Japan PFAS Situation Report

Japan Endocrine Disruption Prevention Action
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Summary

This study examined the country situation for per- and polyfluoroalkyl substances (PFAS) – a large class of substances of increasing global concern. The key findings of this report are:

- PFAS substances are poorly regulated in Japan
- Women and infants are contaminated with PFAS substances
- PFAS substances impact Japanese infants
- Breast milk is contaminated with PFAS substances
- PFAS water pollution is widespread
- Wastewater treatment plants are PFAS pollution sources
- Marine and terrestrial organisms are contaminated with PFAS
- Firefighting foams are a likely major source of PFAS pollution
- Dust is contaminated with PFAS substances
- PFAS contaminates consumer products
- US military bases cause PFAS pollution
- Japan is an important PFAS manufacturer
- PFAS elimination contributes to achievement of the Sustainable Development Goals

PFAS substances are poorly regulated in Japan

Most PFAS substances are not regulated in Japan, but there some regulations partially govern PFOA, PFHxA, PFHxS, and PFOS. The Stockholm Convention listing of PFOS went into legal force in Japan in 2010, but the country has registered the following time-unlimited acceptable purposes: photo-imaging; photo-resistant and anti-reflective coatings for semi-conductors; etching agent for compound semi-conductors and ceramic filters; and certain medical devices. No maximum levels for PFOS, PFOA or any other PFAS substance are set for tap water in Japan. There are no regulations regarding the use of firefighting foam extinguishing agents containing PFOS or any other PFAS substance at the firefighting site. PFOA, PFHxA and PFHxS are categorized as "General Chemical Substances" under the Chemical Substances Control Law which requires those who manufacture or import more than 1 ton of them to notify their amount to the Ministry of Economy, Trade and Industry. Under its Status of Forces Agreement with the US, Japan cannot enforce any regulations on polluting US military bases such as the bases in Okinawa.

Women and infants are contaminated with PFAS substances

A 2018 study found PFHxS, PFOS, PFOA, PFNA, PFDA, PFUnDA, PFDODA, and PFTrDA in pregnant women. The highest levels were found for PFOS (30.28 ppb), PFOA (24.88 ppb), and PFNA (13.19 ppb). This confirmed an earlier study that found PFOA, PFNA, PFDA, and PFUnDA, and PFOS in 100% of the pregnant women that were tested in 2003, 2005, 2007, and 2009. In 2011, significant detection frequencies of other PFAS were detected including PFHxA (20%), PFHpA (50%), PFDoDA (97%), PFTrDA (97%), PFTeDA (13%), and PFHxS (77%). Between 2003 and 2011, plasma concentrations of PFOS and PFOA decreased, but PFNA and

PFDA concentrations increased by 4.7% and 2.4% per year respectively. In 2004, the first study showing PFAS contamination of fetal cord blood found PFOS in all infants tested. Since then, cord blood has been tested for impacts of prenatal exposure as described in the next section.

PFAS substances impact Japanese infants

A 2018 Japanese government review based on studies of a large cohort of pregnant women and their infants found that prenatal exposures to PFAS, such as PFOS and PFOA, may affect birth size, disrupt the homeostasis of several hormones (thyroid, steroid, sex hormones et al.), and affect the development of the nervous system, allergies, and infectious diseases. In males, there are negative correlations between prenatal exposure of PFAS and production of testosterone, estradiol, inhibin B, and insulin-like factor 3. In females, there are negative correlations between prenatal exposures to PFAS and production of progesterone, prolactin, and sex hormone-binding globulin (SHBG). A study of this same cohort noted that, "even low levels of PFOS and PFOA exposure can disrupt reproductive hormone imbalance in the fetus." A 2017 study found prenatal exposures to PFHxS and PFOS through the mother, were associated with a higher risk of infectious diseases in their children.

Breast milk is contaminated with PFAS substances

A 2008 study found PFAS in breast milk in women from Ehime Prefecture. PFOS was found in 100% of the samples from Japan. PFOA and PFHxS were found in 92% of the samples; PFNA in 13% of them; and PFHpA in 25% of them. The results showed significant PFAS levels for PFOS, PFOA, PFHxS, PFNA, and PFHpA. Overall, average PFOS levels in Japanese breast milk averaged 232 ppt – more than 10 times higher than the drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US State of Vermont. Average levels of PFOS in Japanese breast milk were the highest in all countries examined. The highest level of PFOS in Japanese breast milk (523 ppt) was more than 26 times higher than this drinking water health advisory limit.

PFAS water pollution is widespread

PFOS pollution has been found in 64 rivers and PFAS pollution is widely present in coastal areas including the Sea of Kushiro, Mutsu Bay, Hachinohe Bay, Jyodoga Coast, Miyako Bay, Kamaishi Bay, Honjyo Marina, Souma Bay, Nagasaki Bay, Chiba-Funahashi Bay, Yamashita Bay, Pacific Ocean, Nagoya Bay, Koshien Bay, Motoujina Shima coast, and Hakata Bay. A subsequent study of 18 rivers in Hokkaido, Tohoku, Kanto, Hokuriku, Chubu, Kinki, Chugoku, Shikoku, and Kyushu found PFAS in all of them and more than half of them had levels higher than 40 ppt. Predominant PFAS were PFOS, PFHpA, PFOA, and PFNA. In Hokkaido rivers, PFOS and/or PFHxS were detected around airports, probably due to the use of fire extinguishers. PFAS contamination from industrial facilities was also detected in paper mill effluents. These compounds are used in surface treatment of paper products.

In Tokyo Bay, 11 PFAS substances were found in pore water including PFHxS, PFOS, TH-PFOS, N-EtFOSAA, PFBA, PFPeA, PFHxS, PFHpA, PFOA, PFNA, and PFDA. Freshwater inputs are the likely source of these substances. A spatial survey of PFAS pollution in Tokyo Bay showed that PFNA was the most prevalent PFAS substance and that levels of PFHxA, PFHpA, PFOA and PFNA were 10 – 100-fold higher near production plants than in other areas. PFCA levels in coastal water (491 ng/L) and plant effluent (6024 ng/L) were described as

"extraordinarily high." Estimated loadings from the six major rivers into Tokyo Bay were 96.6 kg/year for PFOA and 139.6 kg/year for PFOS. The authors conclude that, "the introduction of PFC [PFAS] regulations on the use/production/emission has not reduced PFC pollution."

A time study of PFAS in Tokyo Bay sediment showed a rapid increase after the early 1970s for PFUnDA, PFDoDA, PFTrDA; gradual increase from the 1950s for PFNA and PFDA; an unexpected peak in the later 1950s – 1960s and rapid increase after the late 1990s for PFOA; gradual increase from the 1950s and gradual decrease after the early 1990s for PFOS; and an unexpected peak in the late 1990s for precursors of PFOS such as N-EtFOSAA and N-MeFO-SAA. The authors note that this might represent releases from firefighting foams or discharge from manufacturing facilities. In Osaka Bay, increasing levels of PFHxA were observed due to pollution from the Kanzaki River.

PFAS substances have been found in all Tokyo groundwater samples and PFOS (0.28-133 ng/L), PFHpA (<0.1-20 ng/L), PFOA (0.47-60 ng/L), and PFNA (0.1-94 ng/L) were the predominant substances. The authors note that, "groundwater contamination by PFOS will continue unless the precursors are regulated." In addition, they observe that, "since manufacturers are switching to shorter-chain PFCAs with <7 fluorinated carbons, these compounds could also contaminate groundwater." Another study found widespread PFAS pollution in Tokyo groundwater, springs, and confined aquifers with PFHxS, PFOS, PFHpA, PFOA, PFNA, and PFDA the most prevalent substances.

Sampling in the Iruna River upstream of the intake for Tokyo's drinking water treatment plants found PFHxA, PFOS, PFHpA, PFOA, PFNA, PFDA, PFUA, and PFDoDA in all samples. PFAS substances are also found in tap water. In Osaka, PFOS (0.20 – 22 ng/L) and PFOA (2.3 – 84 ng/L) contaminated all tap water samples. The highest levels of PFOS and PFOA in tap water exceeded drinking water limits set in some US states. For example, Vermont sets a drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined. In Tokyo tap water, total PFAS levels ranged from 0.72 to 95 ng/L and 12 PFAS substances were detected in the samples with the highest concentrations.

PFOS and PFOA have both been found in sediment cores from the Suruga Bay at depths of 800 – 850 meters. The authors note that the finding of PFAS in the deep-sea environment could be due to the coastal waters and "garbage such as plastic products, waste cans, and gum products are often observed in the deep-sea environment, and perfluoro organic compounds are considered to be eluted in this garbage."

Wastewater treatment plants are PFAS pollution sources

Wastewater treatment plants receive a mixture of domestic and industrial wastewater and the PFAS removal rate is less than 50% but increases with frequent changes of activated carbon. A study of the processes found that sand filtration and ozonation did not remove PFOS or PFOA and that PFOS (0.51 – 7.6 ng/L) and PFOA (0.78 – 72 ng/L) were also found in activated carbon filter samples. The authors noted that older activated carbon was not effective at PFOS or PFOA removal. This raised concerns since activated carbon at water treatment plants is commonly used for several years At some plants, the effluent has even higher PFAS levels than the influent and they drain into rivers that serve as drinking water supplies. A 2010 study estimated daily

discharges of PFAS from individual sewage treatment plants to range from 0.35–55.9 g/d. A sewage treatment plant discharge site on the Ina River near Osaka led to a level of 67,000 ng/L PFOA with an estimated 18 kg of PFOA discharged daily. Another plant discharging into the Tsurumi River released PFOS (78.7-689.9 ng/L), PFHxA (3.5 – 9.4 ng/L), PFHpA (5.5 – 7.2 ng/L), PFOA (17.8 – 24.9), PFNA (27.5 – 41.8), and PFDA (3.0 – 4.5 ng/L). Sampling at discharge sites in other rivers indicated that sewage effluent was the likely source of PFOS, PFHpA and PFNA. The total fluxes of sewage-derived PFAS were estimated to be: PFOS (3.6 t/year), PFHpA (2.6 t/year), PFOA (5.6 t/year), and PFNA (2.6 t/year). The authors note that these fluxes, especially for PFNA were remarkably high in Japan compared to rivers in Europe.

Marine and terrestrial organisms are contaminated with PFAS

A variety of PFAS substances contaminate marine organisms such as bream, carp, crab, filefish, flounder, jack mackerel, lugworm, medaka, mudskipper fish, mussel, oyster, porpoises, sandfish, sardines, sea bass, shark, and trout. Birds and both wild and domestic terrestrial organisms contaminated with PFAS substances include: brown hawk owl, carrion crow, cattle, cattle egret, chicken, common kestrel, cormorant, dog, eagle, Eurasian sparrowhawk, goat, great egret, gull, horse, Japanese sparrowhawk, large-bill crow, mallard duck, northern goshawk, pig, pintail duck, raccoon dog, swan, turtle, Ural owl and wild rats.

Firefighting foams are a likely major source of PFAS pollution

There are large historical stockpiles of PFOS-containing firefighting foams in Japan, along with current use of PFAS-containing foams. A 2017 research paper reports that there were 15,000 tons of stockpiles of PFOS-containing aqueous film-forming foam in 2012 and 47% of the total amount (7000 tons) was due to stockpiles in car-parking facilities. According to the government, there are more than 20,000 multi-story car-parking facilities in Japan and each one stocks 400 kg of PFAS-containing firefighting foam on average. The second largest PFOS-containing foam stockpile (4200 tons) was held by the petrochemical industry and the third largest was contained in portable fire extinguishers (2000 tons). PFAS contamination likely due to firefighting foams has also been found in the Ina River downstream from the Osaka International Airport. There are no regulations regarding the use of firefighting foam extinguishing agents containing PFOS or any other PFAS substance at the firefighting site.

