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Supporting Information

How much persistent organic pollutants have been produced and released into the global environment?

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Text S1. Literature-reported information on production and environmental releases

Aldrin and dieldrin

Aldrin and dieldrin were first commercialized in the U.S. and The Netherlands in 1950.[1] Before 1968, the U.S. was the only producer of aldrin and dieldrin, with an average annual production of ~9,000 tonnes (~20 million lbs as reported) in the mid-1960s.[2] In addition, the annual global production of aldrin and dieldrin is reported to be 13,000 tonnes in 1972 and decline to <2,500 tonnes in 1984.[1] These two numbers are used in this work to represent the average annual global production volumes in the 1970s and 1980s. In this work, we assume the production volumes comprised 90% aldrin and 10% dieldrin, based on the reported shares of these two compounds in the U.S. in the mid-1960s.[2]

While the U.S., which was historically the dominant producer of aldrin and dieldrin, ceased the production of these two compounds by 1990, the production continued in several other countries including Germany, the United Kingdom, and Belgium until 2016.[3] While aldrin and dieldrin were reported to be "used throughout the world" in the late 1960s and 1970s,[1] it is unclear about the relative shares in the U.S. and Europe. For simplification, we assume that each the U.S. and Europe consumed one-third of the annually produced aldrin and dieldrin throughout history. Aldrin and dieldrin have never been produced or used in China.[4]

Chlordane

Chlordane was first commercialized in the U.S. in 1947.[5] The annual production in the U.S. was estimated to be 12,000 tonnes (~9,500 tonnes for domestic use and ~2,300 tonnes for export)[5] in the 1970s and 45–450 tonnes in the early 1990s.[6] The annual use in the U.S. was reported to be 4,500 tonnes in the early 1980s and 1,600–1,800 tonnes in 1986.[7] The use of chlordane in the U.S. ceased in 1988 when the U.S. Environmental Protection Agency canceled its registration as a pesticide.

Historical data on the annual production and use of chlordane in China were taken from Wang et al.[8] Those data show that China produced a cumulative total of ~1,500 tonnes of chlordane for export throughout the 1970s and a cumulative total of ~3,500 tonnes for domestic use in the late 1980s and throughout the 1990s.

Since chlordane has not been produced in Europe or Japan,[9] we assume that the annual global production and use are equal to the sum of those in the U.S. and China and ignore the use of chlordane in Europe.

Chlordecone

The U.S. was the sole producer of chlordecone between 1956 and 1975.[10] During this period, a cumulative total of ~1,600 tonnes of chlordecone was produced,[11] with more than 1,500 tonnes between 1968 and 1975 and only 12 tonnes in the 1950s.[11] Another estimate indicates that the annual production reached ~500 tonnes in the early 1970s.[5] Brazil became the major producer after the production ceased in the U.S., with cumulative production of 200 tonnes from 1981 to 1991.[12] Therefore, the cumulative global production of chlordecone amounted to 1,800 tonnes throughout history.

Of the historically produced chlordecone, 300 tonnes were used in the archipelago of Antilles,[12] up to 70 tonnes in the U.S. (before 1975),[11] and the rest was exported to Europe (mainly to Germany before the 1980s), Africa, and other regions in Latin America.[11, 12] For simplification, we assume that half the rest of the world's production was used in Europe. Chlordecone has never been used in China.[13]

Commercial polybrominated diphenyl ethers

PBDEs were first commercialized in the early 1970s. Abbasi et al.[14] aggregated data on the annual global production from 1970 to 2020. Their results show that a cumulative total of ~175,000 tonnes of C-pentaBDE, ~130,000 tonnes of C-octaBDE, and ~1,600,000 tonnes of C-decaBDE have ever been produced worldwide. We average their yearly data for decadal estimates of production from the 1970s to 2010s.

Based on the collected data on the annual global production of six congeners (BDE28, 47, 99, 153, 183, and 209), Abbasi et al.[14] simulated the time-variant, geographically-resolved environmental releases of these congeners from 1970 to 2030 using substance flow analysis modeling. The model calculated that 22–345 tonnes of BDE99, 9–116 tonnes of BDE183, and 1,506–15,175 tonnes of BDE209 have been released into the global environment by 2020. Given that the congeners BDE99, BDE183, and BDE209 account for ~49%, ~30%, and 94% of the total mass in C-pentaBDE, C-octaBDE, and C-decaBDE, respectively,[15] we scale these congener-specific release estimates to numbers for three commercial mixtures: 45–705 tonnes of C-pentaBDE, 30–380 tonnes of C-octaBDE, and 1,602–16,143 tonnes of C-decaBDE. Our scaled results indicate that the annual environmental releases of all the C-pentaBDE, C-octaBDE, and C-decaBDE in both the U.S. and Europe peaked in the 1990s or 2000s. However, in China, the annual environmental releases of C-pentaBDE and C-octaBDE peaked in the 1990s or 2000s as well, whereas the annual environmental releases of C-decaBDE mixtures peaked later between the 2000s and 2010s.

