scientific community, has therefore been to develop ways of *listing, screening, ranking and prioritising* the thousands of chemicals currently being manufactured, to use 'logic' in helping to assign resources and effort. Such schemes can use an array of approaches, such as: physico-chemical property measurement and estimation; in-silico prediction and computational machine learning to design and 'categorise' compounds and identify problems and unintended consequences early; inventories; non-target screening of compounds in environmental and biological indicator sample; targeted screening; bio-toxicity assays; read-across methods; modelling; hazard ranking; and risk assessment.[144,158-160] These are all vitally important endeavours and will keep us all busy far into the future.

However, over and above this, there is an even bigger agenda and pressure on us, as a global community and civilisation. We are faced with stark realities. How best to prioritise the use of the planet's natural resources – the stocks of carbon, nutrients, industrial and precious metals etc. It has been estimated that humans have now consumed the majority of all the Earth's petroleum reserves.[161,162] So, which organic chemicals (which largely use fossil fuels as the feedstock) should be made and why? Which do we really need, that justify using the planet's resources to make? Plastics, personal care products, agrochemicals, antibiotics and other pharmaceuticals…? How can we decide and – perhaps more importantly – who should decide? Should that be the chemicals industry, who can create and 'market' demand? Should it be the 'resource holders' – governments, multi-national companies, other stakeholders, or customers and the free market, or international agencies? How can scientists become involved and help drive the debate?

Our global community has identified and committed to 17 Sustainable Development Goals.[163] Pollution and chemicals management is relevant to most, if not all of them (e.g. number 3 – good health and wellbeing; 6 – clean water and sanitation; 12 – responsible consumption and production). Delivering these goals by 2030, or even coming close to delivering some of them, needs knowledgeable, passionate and committed environmental scientists with great communication skills. It's time to get involved!

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Table 1: POPs currently listed in the Stockholm Convention (October 2020)

Annex A (Elimination)

Aldrin*
Chlordane*
Chlordecone*
Decabromodiphenyl ether+
Dicofol*
Dieldrin*
Endrin*
Heptachlor*
Hexabromobiphenyl+
Hexabromocyclododecane+
Hexabromodiphenyl ether & heptabromodiphenyl ether+
Hexachlorobenzene*+
Hexachlorobutadiene+
Alpha hexachlorocyclohexane*
Beta hexachlorocyclohexane*
Lindane*
Mirex*
Pentachlorobenzene*+
Pentachlorophenol*
Polychlorntaed biphenyls (PCBs)+
Polychlorinated naphthalenes+
Perfluorooctanoic acid (PFOA) and related compounds+
Short-chain chlorinated paraffins+
Technical endosulfan*
Tetrabromodiphenyl ether and Pentabromodiphenyl ether+
Toxaphene*

Annex B (Restriction)

DDT*
Perfluorooctane sulfonic acid and related compounds+

Annex C (Unintentional production)^

Hexachlorobenzene
Hexachlorobutadiene
Pentachlorobenzene
PCBs
Polychlorinated dibenzodioxins
Polychlorinated dibenzofurans
Polychlorinated naphthalenes

Proposed for listing under the Convention (as of October 2020)

Dechlorane Plus+
Methoxychlor*
UV-328+

KEY:
*Pesticide
+Industrial chemical
^Note that some chemicals are deliberately manufactured as well as being unintentionally produced.

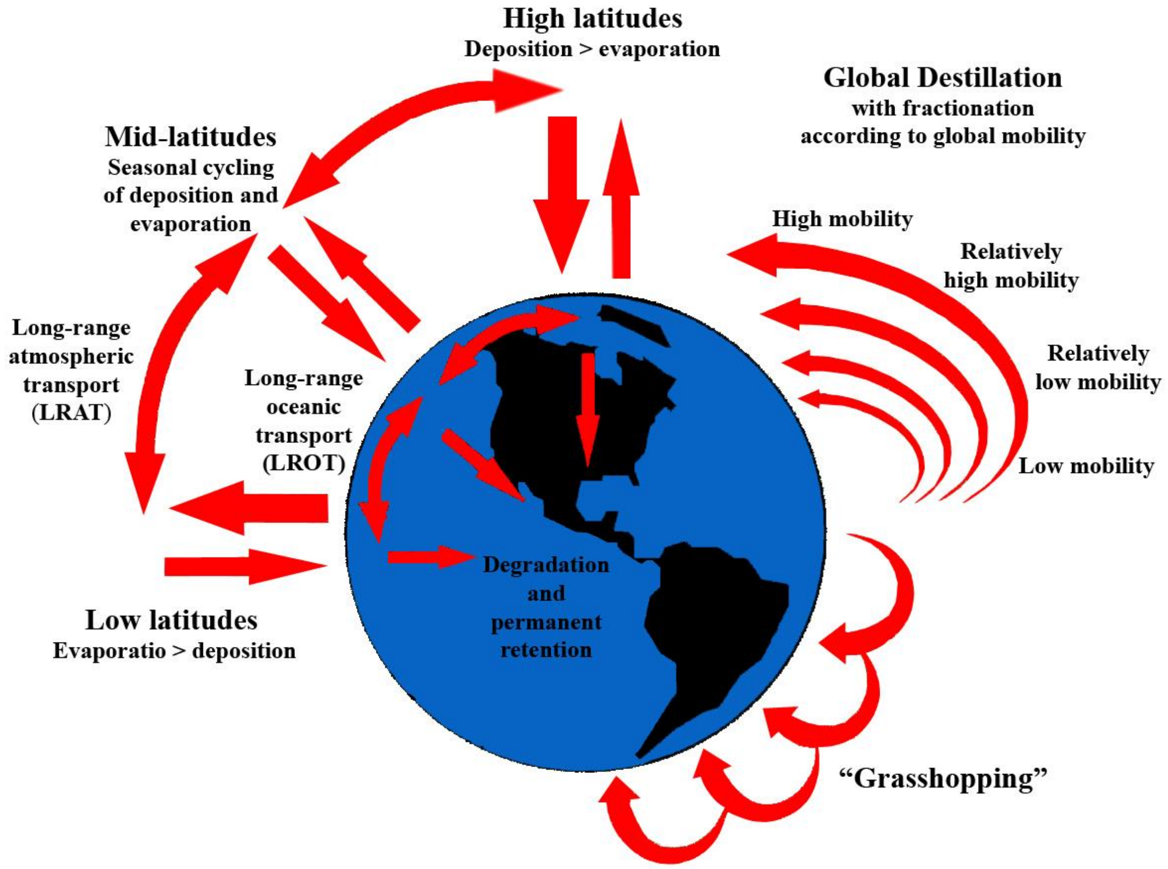

Figure 1. Global cycling of POPs (after Wania and Mackay 1996, adapted by Schuster, 2008)

Figure 2: Illustration of some of the key compartments and processes controlling the environmental cycling of POPs (prepared by Jasmin Schuster).