Dust is contaminated with PFAS substances

High levels of PFOS (15 – 2500 ng/g) and PFOA (69 – 3700 ng/g) were found in all samples of house dust. The authors note that "small children spend their life in the space near the floor, and the compounds have been used in floor wax and carpets." PFAS precursor substances such as polyfluorinated phosphate esters (PAPs) were frequently detected in household dust samples from Canada, the Faroe Islands, Sweden, Greece, Spain, Nepal, Japan, and Australia. The median levels of monoPAPs and diPAPs ranged from 3.7 ng/g to 1,023 ng/g and 3.6 ng/g to 692 ng/g, respectively, with the highest levels in Japan. Dust from Japan had the highest level of PFCAs (230 ng/g) and unlike other countries, PFNA was the predominating substance instead of PFOA. PFOS, PFOA, PFNA, PFDA, and PFUA have also been found in street dust in Tokyo,

PFAS contaminates consumer products

A 2015 study found PFAS in sprays for textiles, car wash / coating products, and rust inhibitor products. The highest PFAS concentration was 25,000 ng/g for N-ethyl perfluorooctane

sulfonamidoethanol (EtFOSE), a PFOS precursor, in a spray for textiles. Sprays for textiles and car wash / coating products were the categories that contained PFAS levels greater than 1000 ng/g. PFAS were detected in two out of three rust inhibitor products, including PFPeA. The PFAS substance with the highest detection frequency was PFNA (16%). PFNA also had the second highest average concentration of 220 ng/g. The authors suggest that PFNA may have been substituted for PFOA in Japan.

US military bases cause PFAS pollution

Surveys by the Okinawa Prefectural Enterprise Bureau since 2013 showed that Kadena Air Base and Marine Corps Air Station at Futenma are major sources of PFAS pollution that have contaminated water outside the base. Internal US government documents reveal large spills and leaks of firefighting foams and US military officials have admitted using PFOS-containing firefighting foams at the base, after the substances were detected in groundwater wells and streams running through the base. Internal US government documents described in a 2019 report indicate extreme PFAS contamination inside and outside of the Kadena Air Base. For example, foam fire extinguisher located around a pond situated 200 meters from the Kadena town hall contained 90,000 ppt. A map shows the growing number of polluted sites. Although the Japanese government has requested the US military to allow access for investigation, the US has continued to refuse the requests saying that, "PFOS is not a regulated substance in the US and Japan."

Japan is an important PFAS manufacturer

PFAS manufacturing began in 1947 in Japan and between 1988 and 2017, production nearly doubled, rising from 16,398 tons to 30,151 tons. Three prominent manufacturers include AGC, Daikin, and Mitsui-Chemours.

In Japan, Mitsui-Dupont polluted water with PFOA at their factory in Shimizu. DuPont tracked PFOA in the blood of the workers at the facility as revealed in US EPA filings and internal company documents. PFOA was also found in water at 10 wells at the factory site with levels up to 1,540,000 ppt. The site was sold to DuPont spin-off company, Chemours and Chemours-Mitsui Fluoroproducts is a 50:50 joint venture between the two companies. In 2019, US EPA filed a notice of violation against Chemours – its first under the new Toxic Substances Control Act. The company had five serious violations including manufacturing a substance for commercial purposes that was not even on the regulator's inventory list. In China,

Daikin is located in the Changshu Industrial Park in Jiangsu Province along with other foreign PFAS manufacturers such as Solvay and Arkema. In 2013, scientists measured some of the highest PFAS levels ever reported in China in the industrial park. In 2016, the US Northern Alabama water and sewer authority sued 3M and Daikin America due to pollution of the Tennessee River drinking water supply with PFOS and PFOA. The area is downstream of Daikin's factory in Decatur, Alabama. Local residents were warned by authorities not to drink the water. Daikin settled their part of the lawsuit for USD$4 million in 2018 to pay for a carbon filtration system. However, the settlement does not prevent individual residents from pursuing legal action.

In 2018, Kevin Hardwick, a US firefighter, filed a national class-action lawsuit against nine PFAS manufacturers including AGC and Daikin, along with 3M, Archroma, Arkema, Chemours, Dyneon, DowDuPont, and Solvay. The purpose of the lawsuit is to fund an independent investigation of links between PFAS exposure and health impacts, including the entire class of PFAS substances.

PFAS elimination contributes to achievement of the Sustainable Development Goals (SDGs)

Actions to control and phase-out PFAS as a class contribute to achievement of several key Sustainable Development Goals (SDGs) due to the impacts of the substances on health and ecosystems including water pollution. These include SDGs 3, 6, 9, 12, 14, 15, and 16.

What are per- and polyfluoroalkyl substances (PFAS)?

PFAS is a large class of more than 4,500 persistent fluorinated chemicals. PFAS are both hydrophobic and lipophobic in nature and extremely persistent due to the strength of the carbon-fluorine bond. They are widely distributed in the global environment due to their high solubility in water, low/moderate sorption to soils and sediments and resistance to biological and chemical degradation. The properties of PFAS have resulted in extensive use as surfactants and surface-active agents in products. Two widely-used members of this class have been perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA). As these two substances have come under regulatory pressure, the industry has shifted to other PFAS with similar properties.

Human exposure to PFAS is mainly by ingestion of contaminated food or water. These substances bind to proteins (not to fats) and persist in the body where they are mainly detected in blood, liver and kidneys. Studies indicate that PFOA and PFOS can cause reproductive and developmental, liver and kidney, and immunological effects in laboratory animals. Both chemicals cause tumors in animal studies along with a variety of other effects on infant birth weight, growth, learning, infant behavior, pregnancy, endocrine system, increased cholesterol, and thyroid function. Recent studies have linked a variety of PFAS substances to many human health effects: cardiovascular disease, markers of asthma, damage to semen quality, ovarian insufficiency, altered glucose metabolism, lower testosterone levels in male adolescents, association with shorter birth length in girls, elevated blood pressure, abnormal menstruation, lower birth weight in infants, possible increased risk of female infertility due to endometriosis, and decreased lung function in children with asthma.

The manufacture and use of PFAS and their use in a multitude of products has caused widespread pollution. PFAS are found in wildlife, accumulating in the blood, liver and kidneys of wildlife such as dolphins, polar bears, seals, birds, fish, and other marine wildlife. PFAS substitutes for PFOS and PFOA have been identified as potential global surface water contaminants and they have been found in more than 80% of 30 surface seawater samples from the North Pacific to Arctic Ocean. PFAS use in firefighting foams at military bases and airports is responsible for water pollution and contaminated communities in many countries, including Australia, Canada, China, Germany, Italy, Japan, Netherlands, New Zealand, South Korea, and Sweden.

Safer cost competitive non-fluorinated alternatives for PFAS use in firefighting foams have been adopted by an increasing number of major airports, including Auckland, Copenhagen, Dubai, Dortmund, Stuttgart, London Heathrow, Manchester, and all 27 major airports in Australia. Increasing awareness about the negative characteristics of PFAS has driven efforts to identify and market safer substitutes for other uses. Increasing awareness about the negative characteristics of PFAS has driven efforts to identify and market safer substitutes for other uses. Due to the complexity and negative characteristics of PFAS, there is increasing interest in regulating PFAS as a class rather than as individual substances.

PFOS

PFOS and its related substances have been used in a variety of products and processes including firefighting foams, carpets, leather goods, upholstery, packaging, industrial and household cleaning products, pesticides, photographic applications, semiconductor manufacturing, hydraulic fluids, catheters and metal plating. PFOS is extremely persistent and has shown no degradation under any environmental condition that has been tested. It is toxic to mammals and high concentrations have been found in Arctic animals, far from anthropogenic sources. PFOS is regularly detected in human blood and breast milk. For example, in one study of 299 infants, PFOS was found in the blood of 297 of them and PFOA was found in all of them.

PFOA

PFOA has been used to make non-stick pans, and is found in textiles, fire-fighting foams, and medical devices, and is used in many other products and processes. In 2017, the Stockholm Convention POPs Review Committee noted the link between PFOA and serious illnesses in humans, including diagnosed high cholesterol, ulcerative colitis, thyroid disease, testicular cancer, kidney cancer and pregnancy-induced hypertension. PFOA has contaminated the global environment, including wildlife and people of remote regions such as the Arctic and Antarctic.

For more information about recent research on the impacts of PFAS, including fluorinated substitutes for PFOS and PFOA, please see Annex 1. Information about the high cost of PFAS pollution cleanup is available in Annex 2. Global regulation of PFAS through the Stockholm Convention and evaluations of its expert committee is discussed in Annex 3.

Actions on PFAS and the Sustainable Development Goals

Actions to control and phase-out PFAS as a class contribute to achievement of several key Sustainable Development Goals (SDGs) due to the impacts of the substances on health and ecosystems including water pollution. These include

Sustainable Development Goal 3: Ensure healthy lives and promote well-being for all at all ages. Targets under SDG3 include:
3.4: *"reduce by one third premature mortality from non-communicable diseases through prevention and treatment and promote mental health and well-being"*
3.9: *"substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination."*

Sustainable Development Goal 6: Ensure availability and sustainable management of water and sanitation for all. Targets under SDG6 include:
6.3: *"improve water quality by reducing pollution, eliminating dumping and minimizing release of hazardous chemicals and materials, halving the proportion of untreated wastewater and substantially increasing recycling and safe reuse globally."*

Sustainable Development Goal 9: Build resilient infrastructure, promote inclusive and sustainable industrialization and foster innovation. Targets under SDG9 include:
9.4: *"greater adoption of clean and environmentally sound technologies and industrial processes."*

Sustainable Development Goal 12: Ensure sustainable consumption and production patterns. Targets under SDG12 include:
12.4: *"By 2020, achieve the environmentally sound management of chemicals and all wastes throughout their life cycle, in accordance with agreed international frame works, and significantly reduce their release to air, water and soil in order to minimize their adverse impacts on human health and the environment."*
12.5: *"substantially reduce waste generation through prevention, reduction, recycling and reuse."*
12.6: *"Encourage companies, especially large and transnational companies, to adopt sustainable practices and to integrate sustainability information into their reporting cycle."*
12.7: *"Promote public procurement practices that are sustainable, in accordance with national policies and priorities."*

Sustainable Development Goal 14: Conserve and sustainably use the oceans, seas and marine resources for sustainable development. Targets under SDG14 include:
14.1: *"By 2025, prevent and significantly reduce marine pollution of all kinds, in particular from land-based activities, including marine debris and nutrient pollution."*

Sustainable Development Goal 15: Protect, restore and promote sustainable use of terrestrial ecosystems, sustainably manage forests, combat desertification, and halt and reverse land degradation and halt biodiversity loss. Targets under SDG15 include:
15.1: *"By 2020, ensure the conservation, restoration and sustainable use of terrestrial and inland freshwater ecosystems and their services, in particular forests, wetlands, mountains and drylands, in line with obligations under international agreements."*
15.5: *"Take urgent and significant action to reduce the degradation of natural habitats, halt the loss of biodiversity and, by 2020, protect and prevent the extinction of threatened species."*
15.9: *"By 2020, integrate ecosystem and biodiversity values into national and local planning, development processes, poverty reduction strategies and accounts."*

Sustainable Development Goal 16: Promote peaceful and inclusive societies for sustainable development, provide access to justice for all and build effective, accountable and inclusive institutions at all levels. Targets under SDG16 include:
16.7: *"Ensure responsive, inclusive, participatory and representative decision-making at all levels."*

16.10: *"Ensure public access to information…"*

PFAS production, use, and waste management in Japan

Industry information shows increases in production, import and export

Industry information on PFAS production and import is available from the Japan Fluoropolymers Industry Association (JFIA). JFIA is an association of 22 regular members (users of fluoropolymers), 4 affiliate members (manufacturers of fluoropolymers) and 24 supporting members (other fluoropolymers-related industries).[1]

Between 1988 and 2017, production of PFAS nearly doubled in Japan, rising from 16,398 tons to 30,151 tons. There was also a large increase in "domestic demand" from 10,464 tons in 1988 to 17,930 tons in 2017 a 1.7-fold increase. Exports increased three-fold from 1988 reaching 22,030 tons in 2017. Over this same period, imports increased 6-fold from 1,643 tons to 10,596 tons.