DDT

Schenker et al.[16] compiled, compared, and curated estimates of annual global production of DDT from 1940 to 2005. Those results show that the annual global production of DDT increased from ~2,000 tonnes in 1941 to ~150,000 tonnes in the early 1960s and then declined to ~7,000 tonnes in 2005, with a total cumulative volume of 4.5 million tonnes. These estimates were assumed to be associated with an uncertainty following the log-normal distribution, with its geometric standard deviation to be 1.14 times central tendency estimates.[17] Based on this number, we calculate the 95% confidence interval (i.e., the fold change between 97.5th and 2.5th percentiles as the upper and lower bounds, respectively) of the uncertainty range to be 2.5, which is therefore assigned as the uncertainty range factor in this work.

Schenker et al.[16] also critically compared different sources of data on the regional distribution of the historical global use, indicating that North America (mainly the U.S.) and Europe consumed 20% and 7% of the historically produced DDT worldwide, respectively. We use these ratios to allocate the global DDT use during the period 1940–1990 to the U.S. and Europe. The use of DDT in the U.S. and Europe has been rare since the early 1990s due to restrictions in these two regions.

After the ban of its agricultural use by the Stockholm Convention, DDT continued to be produced in China and India for malaria control. The average annual production was 5,144 tonnes between 2003 and 2007 and 3,491 tonnes between 2008 and 2014.[18] Most of the produced DDT was used by India and African countries such as Ethiopia, Mozambique, and Zimbabwe.[18] China banned the agricultural use of DDT in 1983 but kept producing DDT until 2005. In the 2000s, the production was for export for malaria control (~20%), manufacturing technical dicofol (~70%), and producing DDT-based antifouling paints for domestic use (~5%).[13] Here, we do not consider the environmental releases of unreacted DDT impurities along with the use of technical dicofol. We assume that DDT in antifouling paints is completely released into the environment. Limited data between 2003 and 2006 show that North Korea produced 1,000 tonnes per year for domestic agricultural use; however, data after 2006 are not available.[18]

Dicofol

Dicofol was first introduced commercially in 1955. In the U.S., the annual sales of dicofol amounted to 1,400 tonnes in the 1980s.[19] The U.S. Geological Survey estimated that on average 410–450 tonnes of dicofol were used across the conterminous U.S. in the 1990s.[20] A survey in 2003[13] indicates that China started to produce dicofol in 1976, with the annual production increasing from ~2,000 tonnes in the late 1980s to ~5,600 tonnes in 1999 and then remaining 3,000 tonnes since 2000.

A survey conducted most likely in the late 1990s indicates that Europe and North America (mainly the U.S.) contribute to equally ~10% of the annual global dicofol consumption.[21] Based on this estimate, we assume that the annual use of dicofol in Europe was equal to that in the U.S. between the 1980s and 1990s.

Li et al.[22] compiled and curated estimates of the annual global production and use of dicofol between 2000 and 2012. The data show that the annual global production decreased from 3,350 tonnes in 2000 to 725 tonnes in 2012. During that period, the annual production declined from

~2,500 tonnes to 600–700 tonnes in China, from 300 tonnes to 30 tonnes (discontinued in 2009) in Europe, and from 300 tonnes to 30 tonnes in North America (mostly in the U.S.).[22]

Endrin

Endrin was produced by two U.S. manufacturers between the 1950s and 1980s.[23] The U.S. sold 2,300–4,500 tonnes of endrin in 1962 alone.[23] In 1975, the then sole U.S. manufacturer produced 1,360–2,700 tonnes (3 to 6 million lbs as reported) of endrin.[24] The U.S. exported endrin for use in other countries; for instance, up to 10 tonnes of endrin were reported to be used each year in Bail, Indonesia between 1963 and 1972.[23] After the production ceased in the U.S. in 1982, Mexico continued producing endrin for years.[23] However, since detailed information on the annual production in Mexico is unavailable, we ignore Mexico' s share in this work. Endrin has never been produced or used in China.[4]

Heptachlor

Heptachlor was first commercialized in the U.S. in 1952. The U.S. was the leading producer of heptachlor, and it produced 2,700 tonnes in 1971, 900 tonnes in 1974, 590 tonnes in 1978, 180 tonnes in 1980, and 45 tonnes in 1982.[7] By comparison, China only produced a cumulative total of 17 tonnes of heptachlor between 1967 and 1978.[13] Since there is no record of heptachlor production in Europe, we assume that the annual global production and use are equal to the sum of those in the U.S. and China. Based on the numbers outlined above, we assume that the annual production of heptachlor was on average 2,700 tonnes in the 1960s, 1,000 tonnes in the 1970s, and 500 tonnes in the 1980s.

Most of the U.S. produced heptachlor was exported for use in other countries. The use of heptachlor in the U.S. was reported to be 340 tonnes in 1986 and then ceased in 1988.[7] An earlier estimate shows that the U.S. and Europe used 5% and 60%, respectively, of heptachlor produced globally in the year 1970[25], based on which we allocate the annual global use to the U.S. and Europe.