Trend of Demand and Supply of Materials for Fluorocarbon Resin
Ministry of Economy Trade and Industry Current Production Statistics & Ministry of Finance Customs Statistics compiled by JFIA[2]

	Production in Japan		Shipment in Japan		Import		Export		Domestic Demand	
FY	Volume (Ton)	Comparison with the previous year	Volume (Ton)	Value (1 mil. Yen)	Volume (Ton)	Value (1 mil. Yen)	Volume (Ton)	Value (1 mil. Yen)	Volume (Ton)	Value (1 mil. Yen)
1956	---	---	---	---	12	57				
57	---	---	---	---	23	105				
58	11	---	7	69	37	156				
59	38	345.5	29	173	52	206				
60	79	207.9	62	306	99	350				
61	104	131.6	96	412	59	199				
62	120	115.4	104	455	85	325				
63	160	133.3	150	629	100	304				
64	267	166.9	196	824	212	614				
65	290	108.6	236	870	83	336				
66	608	209.7	572	1,815	99	391				

[1] The member list is available at http://www.jfia.gr.jp/members/member.htm#kyou.
[2] http://www.jfia.gr.jp/jfiadata/jyukyu.htm translated by Japan Endocrine Disruption Prevention Action

67	984	161.8	961	2,387	119	483				
68	861	87.5	769	2,244	127	643				
69	1,143	132.8	1,078	3,581	164	753				
70	1,403	122.7	1,202	3,940	353	1,467				
71	1,099	78.3	1,161	3,614	269	1,083				
'72	1,263	114.9	1,286	3,880	349	1,121				
'73	1,835	145.3	1,883	5,624	630	1,903				
'74	1,737	94.7	1,597	4,792	893	2,974				
'75	1,171	67.4	1,145	3,736	565	1,884				
'76	2,418	206.5	2,116	6,911	550	2,349				
'77	2,797	115.7	2,550	8,095	694	3,032				
'78	3,084	110.3	2,969	9,268	615	2,258				
'79	3,983	129.1	3,883	12,128	871	3,093				
'80	4,552	114.3	4,454	15,605	844	3,101				
'81	5,227	114.8	5,027	18,017	1,057	3,677				
'82	5,481	104.9	5,748	20,010	1,074	3,637				
'83	6,239	113.8	6,489	20,467	1,031	3,725				
'84	10,320	165.4	10,385	31,772	1,432	5,104				
'85	12,915	125.1	12,527	37,902	1,604	5,995				
'86	12,677	98.6	12,397	33,593	1,431	4,969				
'87	13,691	108.4	13,804	35,223	1,368	3,950				
'88	16,398	119.8	16,095	41,132	1,643	4,188	7,092	10,464	10,646	34,856
'89	16,769	102.3	16,157	43,877	1,913	5,084	6,675	10,643	11,286	38,280
'90	16,065	95.8	15,774	41,131	2,018	5,638	6,422	12,136	11,370	34,633
'91	15,832	98.5	15,654	41,395	2,032	5,221	6,814	11,687	10,882	34,929
'92	16,505	104.3	16,844	42,324	1,685	4,324	8,953	13,990	9,571	32,658
'93	15,916	96.4	16,203	40,051	1,743	4,269	8,651	12,694	9,295	31,826
'94	16,627	104.5	17,616	42,531	2,175	5,215	9,324	13,681	10,467	34,065
'95	19,487	117.2	19,403	47,850	2,671	6,082	9,498	15,648	12,576	38,283
'96	20,360	104.5	20,447	54,888	2,956	6,937	10,138	20,294	13,264	41,466

'97	21,826	107.2	20,568	55,457	3,598	8,829	11,526	23,399	12,640	40,887
'98	21,958	100.6	21,486	58,907	3,051	8,018	14,134	30,076	10,403	36,849
'99	22,093	100.6	22,426	55,683	2,409	4,509	14,303	26,975	10,532	33,217
2000	26,408	119.5	26,184	61,021	3,481	6,101	15,438	29,439	14,227	37.638
'01	24,175	91.5	21,862	51,716	2,724	5,268	13,733	28,321	10,853	28,663
'02	21,399	93.5	20,937	47,398	2,520	5,281	13,096	24,533	10,361	28,146
'03	22,894	107	22,360	49,322	2,986	5,974	14,370	26,251	10,976	29,045
'04	21,945	95.9	23,352	56,235	6,413	10,508	13,157	26,719	16,608	40,024
'05	25,779	117.5	24,487	59,977	5,524	10,463	14,847	30,148	15,164	40,292
'06	27,779	107.8	27,389	69,172	5,747	12,230	16,393	36,038	16,743	45,364
'07	28,498	102.6	27,146	68,230	6,768	14,559	17,165	38,314	16,749	44,475
'08	29,276	102.7	28,267	68,127	7,616	14,725	18,810	38,848	17,073	44,004
'09	14,687	50.2	16,817	37,217	3,933	6,836	12,247	21,646	8,503	22,407
'10	28,173	191.8	27,430	63,762	7,911	13,739	19,296	33,716	16,045	43,784
'11	29,046	103.1	27,856	68,872	8,749	17,099	19,957	37,782	16,647	48,189
'12	27,233	93.8	25,463	61,690	6,326	13,663	19,034	37,222	12,755	38,131
'13	25,234	92.7	26,240	66,597	6,402	13,081	19,602	42,765	13,040	36,914
'14	29,201	115.7	28,102	73,060	6,343	14,212	21,550	50,464	12,895	36,808
'15	27,610	94.6	26,552	72,704	7,581	16,932	20,672	53,844	13,461	35,791
'16	28,374	102.8	27,648	73,708	8,433	16,282	20,601	49,066	15,480	40,924
'17	30,151	106.3	29,363	81,508	10,596	20,742	22,030	53,864	17,930	48,386

Note: Before 1988, export of fluoropolymers was listed as a part of export of polymers.

Use and stockpiles of PFAS-containing firefighting foams

There are large historical stockpiles of PFOS-containing firefighting foams in Japan, along with current use of PFAS-containing foams.

For extinguishers, extinguishing agents for fire extinguishers and foam extinguishing agents that are produced using PFOS or its salts (hereafter referred to as ‘foam extinguishing agents containing PFOS’), alternative chemicals already exist, and it is unlikely that foam extinguishing agents containing PFOS will be manufactured or imported in the future. However, large amounts have already been distributed nationwide. According to the survey conducted by the relevant ministries, in March 2016, approximately 17 tons (amount of PFOS or its salts contained) of the foam extinguishing agents containing PFOS were identified.

The Japanese government claims that it is extremely difficult to replace these 17 tons of foam extinguishing agents containing PFOS with alternatives in the short-term, saying that the amounts are large and they have already been distributed nationwide. Under the current regulations, the technical standards and labeling matters at the time of transfer were prepared based on the Chemical Substances Control Law. While the government states that standards based on the Fire Defense Law have been established to prevent leakage of the foam extinguishing agents containing PFOS to the exterior at the time of inspection of fire defense equipment, there are no regulations regarding the use of foam extinguishing agents containing PFOS at the firefighting site. This runs counter to the industry's own recommendations for these products (which recommend containment) and indicates that PFOS and PFAS pollution at these sites is a certainty.

A recent research paper illustrates a pattern of PFOS-containing firefighting foam stockpiles that is quite different from other countries. The study reports that there were 15,000 tons of stockpiles of PFOS-containing aqueous film-forming foam in 2012 and 47% of the total amount derived from stockpiles in car-parking facilities.[3] The second and third highest stockpiles were in petrochemical factories and portable extinguishers. Japan Endocrine Disruption Prevention Action could not find out the reason why there are such huge discrepancies between the figures in the government survey and the figures in this research study. The paper notes that PFOS-containing firefighting foam was produced in Japan until 2007 – four years after 3M stopped producing it in the US.

Estimated PFOS-containing firefighting foam stockpiles in Japan in 2012 (total = 15,000 tons)

Entity	Stockpile (metric tons)	% of Total
Car parking facilities	7,000	47
Petrochemical factories	4,200	28
Portable extinguishers	2,000	13
Self-defense force	1,300	9
Airports	160	1
Gas stations	60	0.4
Others	210	1

Data from Zushi Y, Yamamoto A, Tsunemi K, Masunaga S (2017) Revaluation of stockpile amount of PFOS-containing aqueous film-forming foam in Japan: gaps and pitfalls in the stockpile survey, Environ Sci Pollut Res 24:6736-6745

Japanese fire defense law requires car facilities over 500 m^2 (without multi-stories) or those higher than two stories with 200 m^2 per level or more or parking facilities on the roofs of multi-story structures of 300 m^2 or more to have a fire extinguishing system. The government also has estimated that the probability of a fire at one of these facilities is 5.8×10^{-7}. Suppliers have sold high-priced fluorinated aqueous film forming foams to parking facilities to comply with the

[3] Revaluation of stockpile amount of PFOS-containing aqueous film-forming foam in Japan: gaps and pitfalls in the stockpile survey. https://www.ncbi.nlm.nih.gov/pubmed/28091990

federal law. According to the government, there are more than 20,000 multi-story car-parking facilities in Japan and each one stocks 400 kg of PFAS-containing firefighting foam on average.

In 2012, this resulted in car-parking facilities holding approximately 7000 tons (46.7% of the total inventory) of PFOS-containing firefighting foam – larger than the petrochemical industry which contained 4,200 tons. The third-highest stockpile was in portable fire extinguishers (2,000 tons). The authors estimate that the total stockpile of PFOS-containing firefighting foams should decrease to 3,200 tons by 2019 – with 87.5% of the total in car-parking facilities. The authors estimate that if there is no "efficient reduction" in the PFOS-containing firefighting foam stockpiles at car parking facilities, they will not be depleted until 2024.

The total stockpile in Japan of 15,000 tons of PFOS-containing firefighting foam in 2012 is much higher than in other countries. For example, at its peak, the entire EU had a PFOS-containing firefighting stockpile of 122 tons – and the total population is significantly bigger than that of Japan. The authors suggest that perhaps car parking facilities were not counted in the inventory in the EU – but even if they represent the same contribution to the inventory as in Japan, the total stockpile would still be far less.

Government information on PFOS

There is government information on PFOS or its salts, and perfluorooctane sulfonyl fluoride (PFOSF) contained in the country's National Implementation Plan.[4] PFOS or its salts, and PFOSF as their precursor were used as water/oil repellents and surface acting agents. They were designated as a Class I Specified Chemical Substance in April 2010 under the Chemical Substances Control Law (hereinafter referred to as "CSCL", and their manufacture, import and use are virtually prohibited. However, PFOS or its salts are still approved for following three uses including 1) the manufacture of the etching agent for the piezoelectric ceramic filter or composite semiconductor for high frequency band, 2) the photosensitive film of semiconductors, and 3) photographic film for industrial use.

Japan registered a number of "acceptable purposes" for PFOS under the Stockholm Convention listing. These include photo-imaging, photo-resistant and anti-reflective coatings for semiconductors, etching agent for compound semi-conductors and ceramic filters, and certain medical devices.

According to the survey conducted in FY2011 by the government, approximately 1.5 tons (approximately 30 kg in PFOS equivalent) of PFOS or its salts in stock were identified for use in the etching agent and photosensitive film of semiconductors. By FY2015, it was reported that all identified PFOS or its salts were disposed by business entities and no stock is remaining.

[4] The National Implementation Plan of Japan under the Stockholm Convention on Persistent Organic Pollutants Modified in October 2016 https://www.env.go.jp/chemi/pops/plan/en_full-161006.pdf

Information about PFOA

Three manufacturers in Japan; AGC, Daikin Industries, Ltd., and Mitsui-DuPont Fluoroproducts Co., Ltd announced in August 2014 that they joined US EPA's 2010/2015 PFOA Stewardship Program and completely stopped use of PFOA for fluoropolymers products by the end of 2013.

Daikin announced that they ceased manufacture and use of PFOA and related substances, and manufacture of products which use PFOA and related substances as a raw material by the end of 2015.[5] Daikin states that they have developed new C6 fluorinated telomer products including PFHxA to substitute for PFOA.

AGC also announced that the company introduced new products with the perfluoroalkyl group having 6 or less carbon atoms, in response to the Stewardship Program, so that they do not produce PFOA or long-chain PFCAs even by decomposition.[6]

However, Daikin requested an exemption under the Stockholm Convention for use of PFOI, one of the PFOA-related substances, for reprocessing into PFOB for production of pharmaceuticals.[7]

PFAS notification requirements

PFOA, PFHxA and PFHxS are categorized as "General Chemical Substances" under CSCL. Since 2011, those who manufacture or import more than 1 ton of General Chemical Substances must notify their amount to the Ministry of Economy, Trade and Industry. The data collected before 2010 was based on a survey conducted by METI.

For the purpose of CSCL, the substances are regulated by "METI Numbers" which are a series of numbers given for chemical substances that existed already in 1973 when CSCL was introduced along with newly registered chemical substances.

Since one METI number may be given to PFAS with different number of carbon atoms, it is not possible to find the amounts by carbon atoms from the statistics. Thus, Japan Endocrine Disruption Prevention Action could not confirm from the data based on CSCL notification that PFOA has been substituted by PFHxA and PFHxS, as Daikin and AGC claim on their websites.

On the other hand, the statistics for chemical substances with the METI number 2-1195 including e.g., ammonium perfluorooctanoate stopped after 2012. This is consistent with the announcement of the three companies above that they stopped the use of PFOA for fluoropolymers products by the end of 2013. However other PFOA-related substances such as the ones covered by METI numbers 2-1182 and 2-2659 continue to be manufactured even after 2016. These gaps are explained by the scope of substances covered by the METI number as mentioned above. The METI number 2-1182 substance, for example, is fluoroalkyl(C=2-10) carboxylic acid which includes both PFOA (C=8) and PFHxA (C=6).

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[5] Daikin https://www.daikinchemicals.com/company/sustainability.html
[6] AGC Seimi Chemical Co. Ltd. http://www.seimichemical.co.jp/eng/product/fluoro/PFOA/
[7] Materials prepared by Daikin to request for an exemption. http://chm.pops.int/Portals/0/download.aspx?d=UNEP-POPS-POPRC12CO-SUBM-PFOA-Daikin-01-20170602.En.pdf

According to METI, although the government has more detailed information on the use of substances, especially for the purpose of Stockholm Convention, the publicly available statistics do not show such details. It is considered that the figure for the fluoroalkyl carboxylic acid with METI number 2-1182 does not contain the amount of production/import for the one with eight carbon atoms according to the companies' announcement. However, as stated, citizens are not able to check the facts by using the current CSCL notification system. In addition, METI does not publicly disclose production information when there are only one or two companies manufacturing the substances, citing trade secrets.

Production / import notifications for PFOA, PFHxA, PFHxS and related substances under the CSCL

METI Numbers	name	Year and volume (ton)								
		2001	2004	2007	2011	2012	2013	2014	2015	2016
2-90	Perfluoroalkyl (C4-23) iodide	N/A	N/A	N/A	*	*	*	*	*	*
2-1182	Fluoroalkyl (C=2-10) carboxylic acid	N/A	N/A	1000 ～ 10,000	*	*	*	*	*	*
2-1195	Ammonium perfluorooctanoate	10～ 100	10～ 100	N/A	*	-	-	-	-	-
2-2402	2-Perfluoroalkyl (C=4-16) ethanol	N/A	N/A	10～ 100	<1000	<1000	<1000	<1000	<1000	<1000
2-2659	Perfluoroalkyl(C=7-13) carboxylic acid	N/A	N/A	N/A	*	*	*	*	*	*
2-2810	Salt (Na,K,Li) of perfluloroalkyl (C=4-12) sulfonic acid	N/A	N/A	10～ 100	<1000	<1000	<1000	<1000	<1000	<1000

Legends:

"*" METI received notification of manufacture/import, but METI does not disclose due to protect corporate secrets when the number of companies is one or two.

"-" METI did not receive notification (A company may manufacture/import less than 1 ton) when the volume

Source:

- For the data in 2001, 2004, 2007
  http://www.meti.go.jp/statistics/sei/kagaku/result-2.html
- For the date from 2011-2016
  http://www.meti.go.jp/policy/chemical_management/kasinhou/information/volume_index.html

Waste Management

The Waste Management and Public Cleansing Law (Law No.137 of 1970, hereafter referred to as "Waste Management Law") regulates wastes contains fluorine and its compounds. Regarding

wastes containing PFOS, the Ministry of Environment issued "Technical Documents on Treatment of Wastes containing PFOS (Revised March 2011)[8]" for PFOS or its salts. The documents describe detailed interpretations of the Waste Management Law for solid and liquid wastes generated in manufacturing and use of PFOS products and its salts. The documents set de-facto standards for storage, collection/transport, disposal, and facility for disposal.

Concerning import and export of POPs wastes in general, the government explains as below.[9]

> "The Export Trade Control Order and the Import Trade Control Order stipulate that the POPs wastes shall be subject to the requirement of import or export approval. These Orders, together with the relevant laws (the Waste Management Law and the Law for the Control of Export, Import and Others of Specified Hazardous Wastes and Other Wastes (Law No. 108 of 1992) etc. hereafter referred to as the Basel Law) ensure that stockpiles and wastes are disposed of in an environmentally sound manner in compliance with paragraph 1 (d) of Article 6 of the Stockholm Convention."

PFAS impacts

Surveys of PFOA pollution

Professor Akio Koizumi and his group reported in 2004 that high levels of PFOA contamination were found in rivers around Osaka and in the blood of residents. In 2003, a survey of 80 rivers located from Hokkaido, the northern part of Japan, to Kyushu, the southern part of Japan showed that PFOA was found in all the rivers. Most of them were present at low levels but 456 ng/L was detected at Inagawa River in Hyogo prefecture, and 140 ng/L in Yodogawa River. As a result of further research, 67,000 - 87,000 ng/L was detected from the sample taken around a sewage treatment facility located in Aigawa River, a branch of Yodogawa River. The survey of blood contamination in 200 people from 10 areas in Japan showed that the contamination levels of residents in Kyoto, Osaka, Nishinomiya are significantly higher than residents in other areas. Because PFOA contamination of residents in Osaka is 300 times higher than in other areas, blood contamination is considered to derive from contaminated drinking water. Levels of 8,300 ng/L and 57,000 ng/L of PFOA were detected in wells near Aigawa River in Osaka, which indicated that the ground water was severely contaminated with PFOA. Osaka and Hyogo Prefecture governments have continued to monitor PFOA contamination along the Aigawa River and Kanzakigawa River.

The Ministry of Environment also conducted surveys of PFOS contamination all over Japan. PFOA contamination in 2007 is shown below.

[8] https://www.env.go.jp/recycle/misc/pfos/tptc.pdf
[9] The National Implementation Plan of Japan under the Stockholm Convention on Persistent Organic Pollutants Modified in October 2016, 43p. http://www.env.go.jp/chemi/pops/plan/en_full-161006.pdf

PFOA detected in various media in 2007[10]

媒体 Media		幾何 平均値 Geometric Average	算術 平均値 Arithmetic Average	最小値 Minimum	最大値 Maximum	検出 下限値 Limit of detection	検出率 Detection rate	調査 地域 Area	測定年度 Year	文献 Literatu
一般環境大気 General atmosphere	μg/m³	0.0000090	0.00014	0.00000060	0.0025	0.00000014	20/20	Japan	2004	1)
室内空気 Indoor atmosphere	μg/m³									
食物 Food	μg/g	<0.000010	<0.000010	<0.000010	0.000024	0.000010	10/50	Japan	2004	1)
飲料水 Drinking water	μg/L	0.0018	0.0098	0.00012	0.04	-	6/6	Japan	2003	2)*)
		-	0.0086	0.0037	0.013	-	-/9	Kyoto	2007	3)
		0.029	0.03	0.019	0.041	*0.002* b)	3/3	Osaka	2007	4)
		0.0061	0.012	<0.001	0.038	*0.001* b)	31/35	Hyogo	2007	5)
		0.031	0.039	0.0079	0.11	*0.0001* b)	14/14	Osaka	2006	6)
		0.029	0.029	0.029	0.029	*0.002* b)	1/1	Osaka	2006	4)
		0.046	0.056	0.024	0.11	*0.002* b)	3/3	Osaka	2005	4)
		<0.005	0.0066	<0.005	0.025	*0.005* b)	9/19	Tokyo	2005	7)
地下水 ground water	μg/L	0.20	3.4	0.003	26	0.0004	8/8	Osaka	2007	8)
		22	33	8.3	57	0.1	2/2	Osaka	2006	9)
土壌 soil	μg/g									
公共用水域・淡水 public water area fresh water	μg/L	0.0035	0.0093	0.00030	0.034	0.00004	5/5	Japan	2005	10)
		0.0024	0.011	0.0001	0.46	0.00006	79/79	Japan	2003	2)
		0.0045	0.018	0.00044	0.096	0.00004	9/9	Japan	2002	11)

[10] https://www.env.go.jp/chemi/report/h19-03/pdf/chpt1/1-2-2-18.pdf

Media 媒体		Geometric Average 幾何平均値	Arithmetic Average 算術平均値	Minimum 最小値	Maximum 最大値	Limit of detection 検出下限値	Detection rate 検出率	Area 調査地域	Year 測定年度	Literature 文献
		0.16	1.9	0.016	31	0.0004~*0.002*	25/25	Osaka	2007	12)
		0.091	0.10	0.030	0.25	*0.002*[b]	16/16	Osaka	2007	13)[c]
		0.010	0.058	<0.001	0.67	0.001~0.003	42/59	Hyogo	2007	14)
		0.0039	0.031	<0.003	0.41	0.003	5/17	Hyogo	2006	14)
		0.0040	0.0082	0.0003	0.028	-	14/14	Tokyo	2005	15)
		0.013	0.014	0.0057	0.021	*0.00005*[b]	6/6	Tokyo Kanagawa	2004	16)[d]
		0.18	4.3	0.0045	67	0.00006	52/52	Osaka	2003	2)
		1.1	25	0.017	87	-	10/10	Osaka Kyoto	2003~2004	17)
公共用水域・海水 public water area / fresh water	μg/L	0.024	0.029	0.013	0.044	0.00004	2/2	Aichi Osaka	2005	10)
		0.011	0.083	0.0019	0.45	0.00006	6/6	Japan	2003	2)
		0.0033	0.015	0.0006	0.070	0.00004	11/11	Japan	2002	11)
		0.013	0.016	0.0063	0.03	-	3/3	Osaka	2007	18)
		0.027	0.032	0.018	0.066	*0.002*[b]	4/4	Osaka	2007	13)[c]
		0.023	0.023	0.023	0.023	-	1/1	Hyogo	2007	14)
		0.013	0.013	0.011	0.017	*0.00005*[b]	10/10	Chiba Tokyo Kanagawa	2004	16)[d]
底質(公共用水域・淡水) Sediment (public water area/fresh water	μg/g	0.000078	0.00039	<0.000024	0.0012	0.000024	3/4	Japan	2005	10)
		<0.00007	0.000083	<0.00007	0.00017	0.00007	5/9	Japan	2003	19)
		0.0028	0.025	0.0005	0.073	0.0003	3/3	Osaka	2007	20)
底質(公共用水域・海水) Sediment (public water area/sea water)	μg/g	0.00039	0.00057	0.00015	0.00098	0.000024	2/2	Kawasaki Aichi Osaka	2005	10)
		0.000072	0.00011	<0.00007	0.00041	0.00007	5/11	Japan	2003	19)
魚類(公共用水域・淡水) Fish (public water area/fresh water)	μg/g	0.000069	0.000070	0.000059	0.000085	0.000034	3/3	Niigata Tottori Kochi	2005	10)
		<0.000059	<0.000059	<0.000059	<0.000059	0.000059	0/3	Shiga Tottori Kochi	2003	19)
魚類(公共用水域・海水) Fish (public water area/sea water)	μg/g	0.000052	0.000075	<0.000034	0.00038	0.000034	12/16	Japan	2005	10)
		<0.000059	<0.000059	<0.000059	0.000062	0.000059	1/6	Japan	2003	19)
貝類(公共用水域・淡水) Shellfish (public water area/fresh water)	μg/g									
貝類(公共用水域・海水) Shellfish (public water area/sea water)	μg/g	0.00011	0.00013	0.000048	0.00025	0.000034	6/6	Japan	2005	10)

Since then, surveys on organofluorine compounds all over Japan have continued. In Okinawa, Okinawa, the local government started surveying the water supply source in 2013 and high levels

of PFOS and PFOA were detected. The contamination was caused by the US military base and the local government has conducted detailed surveys of PFAS water pollution since FY2016 (see more about the US military base contamination below).