Hexabromobiphenyl

The U. S. Environmental Protection Agency conducted a comprehensive survey on the production, market, use, and environmental releases of hexabromobiphenyl.[26] The U.S. was the sole producer of hexabromobiphenyl, with the production occurring only between 1970 and 1976.[27] During this period, a cumulative total of 5,350 tonnes (11,197,900 lbs as reported) of hexabromobiphenyl was produced for domestic use as a flame retardant in ABS plastics, coatings and lacquers, and polyurethane foam.[26] There was no record of hexabromobiphenyl export to other countries.[26] Historical information indicates that manufacturing 1 tonne of hexabromobiphenyl led to the release of 0.051 tonnes, primarily as solid waste to landfills.[26]

Hexabromocyclododecane

Li et al.[28] aggregated data on the annual production of technical HBCDD in seven regions worldwide between 1965 and 2016 and extrapolated the annual production to the year 2020 assuming a linear decline to zero after the implementation of the Stockholm Convention. Their results demonstrate a cumulative production of 703,000 tonnes during that period, with a steady increase over years before a peak at 41,000 tonnes in 2013 and a sharp decline thereafter. We average their yearly data for decadal estimates of production from the 1960s to 2010s. The production of technical HBCDD in Europe and North America started in the late 1960s and plateaued in the early 2000s (at 6,200 and 10,000 tonnes per year, respectively), before declining to ~3,000 and ~7,000 tonnes in 2016, respectively. By contrast, the production of technical HBCDD in China started in 2000 and peaked at 30,000 tonnes per year in 2013.

Using a dynamic substance flow analysis model, Li et al.[28] estimated the annual environmental releases of individual HBCDD isomers in different regions from 1965 to 2100. The model calculated that 400–1,200 tonnes of HBCDD have been released into the environment by 2020; it predicted that another 200–400 tonnes of HBCDD will be released from the "stock" of in-use construction materials and demolition waste during the future period 2021 through 2100. In Europe and North America, the annual environmental releases of HBCDD peaked in the 2000s, with annual averages of 3.1–8.6 tonnes and 5.3–14.6 tonnes, respectively. In China, the annual environmental releases of HBCDD peaked in the 2010s, with an annual average of 8.7–26.5 tonnes.

Hexachlorobenzene (HCB)

HCB can be intentionally produced for use as a fungicide, unintentionally formed during thermal or combustion processes, or unintentionally generated as a by-product or impurity in the manufacture of several chlorinated chemicals or solvents, such as dimethyltetrachloroterephthalate (at a typical concentration of 1,000 ppm), pentachloronitrobenzene (500 ppm), lindane (50 ppm), and pentachlorophenol (50 ppm).[29] In this work, we consider only the intentional production and associated environmental releases, given that the unintentional formation and generation are relatively smaller (accounting for <20% of the global total environmental releases[30]) and are also out of the scope of this work.

The global production of HCB dated back to 1945. The use of HCB as a fungicide ceased in the U.S. in 1985, in Mexico in 1991, and probably last in Russia in 1996.[30, 31] Numbers collected and curated by Barbere et al.[32] indicate that the annual global production was 1,000–2,000 tonnes in the mid-1970s and then increased to ~10,000 tonnes between 1978 and 1981. Although the annual global productions in the 1950s and 1960s were not available, they are likely to be 1,000–2,000 tonnes, similar to the level in the mid-1970s, given that the annual production in the U.S. was surveyed to be stable throughout this period: 345 tonnes in 1958, 327 tonnes in 1959, 200 tonnes in 1960, and 317 tonnes in 1973.[31]

Approximately 10,000 tonnes of HCB were produced each year in the late 1970s, in which the shares by Europe, the U.S., and Mexico were ~8,000 tonnes (based on data for 1978–1981), 454 tonnes (data for 1977), and ~3,500 tonnes (data for the 1970s), respectively.[32, 33] Based on these data, we assume that Europe and the U.S. contributed to 80% and 4% of the annual global production between the 1940s and 1980s due to the lack of temporally resolved region-specific data. We further assume that the production of HCB was for domestic use only, whereby we estimate annual environmental releases in Europe and the U.S.

Hexachlorocyclohexane

Environmental releases of hexachlorocyclohexane result from the use of both technical HCH and technical lindane products. The technical HCH product typically comprises 55–80% of α-HCH (an average of 67.7% was recommended for use in calculations), 5–14% of β-HCH (an average of 5.5% was recommended for use in calculations), and 8–15% of γ-HCH (an average of 14% was recommended for use in calculations).[34, 35] By contrast, γ-HCH is almost the single dominant component in the technical lindane product.[34, 35] Therefore, we calculate the annual productions of α-HCH and β-HCH as the annual production of technical HCH multiplied by corresponding mass fractions (67.7% and 5.5%, respectively), and we calculate the annual production of γ-HCH as a combination of the annual production of technical HCH multiplied by the corresponding mass fraction (14%) and the annual production of technical lindane multiplied by 100%.