PFAS pollution by Japanese companies

Daikin, AGC, and Mitsui-DuPont are the principal PFAS manufacturers in Japan. Daikin is the largest manufacturer and also operates factories in the US, France, and China.

In Japan, Mitsui-Dupont polluted water with PFOA at their factory in Shimizu. DuPont tracked PFOA in the blood of the workers at the facility as revealed in US EPA filings and internal company documents. High levels were found, including one worker with 8,370,000 ppt PFOA. PFOA was also found in water at 10 wells at the factory site with levels up to 1,540,000 ppt. The site was sold to DuPont spin-off company, Chemours who described the area as, "highly industrialized and the groundwater is brackish, and not a source of drinking water." The company also has a fluorochemical manufacturing facility in Chiba. Chemours-Mitsui Fluoroproducts is a 50:50 joint venture between the two companies.

In 2019, US EPA filed a notice of violation against Chemours – its first under the new Toxic Substances Control Act. The company had five serious violations including manufacturing a substance for commercial purposes that was not even on the regulator's inventory list. The notice carries a penalty up to USD$50,000 per day or a year in prison.

In China, Daikin is located in the Changshu Industrial Park in Jiangsu Province along with other foreign PFAS manufacturers such as Solvay and Arkema. In 2013, scientists measured some of the highest PFAS levels ever reported in China in the industrial park.

Daikin has received permits for PFAS pollution from the US Food and Drug Administration (FDA). FDA approved applications filed in 2009 – 2010 for Daikin's PFAS to be used in grease-repelling food packaging such as pizza boxes, sandwich wrappers and microwave popcorn bags. One application estimated release of 180 pounds/day (82 kg) PFAS in the wastewater discharge with a concentration of 83,000 ppt. Another estimated 225 pounds/day (102 kg) in the wastewater discharge with a concentration of 103,000 ppt. As a point of comparison Vermont sets a drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined.

AGC has also submitted proposals for PFAS in food contact materials to US FDA, providing limited toxicology data. The substances themselves are also complex. For example, an approved food contact material substance contains 15 substances including PFAS and FDA admits that four of them would remain in the food contact paper. However, the FDA hid this information along with the typical amount of impurities remaining in the paper contact material.

US regulators granted permits for PFAS substances in food contact materials to Daikin, AGC, and other companies despite knowing that they are released into food since 2008. A US FDA study tested PFAS-containing popcorn bags, muffin/croissant bags, hamburger bags, sandwich wrappers, pizza box liners and French fry/hash brown bags and found PFOA at 0.3 – 1.2 ppm. The agency tested short-time migration of PFAS into food and found migration into butter,

chocolate spread and microwaveable popcorn. Levels of PFAS in microwaveable popcorn were in the ppm range.

Ironically, grease-resistant PFAS-free food contact paper and cardboard has been available for approximately 10 years. Major manufacturers include Akzo Nobel and Imerys. Regulation has played a role in stimulating fluorine-free alternatives. For example, after Denmark prohibited PFAS in microwave popcorn bags, a supplier provided a stronger fluorine-free bag that fulfilled the grease resistant function by just boiling the paper cellulose for a longer period of time.

In 2016, the Northern Alabama water and sewer authority sued 3M and Daikin America due to pollution of the Tennessee River drinking water supply with PFOS and PFOA. The area is downstream of Daikin's factory in Decatur, Alabama. Local residents were warned by authorities not to drink the water. Daikin settled their part of the lawsuit for USD$4 million in 2018 to pay for a carbon filtration system. However, the settlement does not prevent individual residents from pursuing legal action.

In 2018, Kevin Hardwick, a firefighter, filed a national class-action lawsuit against nine PFAS manufacturers including AGC and Daikin, along with 3M, Archroma, Arkema, Chemours, Dyneon, DowDuPont, and Solvay. The purpose of the lawsuit is to fund an independent investigation of links between PFAS exposure and health impacts, including the entire class of PFAS substances. The suit is modeled after the class action litigation against DuPont for PFOA pollution which resulted in formation of the C8 Science Panel. The Panel associated PFOA exposure with high cholesterol, ulcerative colitis, thyroid disease, testicular cancer, kidney cancer, pregnancy-induced hypertension. Under the conditions of the lawsuit, DuPont could not contest these links in court and that led to subsequent verdicts against the company. As noted by the attorney filing the current lawsuit, "There is tremendous fear, anxiety, and uncertainty across the country as to the serious public health threat posed by PFAS contamination. This lawsuit could provide a mechanism for addressing and resolving those concerns through a truly comprehensive and independent, science-based process paid for by those that actually created the problem — and not by the American taxpayers."

US military base pollutes Okinawa

The US military base was reported as a source of environmental contamination. However, it was Mr. Jon Mitchell's reports[11] based on his investigation concerning a defoliant, Agent Orange, in Okinawa that made people aware of the seriousness of the contamination by the US military base. Some veterans of the US military testified that Agent Orange, which was used in the Vietnam War in the 1960's and 1970's, was brought to the base in Okinawa in spite of the fact that it was prohibited and that it caused health problems and environmental pollution. Mitchell has also reported extensively on PFAS contamination from the military bases in Okinawa.

In June 2013, oil drums were found in a soccer field of Okinawa City, located in an area used as a part of the Kadena Air Base and returned to Okinawa in 1987. The drums were labeled as "THE DOW CHEMICAL COMPANY," the biggest supplier of Agent Orange defoliants. PCBs

[11] https://www.jonmitchellinjapan.com/agent-orange-on-okinawa.html

and arsenic were detected from soil of the Okinawa City Soccer Field. The soil of Camp Kinser to be returned to Okinawa has been also contaminated.

In September 2015, an environmental agreement to supplement the Japan-U.S. Status of Forces Agreement (SOFA) was finalized. It was agreed that US Forces would develop Environmental Governing Standards for its facilities and military activities and comply with them. In addition, the agreement allowed for the US-Japan joint committee to access the site in case of accidents.

In advance of the environmental supplementary agreement, contamination due to use of foam extinguishing agents containing PFOS and PFOA at US bases had been regarded as social issues in Japan. Surveys by the Okinawa Prefectural Enterprise Bureau since 2013 showed that Kadena Air Base and Marine Corps Air Station at Futenma are considered the sources of the pollution. The Okinawa prefectural government has been conducting detailed surveys around the bases since 2016 and high levels of PFOS and PFOA contamination have been detected as stated in the next paragraphs.

Water Pollution Survey by the Okinawa Prefectural Enterprise Bureau

The Okinawa Prefectural Enterprise Bureau has been conducting surveys of raw water for water supply and filtered water since FY2013. In FY2013, the Bureau did not monitor the contamination frequently, but PFOS and PFOA level was over 70 ng/L at the Hija River Intake Pumping Station and Dakujaku River. The Bureau increased the frequency and sites of surveys since then. Average contamination levels for PFOS and PFOA between 2013 and 2018 were 170-293 ng/L and 346-706ng/L at the Hija River Intake Pumping Station and the maximum level was 1379 ng/L. The maximum level of PFOS and PFOA exceeded 70 ng/L at least one time at three sites: Nagata River Intake Pumping Station, Kawsaki Intake Pumping Station, and Kadena Well. Filtered water (treated for water supply) taken by citizens should not exceed 70 ng/L of PFOS and PFOA, but, at the Chatan Filiter Plant, the annual average of PFOS and PFOA for filtered water is 14-44 ng/L and two exceeding results: 82 ng/L and 120ng/L were recorded in FY2015. To specify the source of contamination at the Hija River Intake Pump Station, the Bureau surveyed ground water near Kadena Air Base in 2018 and the contamination levels were exceeded at two locations.

The Bureau stated as follows[12].

> Perfluorinated Compounds (PFOS and etc.) are detected at higher levels in comparison with other rivers at the Chatan Filtering Plant taking water from the Hija River and the Kadena Wells and they were found in filtered water.
>
> The toxicity and impact on human beings of PFOS exposure are not determined clearly. There are no standards for water quality regarding PFOS and PFOA contamination but PFOS was recognized as a factor to be reviewed. In May 2016, EPA lowered the contamination level from 200ng/L to 70 ng/L as a temporary health advisory. The level of 70ng/L is considered as sufficiently low for any person to drink the water for lifetime without problems.

[12] https://www.eb.pref.okinawa.jp/opeb/309/619

The bureau set the target level of the Chatan Filtering Plant at less than 70 ng/L PFOS and PFOA to control water quality and supply safe water to people in Okinawa.

The total concentration of PFOS and PFOA averages 30 ng/L at the Chatan Filtering Plant and accordingly the Bureau confirms that PFOS and other substances are reduced to the safe level. By period replacement of granulated activated carbon absorbing PFOS and other substances, the Bureau will take every measure to supply safe water.

Sites for surveys
A: Hija River Intake Pump Station
B: Nagata River Intake Pump Station
C: Kawakaki Intake Pump Station
D: Kadena Well
E: Dakujaku River
F: Raw Water at Chatan Filtering Plant
G: Filtered Water at Chatan Filtering Plant
H: Raw Water at Nago Filtering Plant
I: Filtered Water at Nago Filtering Plant
J: Raw Water at Ishikawa Filtering Plant
K: Filtered Water at Ishikawa Filtering Plant
L: Raw Water at Nishihara Filtering Plant
M: Filtered Water at Nishihara Filtering Plant

The data of surveys between 2013-2017

Okinawa Prefectural Enterprise Bureau Survey on PFOS and PFOA

Unit: ng/L

2013

	A	B	C	D	E	F	G
	比謝川取水ポンプ場			嘉手納井戸集合	大工廻川	北谷浄水場原水	北谷浄水場浄水
Max.	195			81	687	59	41
Min.	144			81	687	58	38
Ave.	170			81	687	59	40
Freq.	2			1	1	2	2

2014

	A	B	C	D	E	F	G	H	I	J	K	L	M
	比謝川取水ポンプ場	長田川取水ポンプ場	川崎取水ポンプ場	嘉手納井戸集合	大工廻川	北谷浄水場原水	北谷浄水場浄水	名護浄水場原水	名護浄水場浄水	石川浄水場原水	石川浄水場浄水	西原浄水場原水	西原浄水場浄水
Max.	414	218	87	77	1379	40	36	<1	<1	1	<1	<1	<1
Min.	121	8	61	52	436	2	18	<1	<1	1	<1	<1	<1
Ave.	209	130	75	65	706	20	26	<1	<1	1	<1	<1	<1
Freq.	14	4	3	11	12	11	11	1	1	1	1	1	1

2015

	A	B	C	D	E	F	G	H	I	J	K	L	M
	比謝川取水ポンプ場	長田川取水ポンプ場		嘉手納井戸集合	大工廻川	北谷浄水場原水	北谷浄水場浄水	名護浄水場原水	名護浄水場浄水	石川浄水場原水	石川浄水場浄水	西原浄水場原水	西原浄水場浄水
Max.	590	580		143	744	112	120	<1	<1	<1	2	<1	<1
Min.	26	4		50	394	2	22	<1	<1	<1	2	<1	<1
Ave.	242	73		86	579	35	44	<1	<1	<1	2	<1	<1
Freq.	12	12		12	4	12	12	1	1	1	1	1	1

2016

	A	B	C	D	E	F	G	H	I	J	K	L	M
	比謝川取水ポンプ場	長田川取水ポンプ場	川崎取水ポンプ場	嘉手納井戸集合	大工廻川	北谷浄水場原水	北谷浄水場浄水	名護浄水場原水	名護浄水場浄水	石川浄水場原水	石川浄水場浄水	西原浄水場原水	西原浄水場浄水
Max.	506	154	90	87	621	63	30	<1	<1	2	1	<1	<1
Min.	137	10	48	55	316	<1	3	<1	<1	2	1	<1	<1
Ave.	293	45	62	63	447	16	14	<1	<1	2	1	<1	<1
Freq.	12	12	10	13	4	13	13	1	1	1	1	1	1

2017

	A	B	C	D	E	F	G	H	I	J	K	L	M
	比謝川取水ポンプ場	長田川取水ポンプ場	川崎取水ポンプ場	嘉手納井戸集合	大工廻川	北谷浄水場原水	北谷浄水場浄水	名護浄水場原水	名護浄水場浄水	石川浄水場原水	石川浄水場浄水	西原浄水場原水	西原浄水場浄水
Max.	498	241	93	68	491	74	59	1	1	2	2	1	1
Min.	98	12	42	49	213	12	15	1	1	2	2	1	1
Ave.	204	81	60	58	346	40	27	1	1	2	2	1	1
Freq.	12	12	12	12	9	13	13	1	1	1	1	1	1

The data of surveys in 2018

Okinawa Prefectural Enterprise Bureau
Survey on PFOS and PFOA Unit: ng/L

2018		A	B	C	D	E	F	G	H	I	J	K	L	M
		北谷浄水場	北谷浄水場	北谷浄水場	嘉手納水道企業団	大工廻川	北谷浄水場	北谷浄水場	石嶺浄水場	石嶺浄水場	石川浄水場	石川浄水場	西原浄水場	西原浄水場
		取水ポンプ場	取水ポンプ場	取水ポンプ場			原水	浄水	原水	浄水	原水	浄水	原水	浄水
Max.		375	684	91	105	971	72	63	<1	<1	<1	<1	<1	<1
Min.		67	5	34	51	92	1	13	<1	<1	<1	<1	<1	<1
Ave.		190	98	59	71	458	32	30	<1	<1	<1	<1	<1	<1
Freq. of surveys		44	45	43	46	35	46	46	1	1	1	1	1	1
4月3日	4/3	211	341	62	71	841	64	47						
4月10日	4/10	181	334	59	67	634	58	42						
4月18日	4/18	103	12	53	79	201	17	24						
4月23日	4/23	80	35	36	61	182	18	29						
5月8日	5/8	203	336		74	372	69	47						
5月15日	5/15	196	442	62	72	501	67	52						
5月22日	5/22	184	425	61	71	307	64	53						
5月28日	5/28	199	482	50	60	506	63	63						
6月5日	6/5	134	104	39	58	306	30	27						
6月11日	6/11	196	684	57	58	390	72	48						
6月18日	6/18	106	26	70	51	269	31	27						
6月26日	6/26	225	9	54	57		34	31						
7月3日	7/3	85	5	41	58	320	10	19						
7月9日	7/9	155	8	85	63		12	20						
7月17日	7/17	171	14	48	63		23	24						
7月25日	7/25	173	41	79	67	482	30	23						
7月30日	7/30	319	73	71	62		12	22						
8月7日	8/7	320	72	91	57		29	22						
8月14日	8/14	123	14	60	57		16	20						
8月20日	8/20	176	14	76	63		21	18						
8月27日	8/27	217	15	53	56	340	23	19						
9月3日	9/3	127	34	63	62		8	19						
9月11日	9/11				61		9	18						
9月18日	9/18	164	19	66	76		24	30						
9月25日	9/25	174	47	72	70	275	23	25						
10月2日	10/2	113	17	47	71	448	23	22						
10月10日	10/10	128	7	45	64	378	4	13						
10月16日	10/16	113	5	41	73	428	1	13						
10月23日	10/23	178	11	61	77	351	14	16						
10月30日	10/30	196	15	65	90	509	37	25						
11月5日	11/5	136	12	62	105	439	31	28						
11月13日	11/13	224	14		92	92	41	32						
11月20日	11/20	163	11	58	84	431	34	30						
11月28日	11/28	351	35	68	76	463	56	36						
12月3日	12/3	204	60	67	82	490	54	34						
12月11日	12/11	159	7	48	81	330	15	24	<1	<1	<1	<1	<1	<1
12月17日	12/17	294	9	60	75	495	48	31						
12月25日	12/25	238	40	61	81	507	47	39						
1月9日	1/9		45	60	86		13	26						
1月15日	1/15	375	84	67	87	659	52	39						
1月22日	1/22	291	92	59	75	707	48	35						
1月28日	1/28	300	261	61	82	764	56	45						
2月5日	2/5	206	55	69	76	971	24	29						
2月12日	2/12	241	14	62	73	913	42	32						
2月20日	2/20	148	15	35	75	414	15	24						
2月25日	2/25	67	8	34	72	321	12	32						

Detailed Survey of the Marine Corps Air Station Futenma by the Okinawa Prefectural Government

The Okinawa Prefectural Government has conducted detailed surveys around the Marin Corps Air Station Futenma since 2016. The results for summer and winter in 2016 and 2017 and summer in 2018 are published.[13]

(ng/L)

Result List

	Municipality	Location	Summer Survey(JFY2016)			Winter Survey(JFY2016)			Summer Survey(JFY2017)			Winter Survey(JFY2017)			Summer Survey(JFY2018)				
			PFOS	PFOA	Total Value	PFOS	PFOA	Total Value	PFOS	PFOA	Total Value	PFOS	PFOA	Total Value	PFOS	PFOA	Total Value	6:2FTS	8:2FTS
①	Chatan Town	Surrounding area of Camp Zukeran, Western Drainage	30	11	41	57	7.5	64	29	9.1	38	30	8.0	38	27	8.3	35	5.4	1.4
②	Ginowan City	Surrounding area of Futenma Air Station, Chunnaga (spring)	1200	190	1300	730	150	880	740	140	880	900	130	1000	1800	200	2000	390	40
③	Ginowan City	Surrounding area of Futenma Air Station, Hunshinga (spring)	38	21	59	39	22	61	37	25	62	39	22	61	39	23	62	4.4	0.4
④	Ginowan City	Surrounding area of Futenma Air Station, Hiyakaga (spring)	180	31	210	94	26	120	120	33	150	160	36	190	150	29	170	75	6.9
⑤	Ginowan City	Surrounding area of Futenma Air Station, Mendakarihiga (spring)	680	35	710	670	42	710	590	43	630	640	42	680	600	50	650	150	31
⑥	Ginowan City	Surrounding area of Futenma Air Station, Morinokawa (spring)	30	9.4	39	40	5.4	45	39	11	50	71	25	96	46	6.4	52	2.0	<0.1
⑦	Ginowan City	Surrounding area of Futenma Air Station, Samashita Ubuga (spring)	24	9.0	33	30	11	41	18	8.8	26	13	7.2	20	25	9.9	34	<0.1	<0.1
⑧	Ginowan City	Surrounding area of Futenma Air Station, Isaufuga (spring)				130	62	190	120	35	150	250	42	290	220	60	280	17	14
⑨	Ginowan City	Surrounding area of Futenma Air Station, Furuchinga (spring)				96	22	110	66	17	83	49	14	63	30	11	41	21	0.8
⑩	Ginowan City	Surrounding area of Futenma Air Station, Aragusuku A (groundwater)							15	4.4	19	17	4.8	21	15	4.3	19	<0.1	<0.1
⑪	Ginowan City	Surrounding are Futenma Air Station, Kyuna A (groundwater)							260	26	280	320	29	340	280	24	300	<0.1	<0.1
⑫	Ginowan City	Surrounding area Futenma Air Station, Kyuna B (groundwater)							40	31	71	34	23	57	76	72	140	0.1	<0.1
⑬	Ginowan City	Surrounding area Futenma Air Station, Aragusuku B (groundwater)							40	15	55	35	12	47	42	13	55	<0.1	<0.1
⑭	Ginowan City	Surrounding area of Futenma Air Station, In Civic Park (Upstream Surface-Water)				6.6	3.8	10	11	4.6	15	6.9	3.9	10	6.8	4.1	10	0.2	<0.1
⑮	Ginowan City	Surrounding area of Futenma Air Station, Ginowan Kumaibu Ritual Site (spring)	7.2	3.9	11	6.7	3.0	9.7	11	5.5	16	6.2	3.5	9.0	9.4	3.4	12	<0.1	<0.1
⑯	Ginowan City	Surrounding area of Futenma Air Station, Akamichi (Upstream Surface-Water)				12	4.1	16	11	5.1	16	7.8	4.7	12	13	4.9	17	0.2	<0.1

(Note) The results of measurement are shown in two effective digits (disregarding the third digit) in accordance with "Designation of Water Type in Environmental Standards Based on the Environmental Basic Law and Processing Standards Including Continuous Monitoring Based on the Water Pollution Prevention Act (Ref. 1303271 of March 27,2013)." When the total value was below the lowest detectable limit, the detectable value, 0.04ng/L is used to calcurate the value.
(Note) Regarding past measurement results, only the survey measurement points in FY2018 are shown.

The number of sites for survey has been increasing. In the summer of 2016, the number of sites with total of PFOS and PFOA concentration exceeding 70 ng/L was three, but it became 8 in the summer of 2018. The Informed Public Project (IPP) created a map[14] (below) to show the surveyed sites and results based on the survey results in the summer of 2017. US military officials have admitted using PFOS-containing firefighting foams at the base, after the substances were detected in streams running through the base and in groundwater wells. Levels of PFOS/PFOA more than 70 ng/L, (the current US federal health advisory level), were detected in the rivers and groundwater in the northern part of Futenma Air Station and the contaminated area is expanding. A US military publication notes that the Okinawa Prefecture had to pay USD$1.5 million to install a carbon filtration system to deal with US military PFAS pollution.

In 2016, internal US government documents revealed lax safety standards at the Kadena Air Base. As noted, "The internal reports expose a spate of accidents at the base during the past 15 years that have involved at least 21,000 liters of fire extinguishing agents — some of them toxic." The Base has effectively polluted the waterways that supply drinking water to seven municipalities, including the prefectural capital, Naha. The updated Status of Forces agreement between the US and Japan grants Japanese regulators the right to visit military installations in the case of chemicals spills. However, the US has continued to refuse Japanese government requests and the source of contamination has not yet been identified. In fact, US Marine Corps

[13] https://www.pref.okinawa.jp/site/kankyo/hozen/mizu_tsuchi/water/documents/jfy2018s_report.pdf
[14] http://ipp.okinawa/2018/10/22/ikensho/ translated by Japan Endocrine Disruption Prevention Action

Installations Pacific declined the Japanese government request saying that, "PFOS is not a regulated substance in the US and Japan." Without eliminating the source or taking other measures, PFAS pollution will continue. The documents also highlight a loophole in the Japan Environmental Governing Standards pertaining to US military bases in Japan. The Standards require the US to notify the Japanese government immediately when "a significant spill . . . threatens the local Japanese drinking water resource." However, the decision to characterize a spill as significant is left up to the US military. None of the egregious spills described in the internal documents were reported to Japanese government regulators.

In 2018, after the US Agency for Toxic Substances and Disease Registry (ATSDR) updated the Toxicological Profile for Perfluoroalkyls: Draft for Public Comment, the Informed-Public Project in Okinawa sent a letter to the Okinawa Prefecture requesting strong measures to address US military PFAS pollution including monitoring and adopting stronger regulatory limits as done by US states and to widen the focus to PFAS as a class. The letter urges action, noting that ATSDR recommended tightening limits for PFOS to 7 ppt and PFOA to 11 ppt from the current unenforceable US federal advisory limit of 70 ppt combined. The measures requested include:

- Review the prefecture's current policies on the safety standards for drinking water by examining recent policy revisions made by PFAS affected states and local communities in the US;
- Review with municipalities and local communities the present protocols as to how to address the issues of PFOS/PFOA contamination for affected areas;
- Examine epidemiological studies conducted in PFAS affected areas around military bases in U.S. and take a public health approach to PFAS contamination issues in Okinawa;
- Review how the prefectural government's bureaucratic sectionalism has hindered its ability to deal with PFOS/PFOA issues and develop an information-sharing and knowledge-building system to effectively address a whole range of PFAS contamination issues caused by PFOS/PFOA and other compounds;
- Generate and spread information on PFOS/PFOA contamination issues throughout Japan and request the Japanese Government to make a strong commitment to solving the issues.
- We also requested the prefectural government to be transparent and accountable to the people of Okinawa in developing and implementing new policies.