The total cumulative global use of technical HCH was estimated to be 9.7 million tonnes for the years 1948 through 1997, with China (4.5 million tonnes), India (1.1 million), European countries (~1.5 million tonnes, including the Soviet Union), and the U.S. (~0.35 million tonnes) being major consumers.[34] Specifically, the annual global use of technical HCH amounted to on average 110,000 tonnes in the 1950s and 220,000 tonnes in the 1960s, peaked at ~350,000 tonnes in the 1970s, dropped to on average 265,000 tonnes in the 1980s, and remained at 25,000 tonnes between 1990 and 1995.[36] Another estimate indicates that the total global usage of technical HCH was 40,000 tonnes for the year 1980 and 29,000 tonnes for the year 1990.[36]

As for region-specific production or usage, Li et al.[37] estimated the annual technical HCH production in China from 1952 to 1984, with an increase before 1972, two peaks at 280,000 tonnes in the years 1972 and 1980, and a decrease thereafter to zero in 1984. Li et al.[36] estimated that India used on average ~3,000, ~10,000, ~20,000, ~26,000 tonnes per year in the 1950s, 1960s, 1970s, and 1980s, and the U.S. used and released up to ~6,000 tonnes per year only in the 1950s (numbers are extracted from Figure 1a of that paper). In Europe, Breivik et al.[35] estimated that 382,000 tonnes of technical HCH were used from 1970 to 1996, decreasing from ~35,000 tonnes in the 1970s to ~500 tonnes in 1996. The difference between the historical cumulative use in Europe (1.5 million tonnes[34]) between 1948 and 1997 and that between 1970 and 1996 (382,000 tonnes[35]) represent the summed use in the 1950s and 1960s. We assume that this quantity is equally distributed in the 1950s and 1960s due to a lack of time-specific data.

The total cumulative global use of technical lindane was estimated to be 435,000 tonnes for the years 1950 through 2000, with Europe (287,160 tonnes), Asia (73,200 tonnes), and America (63,570 tonnes) being major consumers.[38] The total global usage of technical lindane was estimated to be 5,900 tonnes for the year 1980 and 4,000 tonnes for the year 1990.[36] The production of technical lindane ceased in the early 2000s in most countries, although it may continue in small quantities in several countries.[39]

As for region-specific production or usage, Li et al.[37] reported that China started to produce and use technical lindane in 1990; China produced on average 1,000 tonnes of technical lindane each year in the 1990s and used cumulatively 28% of the production during this period. Li et al.[36] estimated that the U.S. used on average ~4,000, ~1,000, ~150, and ~100 tonnes per year in the 1950s, 1960s, 1970s, and 1980s.[36] In Europe, Breivik et al.[35] estimated that 81,000 t of technical lindane were used from 1970 to 1996. Based on the data presented in Breivik et al.[35], we back-calculated the annual use of technical lindane to be 2,100~3,500 tonnes over the years. The difference between the historical cumulative use in Europe (287,160 tonnes[38]) between 1950 and 2000 and that between 1970 and 1996 (81,000 tonnes[35]) represent the summed use in the 1950s and 1960s. We assume that this quantity is equally distributed in the 1950s and 1960s due to a lack of time-specific data.

We are unclear about the uncertainty associated with these estimates because no information is available in the literature. However, since these estimates were developed based on binned pesticide use density (for instance, 0.1–1 tonnes of technical HCH or lindane per thousands of hectares for the "low" use density bin), where the upper and lower bounds of each bin differ by factors no greater than 10, we believe the overall uncertainty associated with these estimates is likely to be within an order of magnitude.

Mirex

Historically, mirex was produced only in the U.S. and China. In the U.S., mirex was produced as a pesticide for pest control between 1958 and 1976. Historical statistics indicate that a cumulative total of at least 225 tonnes were produced during the period 1958–1967,[5] that a cumulative total of 400 tonnes were used for pest control between 1959 and 1975,[40] and that a cumulative total of 250 tonnes were used for pest control between 1962 and 1975.[40] Based on these numbers, we estimate the total use for pest control to be 150 tonnes during 1958–1962, 75 tonnes during 1962–1967, and 175 tonnes during 1967–1975. In addition, mirex was used under the trade name of Dechlorane, with a cumulative total of 1500 tonnes between 1959 and 1975, for non-agricultural applications, mostly as a flame retardant.[40]

In China, mirex was produced for pesticide control only. The use of mirex dated back to the 1960s. A 2003 national survey shows that on average 2–3 tonnes of mirex were used each year for termite control between 1992 and 2001.[13] At least 10 manufacturers produced mirex in 2003 although China had canceled the national registration of mirex in the 1980s.[13] For simplification, we assume the annual use of mirex in China has remained 2–3 tonnes throughout history.

Pentachlorophenol (PCP) and its salts and esters

Pentachlorophenol and its salts and esters, such as sodium pentachlorophenate (NaPCP) and pentachlorophenyl laurate (PCPL), were first produced in the 1930s.[41] Historical data indicate that the annual global production reached 90,000 tonnes in the 1980s[42] and maintained 8,500 to 50,000 tonnes in the 1990s.[43] The annual global production was likely to be 20,000 tonnes in the 1960s, based on an estimate in the early 1970s.[5] At least 30,000 tonnes were likely to be produced each year in the 1970s, given that European countries produced ~10,000 tonnes each year in the late 1970s, the U.S. produced ~10,000 tonnes each year in the 1970s, and Japan produced 14,500 tonnes in 1966 and 3,300 tonnes in 1971.[42] The annual global production was likely to be 8,000 tonnes in the 2000s and 2010s given that Mexico, as one of few remaining producers in the world, produced 7,257 tonnes for use in North America in 2009.[41]