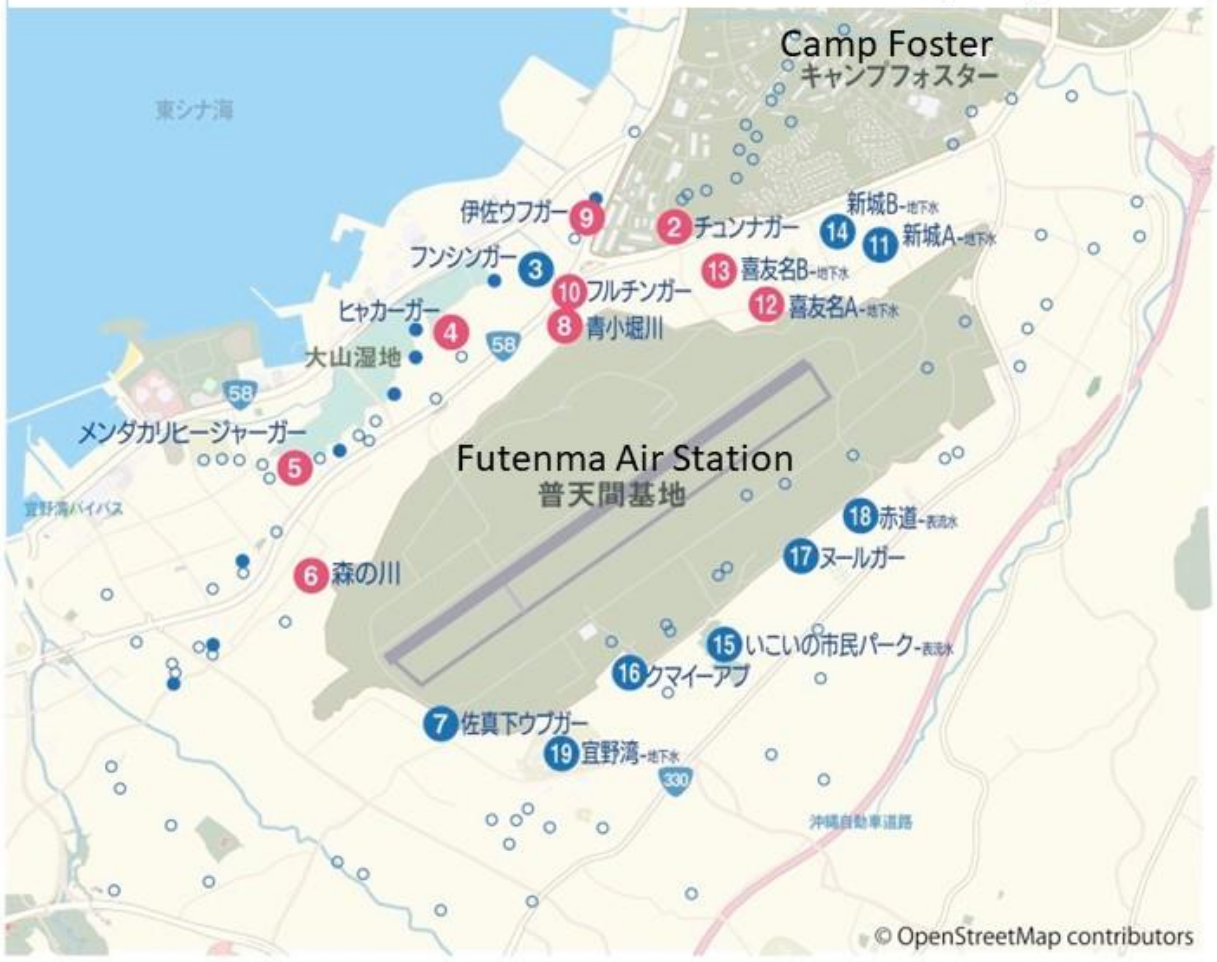
In 2018, Asahi Shimbun reported that the US military denied Okinawa authorities access to the military bases to track the sources of PFAS pollution. The US military confirmed nine leaks of PFAS-containing firefighting foams since 2001. The report notes that representatives of the two governments have met four times to discuss the pollution issues since 2016, but "the US side has not disclosed the results of its study concerning the suspected contamination."

Internal US government documents described in a 2019 report indicate extreme PFAS contamination inside and outside of the Kadena Air Base-. For example, foam-fire extinguisher located around a pond situated 200 meters from the Kadena town hall contained 90,000 ppt PFOS. A survey of fire sprinklers in 11 buildings found up to 9.5 billion ppt PFOS and 99 million ppt PFOA and a fire sprinkler in a building 250 meters from the southern boundary of the

military base contained 28 million ppt PFOS. The report notes that, “the US military has refused to confirm the actual relationship and has also refused to participate in the on-site investigation.”

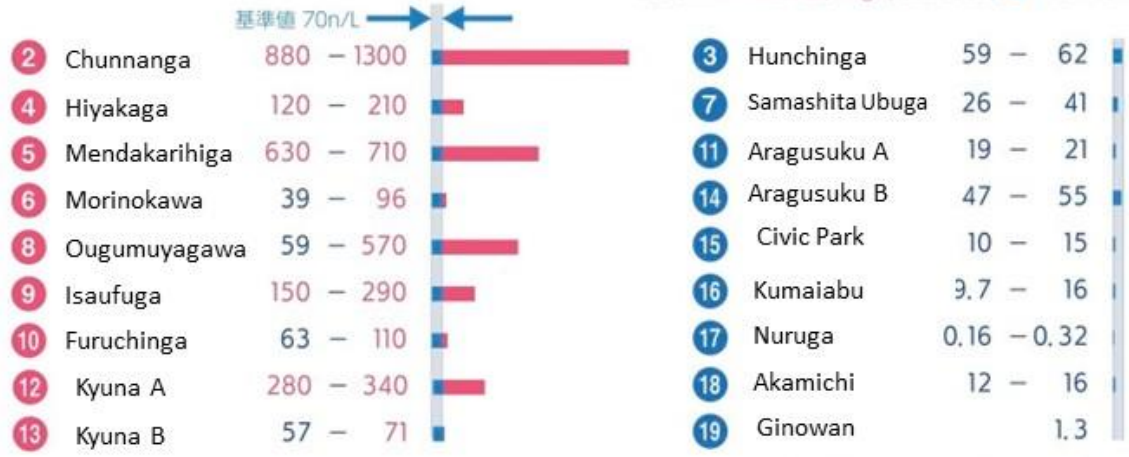
PFOS/PFOA contamination around Futenma Air Station (spring/groundwater)

by IPP, Sep. 7, 2018



Total amount of PFOS/PFOA detected in spring and etc.

数値は2016年度夏季～2017年度冬季間の最大と最小値
赤文字は、EPAの基準値(70ng/L)を上回る部分。●○はその他の湧水など



沖縄県環境部環境保全課 平成29年度PFOS・PFOA調査結果について(冬季結果)より

Scientific studies on PFAS in Japan

Government reports

Japan Ministry of Environment (2018) Annual survey of marine environment after the Great East Japan Earthquake, 2011-2017
https://www.env.go.jp/press/files/jp/109573.pdf
https://www.env.go.jp/press/files/jp/102774.pdf
http://www.env.go.jp/jishin/monitoring/result_me120413.pdf

The Ministry of the Environment surveyed the marine environment for radioactive and pollutant chemicals in 12 sampling locations of the Tohoku Pacific coasts after the Great East Japan Earthquake in 2011. Pollutant chemicals included PFOS and PFOA. The study measured both substances in bottom sediments and surface water.

Bottom sediments: In 2017, concentrations of PFOS and PFOA in 12 sampling locations were 3-140 pg/g dry weight and 30-990 pg/g dry weight respectively. In 2011, concentrations of PFOS, PFOA in 12 sampling locations were 2.6-160 pg/g dry weight and 5.0-180 pg/g dry weight respectively.

Surface water: In 2015, concentrations of PFOS, PFOA in 12 sampling locations were 0.007 ~ 0.098 ng/L and 0.21 ~ 0.63 ng/L respectively. In 2011, concentrations of PFOS and PFOA in 12 sampling locations were ND~0.071 ng/L and 0.13~0.6 ng/L respectively.

The authors suggested that these levels of PFOS and PFOA contamination were almost like other areas of Japan, according to the data collected under the “Chemicals in the Environment” study by Ministry of the Environment (see below).

Kishi R, Araki A (2018) Environmental chemicals and their effects on children based on the first birth cohort studies in Japan, Journal of the National Institute of Public Health, 67: 292 - 305

This review summarized their two cohort studies, which were examined for correlation of child health and environmental chemicals. In the Sapporo cohort, 514 sets of maternal and cord blood were analyzed for environmental chemicals including PFOS and PFOA from 2001 to 2004. In the Hokkaido cohort, 20,926 sets were examined from 2002 to 2012 including 11 PFAS substances: PFHpA, PFHxA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxS, and PFOS.

From their studies, prenatal exposures to PFAS, such as PFOS and PFOA, may affect birth size, homeostasis of several hormones (thyroid, steroid, sex hormones et al.), and development of the nervous system, allergies, and infectious diseases.

In males, there are negative correlations between prenatal exposure of PFAS and production of testosterone, estradiol, inhibin B, and insulin-like factor 3. In females, there are negative

correlations between prenatal exposures to PFAS and production of progesterone, prolactin, and sex hormone-binding globulin (SHBG).

Recently, exposures of PFNA and PFDA are increasing in Japan, instead of PFOS and PFOA.

Japan Ministry of Environment (2017) Survey of the Exposure to chemical compounds in humans, Environmental Risk Assessment Office

The Environmental Risk Assessment Office, Environmental Health Department, Ministry of the Environment, Japan (MOE) has conducted a project entitled, "Survey of the Exposure to Dioxins and other chemical compounds in Humans" to evaluate the exposure and accumulation levels of dioxins and other chemical compounds in Japanese people in FY 2011, and "Survey of the Exposure to chemical compounds in Humans" since FY 2012. The Environmental Risk Assessment Office has compiled 6 years of survey results conducted during FY 2011–2016.

Results from the survey of organofluorine compounds (2011-2016) were:

Blood concentration: The average PFOS level in 406 participants was 4.1 ng/ml (ppb) with a range of 0.29-17 ng/mL (ppb), and the PFOA average was 2.2 ng/mL (ppb) with a range of 0.27-13 ng/ml (ppb).

Intake: The average PFOS exposure of 15 participants was 0.57 ng/kg body weight/day with a range of N.D. to 1.7 ng/kg body weight/day, and the average PFOA exposure was 0.69 ng/kg body weight/day with a range of N.D. to 2.9 ng/kg body weight/day. Monitored PFAS included PFOS, PFOA, PFHxA, PFHpA, PFHxS, PFTeDA, PFNA, PFDA, PFUdA, PFDS, PFDoA, and PFTrDA.

Shiokawa A, Tamaki F (2017) The survey on environmental pollution of Perfluorinated compounds (PFCs) in rivers and the sea areas of Okinawa island, Annual report of Okinawa Prefectural Institute of Health and Environment, 51: 33-48

PFAS in water were surveyed in 72 places from 18 rivers and 3 sea areas of Okinawa's main island (Okinawa Honto). PFAS were not detected in the 3 sea areas. In river samples, 4-8 carbon chains of PFCs were detected, and more than 10-carbon polymers of PFAS were not found. Examined PFCs were PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFHpS, and PFOS. Among the river samples, PFAS levels in city areas were higher than those in mountain and agricultural areas. In the samples from 3 rivers (Hyjya, Tengan, and Nagata), PFAS levels were higher than those in other rivers.

Japan Ministry of Environment (2016) Chemicals in the environment, Report on Environmental Survey and Monitoring of Chemicals in FY2016

The Ministry of Environment surveyed various chemicals including POPs, PFAS, pesticides etc. in surface water, sediment, wildlife, and air at 49 points from north to south in Japan.

Surface water: In 2016, concentrations of PFOS and PFOA ranged from trace-14,000 pg/L, and 260-21,000 pg/L respectively. In 2010, concentrations of PFOS and PFOA ranged from trace-230,000 pg/L and 190-23,000 pg/L respectively.