Europe stopped the production of PCP and NaPCP in 1992,[44] after which, according to limited data records, Europe imported 408 tonnes (378 tonnes of PCP and 30 tonnes of NaPCP) in 1996 and 339 tonnes (15 tonnes of PCP and 324 tonnes of NaPCP) in 1999. [44] The U.S. produced 10,900–20,400 tonnes (24 million to 45 million lbs as reported) each year between 1983 and 1987, with less than 10% of the production exported to other countries.[44] The U.S. produced 7,300–9,500 tonnes of PCP each year and used 6,800–7,300 tonnes each year between 1995 and 1999.[45] The U.S. produced 1,361–1,815 tonnes of PCP and used 5,000–5,500 tonnes for the treatment of utility poles, lumber, and timbers (construction) in 2002.[41] In addition, the U.S. imported 3,670–7,343 tonnes of PCP from Mexico for use between 2007 and 2011.[41]

China produced "very limited" amounts of sodium pentachlorophenate between 1958 and 1983.[13] The production of sodium pentachlorophenate was resumed in 1988, with an average annual production of 7,000 tonnes between 1988 and 1989, 7,700 tonnes in the 1990s, and 2,800 tonnes in the 2000s.[13] China produced 203 tonnes of pentachlorophenol between 1979 and 1994 (the production completely ceased in 1995).[13] The cumulative production was estimated to be 203 tonnes between 1979 and 1983; the annual production was estimated to be on average ~1,200 tonnes in the 1980s and ~550 tonnes between 1990 and 1994.[13]

Perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOSF)

Wang et al.[46] collected data on the annual global production of PFOSF by the 3M company (in the U.S. and Europe), manufacturers other than 3M in Europe and Japan, and manufacturers in China for the period 1958–2015. Since PFOSF is used to manufacture PFOS, its salts, and other related polymeric and non-polymeric derivatives, we assume that the annual global production of PFOSF is equal to the sum of annual global productions of all these PFOS-related compounds. Overall, Wang et al.[46] show that the annual global PFOSF production increased from 26 tonnes in 1958 to a peak of 4,368 tonnes in 2000; after 3M ceased its global production in 2002 and shortly after China started to produce PFOSF, the annual global PFOSF production maintained 100 to 200 tonnes between 2003 and 2015. The global cumulative production of PFOSF between 1958 and 2015 amounted to 68,000–68,500 tonnes. We average their yearly data for decadal estimates of production from the 1950s to 2010s, assuming that the average annual global production in 2016–2020 remained the same as the level reported for 2011–2015.

Based on the collected production data, Wang et al.[46] estimated the environmental releases of PFOS, perfluorooctanesulfonamides/-sulfonamido ethanols (C8FOSA/Es), and POSF, stemming from the production, use, and disposal of POSF-related products, as well as the degradation of precursors from multiple lifecycle stages worldwide. They estimated that 3,000–14,300 tonnes of these chemicals were cumulatively released into the environment since 1958, with 2,500–11,800 tonnes released between 1958 and 2002, 480–1,600 tonnes released between 2003 and 2015, and 14–830 tonnes anticipated between 2016 and 2030. Since Wang et al. did not provide detailed information on the geographic distribution of the global environmental release estimates, we adopt the following two assumptions to break the global total into region-specific estimates. First, since the 3M company produced PFOSF in both its U.S. and European manufacturing plants, we assume that 80% of the 3M's production occurred in the U.S. and the rest occurred in Europe, given that the dominant U.S. manufacturing plant in Decatur, AL was reported to possess 80% of the global PFOSF market share.[47] Second, we assume that ~20% of the non-3M production occurred in Japan and the rest occurred in Europe, given the average annual production of 5.5 to 14.7 tonnes in Japan[48] compared to the total non-3M annual production of ~50 tonnes.[46]

Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds

Wang et al.[49] collected data on the annual global production of PFOA, and its ammonium and sodium salts (APFO/NaPFO) in Country Groups I (Japan, Western Europe, and the U.S.) and II (Russia, China, Poland, and India) for the period 1951–2015. Their collected data indicate that the annual global production of these chemicals increased from ~0.01 tonnes in 1951 to a peak of 130–760 tonnes between 2006 and 2010. Specifically, the annual production in Country Group I peaked at 82–502 tonnes between 2006 and 2010 and declined to 51–354 tonnes between 2011 and 2015, and the annual production in Country Group II started in the early 1980s and increased to 76–377 tonnes between 2011 and 2015. The cumulative global production of these chemicals between 1951 and 2015 amounted to 3,175–18,260 tonnes. We average their yearly data for decadal estimates of production from the 1950s to 2010s, assuming that the average annual productions in both Country

Groups I and II in 2016–2020 remained the same as the respective average levels reported for 2011–2015.

Based on the collected production data, Wang et al.[49] estimated the environmental releases of PFOA, its salts, and related compounds from multiple lifecycle stages worldwide. They estimated that 2,078–18,366 tonnes (the respective "lower" and "higher" scenarios; the "plausible" scenarios not discussed here) of these chemicals were cumulatively released into the environment since 1951, with 1,344–8,184 tonnes released between 1951 and 2002, 730–4,773 tonnes released between 2003 and 2015, and 3–5,408 tonnes anticipated between 2016 and 2030. Since Wang et al. did not provide detailed information on the geographic distribution of the global environmental release estimates, we adopt the following two assumptions to break the global total into region-specific estimates, based on incomplete data on the production and market of fluoropolymers available for several discrete years collected by Wang et al.[49] First, we assume that the U.S. and Europe contributed to ~50% and ~30%, respectively, of the total environmental releases occurring in the Country Group I. Second, we assume that China contributed to 40%, 50%, 60%, and 70%, and Russia (the Soviet Union) contributed to 20%, 25%, 30%, and 30%, of the total environmental releases occurring in Country Group II in the 1980s, 1990s, 2000s, and 2010s, respectively, and that Russia (the Soviet Union) contributed to 90% of the total environmental releases occurring in Country Group II before 1980.

Polychlorinated biphenyls (PCBs)

PCBs were first commercialized in 1930. Breivik et al.[50] aggregated data on the annual global production of PCBs from 1930 to 1993. Their results show that a cumulative total of 1,325,810 tonnes of technical PCBs have ever been produced worldwide, with ~50% produced in the U.S., ~45% in European countries (including the Soviet Union and Russia), and 5% in Asian countries. We average their yearly data for decadal estimates of production from the 1930s to 1990s. The estimate of the global cumulative production was believed to be associated with an uncertainty range factor of 1.5.[51]

Based on Breivik et al.'s aggregated data on the annual global production of six major PCB congeners (PCB8, PCB28, PCB52, PCB118, PCB153, and PCB180; accounting for 22% – 24% of the total mass of technical PCBs), Li and Wania[52] simulated long-term time-variant, geographically-resolved environmental releases of these PCB congeners from 1930 to 2100 using substance flow analysis modeling. The model calculated that ~86,000 tonnes of the six PCB congeners (corresponding to 387,000 tonnes of technical PCBs after a scaling) have been released into the environment by 2020; it predicted that another 5,500 tonnes of the six PCB congeners (corresponding to 25,000 tonnes of technical PCBs after a scaling) will be released from the "stock" of in-use industrial or consumer goods and waste stocks during the future period 2021 through 2100. The estimate of the global cumulative environmental releases between 1993 and 2020 was assumed to be associated with an uncertainty factor of 3.5. The annual environmental releases in both Europe (including the Soviet Union) and the U.S. were predicted to peak at ~3,300 tonnes in the 1970s, whereas the annual environmental releases in China were predicted to peak at ~590 tonnes in the 1980s.

It should be mentioned that Breivik et al.[50, 51] introduced the first global dynamic substance flow analysis for PCBs to bridge the gap between production and atmospheric releases. However, their estimates of atmospheric releases were not used here because this work focuses on the total multimedia environmental releases.

Short-chain chlorinated paraffins (SCCPs)

The worldwide production of SCCPs started in the 1930s. Chen et al.[53] aggregated data on the annual global production of chlorinated paraffins from 1930 to 2020 and the relative distribution of the short-, medium-, and long-chain components in the produced chlorinated paraffins. Their results show that a total of 32,500,000 tonnes of chlorinated paraffins were produced during the period, with SCCPs accounting for 28% (8,795,000 tonnes) of the total mass of chlorinated paraffins. The annual global production of SCCPs peaked at 440,000 tonnes in the year 2014 and declined since then. We average their yearly data for decadal estimates of production from the 1930s to 2010s.

Based on the aggregated data, those authors simulated time-variant, geographically-resolved environmental releases of SCCPs from 1930 to 2020 using substance flow analysis modeling. Their results show that the annual environmental releases of SCCPs have been increasing in China, whereas those in Europe and North America peaked in the 1980s and 1990s, respectively, before steady declines. A cumulative total of ~1,501,800 tonnes of SCCPs was released into the global environment as of 2020.

It should be noted that Glüge et al.[54] previously developed estimates of global production and environmental releases of SCCPs as well. However, their estimates were not used here because the estimates represent a minimum scenario (with a probable underestimation by a factor of 10, ref.[55]) and had been revisited and updated by Chen et al.[53]

Technical endosulfan and its related isomers

Endosulfan was first commercialized in 1954. Li et al.[56] reported the historical annual global production between 1954 and 2000, indicating that the annual global production was on average ~130, ~2,400, ~6,000, 10,500, and 12,800 in the 1950s, 1960s, 1970s, 1980s, and 1990s, respectively. In the 2000s and 2010s, the annual production was ~10,000 tonnes in India, 4,000 tonnes in Germany (ceased in 2007), and 2600–5000 tonnes in China.[57, 58] Therefore, the annual global production is estimated to be ~17,000 tonnes in the 2000s and ~13,500 tonnes in the 2010s.

India was the world's largest user of endosulfan, contributing to 33% of the historical global endosulfan use between 1954 and 2000, followed by the U.S. (8%) and Brazil (7%). The Soviet Union consumed 4% of the total historically produced endosulfan worldwide between 1954 and 2000.[56] In 1993, 416 tonnes of endosulfan were used in Western Europe (266 tonnes in France and 150 tonnes in Spain).[59] The annual use in Western Europe was surveyed to be 416 to 1,028 tonnes of endosulfan between 1994 and 1999.[59] In this study, we assume the annual use in Western Europe maintained ~800 tonnes in the 1980s and 1990s. Most European countries stopped the use of endosulfan in the early 2000s.[58] In addition, endosulfan has been widely used in China since 1994,[60] with annual use of on average 2,000, 3,700, and 2,600 tonnes in the 1990s, 2000s, and 2010s, respectively.[60, 61]

Toxaphene

Toxaphene production started in 1946.[62] Li et al.[56] reported the historical annual global use of toxaphene between 1947 and 2000. The total cumulative global use of toxaphene was estimated to be 1.33 million tonnes between 1950 and 1993 and 0.67 million tonnes between 1970 and 1993.[63]

Li[62] collected and curated production and use data in the U.S. The results show that the U.S. produced 720,000 tonnes between 1946 and 1982 and used 491,000 tonnes between 1947 and 1986. The U.S. stopped using toxaphene in 1990. According to survey data, China produced a cumulative total of 20,660 tonnes of toxaphene between 1970 and 1985.[13]

In Europe, a cumulative total of 254,000 tonnes of toxaphene was used in the Soviet Union (and later Russia) between 1952 and 1990,[56] a cumulative total of 26,000 tonnes was used in France between 1952 and 1991,[56] and a cumulative total of 10,000 tonnes of toxaphene was used in Germany during 1960 and 1980 (with the heaviest use in the 1970s).[64] Based on these data, we estimate that the annual use was 5,000, 7,500, 12,000, and 5,000 tonnes in Europe in the 1950s, 1960s, 1970s, and 1980s, respectively. The annual use of toxaphene was estimated to be as low as 10–100 tonnes in the 1990s.[64]

Table S1. Properties of the 25 investigated persistent organic pollutants

Chemical	logK_{OW} (a)	logK_{OA} (a)	HL_{air} (hr) (b)	HL_{water} (hr) (c)	HL_{soil} (hr) (c)	pK_{a} (d)
Aldrin	6.24	8.26	3	30590	61180	–
Chlordane	6.25 (e)	8.83 (e)	105	61370	122740	–
Chlordecone	5.41 (f)	10.07 (f)	127	330370	660740	–
C-decaBDE	9.97	14.78	610	67900	135800	–
C-octaBDE	7.51	12.16	492	23030	46060	–
C-pentaBDE	7.31	10.88	265	8450	16900	–
DDT	6.39	9.72	126	14470	28940	–
Dicofol	6.06	11.29	44	21620	43240	–
Dieldrin	5.48	8.84	77	42560	85120	–
Endrin	4.94	8.28	77	42560	85120	–
Heptachlor	5.94	7.74	3	42110	84220	–
HBB	6.39 (f)	10.15 (f)	657	17480	34960	–
HBCDD	5.77 (g)	10.46 (g)	15	3600	7200	–
HCB	5.64	7.12	9189	14020	28040	–
Mirex	6.89 (f)	8.37 (f)	1259	495290	990580	–
PCPs	5.12 (f)	11.12 (f)	592	6760	13520	4.74 (h)
PFOS	6.27 (i)	4.75 (i)	11702	44851200 (j)	89702400	-1.64
PFOA	2.80	5.12	299	44851200 (j)	89702400	0.34
PCBs	6.50 (k)	8.60 (l)	429	10850	21700	–
SCCPs (m)	6.00 (n)	8.16 (n)	13	10916 (o)	21883 (o)	–
Endosulfan	4.87 (p)	9.09 (p)	25	36750	73500	–
Toxaphene	5.50 (q)	9.25 (q)	3	85910	171820	–
α-HCH	3.88	7.48	637	4390	8780	–
β-HCH	3.91	8.74	637	4390	8780	–
γ-HCH	3.76	7.72	414	2960	5920	–

Notes:

(a) Taken from Li et al.[65] unless indicated otherwise.

(b) Calculated based on the rate constant of reaction with the hydroxyl radical (the geometric mean of values computed by the AOPWIN model in EPI Suite ver. 4.1[66] and OPERA ver. 2.6[67]) and the average atmospheric hydroxyl radical concentration (9.8×10^5 molecules/cm^3 for the northern hemisphere), assuming 12 hours of daylight.

(c) Calculated based on the primary biodegradation probabilities output by the BIOWIN model in EPI Suite ver. 4.1, using empirical relationships by Arnot et al.[68] and then extrapolated to half-lives in water and soil, unless indicated otherwise.

(d) Calculated using OPERA ver. 2.6, unless indicated otherwise.

(e) The geometric mean of *cis*-chlordane and *trans*-chlordane (taken from Li et al.[65])

(f) Experimentally derived data in the training set of EPI Suite ver. 4.1.

(g) The final adjusted value for technical HBCDD.[69]

(h) Experimentally derived data taken from ref.[70].

(i) Experimentally derived data in the training set of OPERA ver. 2.6.

(j) Since PFOS and PFOA are not subject to biodegradation, the photolytic half-life of PFOA in water[71] was used for both PFOS and PFOA in the aquatic environment.

(k) Experimentally derived data for Aroclor® 1254 in the training set of EPI Suite ver. 4.1

(l) Calculated based on K_{OW} and K_{AW} of Aroclor® 1254. The K_{AW} of Aroclor® 1254 was taken from Staudinger and Roberts.[72]

(m) Calculated based on the structural information of a homolog C13Cl6. This homolog was selected as a representative because it is abundant in the technical SCCP mixture and has partitioning coefficients close to the those of the technical SCCP mixture.

(n) Recommended representative value, based on experimentally derived data for SCCP mixtures (chlorinated contents of 55.9% and 68.5%);[73]

(o) Recommended representative value in Krogseth et al.[74]

(p) Average of 70% of the α-endosulfan value and 30% of the β-endosulfan value (taken from Li et al.[65]), based on the respective abundance of these two isomers in technical endosulfan.

(q) Recommended representative value, based on experimentally derived data of technical toxaphene; taken from MacLeod et al.[75]

Table S2. The persistence (overall persistence), bioaccumulation potential (fish bioconcentration factor), toxicity (human toxicological threshold), and long-range transport potential (the fraction of global environmental releases accumulating in surface media of a remote region, ϕ3) of the 25 investigated POPs.

Chemical	Overall persistence (d)	Fish bioconcentration factor (L/kg)(a)	Human toxicological threshold (ng/kg/d)(b)	ϕ3 (unitless)
Aldrin	2,670	5,500	30	1.70×10^{-5}
Chlordane	4,870	22,600	500	5.80×10^{-5}
Chlordecone	9,960	1,500	300	1.10×10^{-3}
C-decaBDE	4,690	251,000(c)	7,000	3.70×10^{-3}
C-octaBDE	2,760	646,000(c)	128(d)	3.10×10^{-3}
C-pentaBDE	1,010	977,000(c)	100(e)	1.10×10^{-3}
DDT	1,710	19,500	500	1.70×10^{-4}
Dicofol	2,570	6,200	400(f)	3.80×10^{-4}
Dieldrin	4,150	8,100	50	5.40×10^{-5}
Endrin	3,310	8,100	300	2.30×10^{-5}
Heptachlor	2,660	8,700	500	9.40×10^{-6}
HBB	2,080	191,000(c)	10,000(g)	1.40×10^{-3}
HBCDD	431	3,700(c)	200,000(h)	1.40×10^{-5}
HCB	1,380	21,400	800	2.60×10^{-3}
Mirex	6,010	20,400	200	1.10×10^{-3}
PCPs	422	460	5,000	1.90×10^{-5}
PFOS	2,610,000	35(i)	2(g)	4.60×10^{-2}
PFOA	2,700,000	3.2(i)	3(g)	4.60×10^{-2}
PCBs	1,180	151,000(j)	37(k)	1.30×10^{-4}
SCCPs	1,140	760(c)	100,000(l)	9.10×10^{-6}
Endosulfan	3,800	1,100(m)	6,000	1.00×10^{-4}
Toxaphene	6,320	1,100	90(n)	9.50×10^{-5}
α-HCH	475	1,100	8,000(g)	1.90×10^{-5}
β-HCH	503	1,100	600(g)	1.60×10^{-5}
γ-HCH	331	470(o)	300	8.60×10^{-6}

Notes:

(a) Experimental data in the training set of the OPEn-QSAR-App (OPERA) model,[67] unless indicated otherwise.

(b) Taken from the U.S. Environmental Protection Agency's Integrated Risk Information System (IRIS), unless indicated otherwise.

(c) Predicted by the OPERA model.

(d) Predicted by the Conditional Toxicity Value Predictor[76] for BDE-183 due to a lack of the experiment-based recommended value.

(e) Value for both BDE-47 and BDE-49.

(f) Taken from the U.S. Environmental Protection Agency Office of Pesticide Programs' database of Human Health Benchmarks for Pesticides.

(g) Value of the minimal risk level (MRL); taken from the U.S. Agency for Toxic Substances and Disease Registry.

(h) Taken from National Research Council (2000).[77]

(i) Experimental data for the whole-organism common carp (*Cyprinus carpio*) recorded in the ECOTOXicology knowledgebase (ECOTOX).

(j) Geometric mean of experimental data for the whole-organism fathead minnows (*Pimephales promelas*) recorded in ECOTOX.

(k) Geometric mean of the RfDs of Aroclor 1254 (20 ng/kg/d) and 1016 (70 ng/kg/d), taken from the IRIS.

(l) Calculated based on the no-observed-adverse-effect-level from European Chemicals Agency (2008),[73] assuming a safety factor of 100.

(m) Geometric mean of experimental data for the whole-organism sheepshead minnow (*Cyprinodon variegatus*) recorded in ECOTOX.

(n) The provisional chronic reference dose from the U.S. Provisional Peer-Reviewed Toxicity Value (PPRTV) database.

(o) Geometric mean of experimental data for the sheepshead minnow (*Cyprinodon variegatus*) (response site was not reported) recorded in ECOTOX.

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