Sediment: In 2016, concentrations of PFOS and PFOA ranged from trace-17,00 pg/g-dry weight and ND-190 pg/g-dry weight respectively. In 2010, concentrations of PFOS and PFOA ranged from 5-690 pg/g-dry weight and ND-180 pg/g-dry weight respectively.

Wildlife: In 2016, concentrations of PFOS and PFOA in fish ranged from ND-5,200 pg/g-wet weight and ND-20 pg/g-wet weight respectively. In 2010, concentrations of PFOS and PFOA in fish ranged from ND-15,000 pg/g-wet weight and ND-95 pg/g-wet weight respectively.

Air: In 2016, concentrations of PFOS and PFOA ranged from 0.7-9.3 pg/m^3 and 4.0-210 pg/m^3 respectively. In 2010, concentrations of PFOS and PFOA ranged from 1.6-14 pg/m^3 and 3.2-140 pg/m^3 respectively.

Yamashita S, Toyofuku S, Towatari H, Uno E, Matsuo Y (2015) Survey on Perfluorinated Compounds (PFCs) in Water and Bottom Sediments in Public Water of Fukuoka City, Fukuoka city institute of health and environment, Annual report, 40: 74-79, 2015

Twenty PFAS substances were surveyed in water and bottom sediment at 22 points in river and sea areas in the city of Fukuoka, Fukuoka Prefecture during 2012-2014. PFOS was detected in all samples. The concentrations of PFOS and PFOA in river water samples were higher than those in sea areas, and their highest concentrations were 5.6 ng/L and 8.1 ng/L, respectively. The concentrations of PFOS and PFOA in bottom sediment of the sea area were higher than those of rivers. Their highest concentrations were about 300 pg/g-dry weight (PFOS) and about 120 pg/g-dry weight (PFOA).

Hirano S, Ohara K, Toyofuku S, Matsuo Y (2013) Survey on perfluorinated compounds (PFCs) and their Emission Sources in the Water Environment of Fukuoka City (Ⅱ), Fukuoka city institute of health and environment, Annual report, 38: 45-49

This study surveyed PFAS in the water environment of the city of Fukuoka and emission sources from the sewage treatment plant. As the result of the survey at monitoring points for Environmental Quality Standard (EQS) and supplemental points of rivers and Hakata bay, perfluorononanoic acid (PFNA), perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS) were <0.2 ~ 100 ng/L, <0.2 ~ 14 ng/L and <0.2 ~ 11 ng/L respectively. In comparison to the previous report, the PFAS concentration is falling slightly. PFAS were also detected in the discharge water of four sewage treatment plants in Fukuoka and the levels of PFNA were high as observed in the monitoring of rivers. Intermittent sampling of discharge and influent intermittently for 24 hours at one plant showed sharp changes in PFAS concentrations but there were no big concentration changes and the levels were equalized by treatment in the plant.

Kawashita H, Fukushima A, Miki T, Yoshikawa M (2013) Survey Research on Perfluorinated Chemicals（4）, Annual report, Fukui Prefectural Institute of Public Health and Environmental Science, 12: 60-63

During 2011 and 2013, PFAS were assayed in water and bottom sediment from 30 rivers in Fukui Prefecture. Nineteen PFAS were measured including 4 FTOHs and precursors of PFOA. The highest concentration of PFOS was more than 10 ng/L in one sample. The highest concentration of PFOA was 800 ng/L. It came from an industrial drainage of a textile dyeing area. The drainage included FTOHs, which were decomposed and resulted in PFOA.

Gamo T (2011) Chemical Oceanography to Elucidate Global Kinetics of Persistent Perfluorinated Chemicals (PFCs), Report of the Environment Research and Technology Development Fund, Japan Ministry of the Environment

A group of PFAS including PFOS and related chemicals were precisely measured in open ocean water samples collected by international cooperative research cruises, using research vessels Hakuho-Maru, Tansei-Maru, and Mirai. In total, 1835 samples of surface and deep sea waters were obtained, covering the Pacific Ocean, Atlantic Ocean, Arctic Ocean, Atlantic Ocean, and Indian Ocean.

In order to understand the global distribution and fate of PFAS, we have developed an ultra-trace analysis method to measure PFAS in open seawater and open ocean air. A new type of air sampler, the "Cryogenic Moisture Sampler (CMS)", enables us to collect large volumes of air with moisture in the open ocean. An international calibration study was carried out during the project. The study revealed that strict quality assurance and quality control (QAQC) is necessary to conduct open ocean monitoring of PFAS. The result was presented at an international conference to recommend QA/QC for UNEP monitoring of PFAS in the ocean in 2015.

Up-to-date world maps were successfully drawn to show the global distribution of PFOS and PFOA for surface waters using the developed analytical method. The cross section of PFOA concentration was first described in the northwestern Pacific Ocean and suggested that most PFOA pollution still remained in sub-surface water down to 300 m deep. In the Asia region, horizontal transport of PFOA in surface water could be traced up to 165E at relatively higher concentration.

Significant temporal changes were found in both PFOS and PFOA in the ocean. Concentrations of PFOS and PFOA in surface waters collected in the Japan Sea increased three-fold and five-fold, respectively, between 2005 and 2010. This is probably due to the input of contaminated surface water from the East China Sea through the Tsushima Strait. Time-series monitoring of PFAS in seawater should be continued not only in the Japan Sea but also in global oceans for the protection of ocean environments in the future.

In order to evaluate PFAS as chemical tracers, their water column profiles were compared with those of plutonium, which is well known as a traditional chemical tracer. The increasing rate of PFOS/PFOA in the Japan Sea was smaller than those of plutonium. It indicated that PFAS substances were more easily transported by ocean currents than plutonium. Over all, these results clearly demonstrated that the PFAS are good chemical tracers for various studies on ocean dynamics.

Japan National Institute for Environmental Studies (2006) Study on the establishment of scientific and technical foundation for assessment of sources, development of distribution method and education of pollution status of POPs-like compounds, especially organofluorine chemicals
A survey of PFOS, PFOA, PFNA, PFDA, PFuDA, PFdDA in water such as rivers and sea, and sewerage of several points in Japan was conducted from 2004 to 2006. In river water, (Tama River in Tokyo), the range of PFOS concentrations was 0.5 ~ 58 ng/L in 2004. In sewerage water (Tokyo), the range of concentrations of PFOS and PFOA were 5~416 ng/L and 7~174 ng/L, respectively. This indicated that wastewater treatment plants pass PFOS and PFOA to effluents and end up downstream. Contamination of PFAS in mussels was examined in nine bay areas. At Kanazawa and North Ibaraki, high concentrations of PFAS were observed in mussels. At Kanazawa, concentrations of PFOS and PFuDA were higher than those of other PFAS. At North Ibaraki, concentrations of PFOA, PFDA, and PFdDA were higher than other PFAS.

Journals

Iwabuchi K, Tatarazako N (2018) Relationship between Occurrences of Perfluoroalkyl Acids in Medaka, Environmental Water, and Sediment in Its Habitat and Bioconcentration, Journal of Japan Society on Water Environment 41: 61-71
Perfluoroalkyl acid (PFAA) is diffused into the environment and detected in wildlife and its environmental persistence and accumulation is becoming a major concern. In this study, we determined the homolog distribution of 15 PFAA (PFAS) in samples of medaka (Olyzias latipes), surrounding water, and sediment collected from 5 sites in Japan. PFAA concentrations in environmental water, sediment, and medaka differed depending on the sampling site; however, each PFAA composition ratio was almost constant within these sampling sites. The PFAA bioconcentration factor (BCF) in medaka was related with the octanol/water partition coefficient (Log Kow) of PFAA. PFAA concentrations in the water samples had better correlations with those in the sediment samples not per unit dry weight, but per unit ignition loss (IL). In addition, between sediment and medaka, good correlations were found when the comparison was done with the PFAA concentrations of sediment per unit IL. The PFAA accumulation in medaka was estimated by multi-regression analysis using the PFAA concentrations in sediment per unit IL, gender, and body length. The PFAA concentrations in sediment per unit IL were significantly related to accumulation in medaka.

Fujii Y, Kato Y, Sakamoto K, Matsuishi T, Kouji HH, Koizumi A, Kimura O, Endo T, Haraguchi K (2018) Tissue-specific bioaccumulation of long-chain perfluorinated carboxylic acids and halogenated methylbipyrroles in Dall's porpoises (Phocoenoides dalli) and harbor porpoises (Phocoena phocoena) stranded in northern Japan, Sci Total Environ 616-617:554-563
This study investigated PFAS and other substances in the liver, blood, and blubber of Dall's porpoises and harbor porpoises in Hokkaido. PFNA, PFUnDA, PFDoDA, PFTrDA and PFTeDA were found in all samples. PFOA was found in 94% of samples and PFOS in 98% of them. PFHxS was found in 54% of the samples. PFUnDA and PFTrDA accounted for 70% of the total PFCAs in all tissues. Among the PFCAs found in the liver of Dall's porpoise, the concentration

of PFUnDA was the highest (226 ng/g-wet), followed by perfluorotridecanoic acid (PFTrDA, 102 ng/g-wet), and that of PFOA was the lowest (5.2 ng/g-wet). For perfluoroalkyl sulfonates mean concentrations were 14 and 5.4 ng/g-wet in liver, 0.6 and 0.5 ng/g-wet in blood, and 0.1 and 0.1 ng/g-wet in blubber from Dall's and harbor porpoises, respectively. The authors note that the higher concentrations of PFCAs observed in the Dall's porpoises indicate that PFASs are vertically distributed in deep seawater. The authors also state that the unusual accumulation profile of long-chain PFCAs and the predominance of PFUnDA and PFTrDA in the liver suggested that FTOHs may be a major source of PFCAs in this area.

Tasi MS, Miyashita C, Araki A, Itoh S, Bamai YA, Goudarzi H, Okada E, Kashino I, Matsuura H, Kishi R (2018) Determinants and Temporal Trends of Perfluoroalkyl Substances in Pregnant Women: The Hokkaido Study on Environment and Children's Health, Int J Environ Res Public Health 15:989-1003
This study is part of the Hokkaido large-scale cohort that recruited 20,926 pregnant women between 2003 and 2011 to study environmental health relationships. This study included 2,163 women. Eight PFAS substances were found in plasma at detection rates of more than 80%: PFHxS, PFOS, PFOA, PFNA, PFDA, PFUnDA, PFDODA, and PFTrDA. The highest levels were found for PFOS (30.28 ppb), PFOA (24.88 ppb), and PFNA (13.19 ppb). Mothers who had given birth previously showed lower levels of PFHxS, PFOS, PFOA, PFNA, and PFDA than first-time mothers. Women who delivered at an older age had higher PFUnDA levels. There was a positive association between maternal education and PFOA levels and higher annual household income was associated with higher PFOS and PFOA levels.

Miura R, Araki A, Miyashita C, Kobayashi S, Kobayashi S, Wang SL, Chen CH, Miyake K, Ishizuka M, Iwasaki Y, Ito YM, Kubota T, Kishi R (2018) An epigenome-wide study of cord blood DNA methylations in relation to prenatal perfluoroalkyl substance exposure: The Hokkaido study, Environ Int 115: 21-28
This study investigated DNA methylation changes in cord blood associated with prenatal exposure to PFAS in 497 women. Median concentrations of PFOS and PFOA in maternal blood were 5.2 ng/ml and 1.4 ng/ml respectively. The results showed shifts in methylation as a result of PFAS exposure; increased methylation from PFOS and decreased methylation from PFOA. The authors note that methylation changes might link health outcomes to PFAS exposure possibly due to their occurrence in various signaling pathways.

Fujii S (2017) The survey and risk assessment of organofluoro compounds in lake Biwa, Annual Report, Lake Biwa-Yodo River Water Quality Preservation Organization
To survey contamination of unknown fluorinated compounds in water of Lake Biwa and the Yodo River, total organic fluoride and perfluoro compounds (PFAS) were examined from 2016 to 2017. In 54 water samples, 12 PFAS substances were detected and their mean concentrations were 15.4% of total organic fluoride. This suggested that contamination with unknown total organic fluoride was higher than accounted for by PFAS by one order of magnitude. In the water of Lake Biwa, the concentrations of 12 PFAS averaged 11.9 ng/L with a median of 12.2 ng/L. In 25 water samples from the Yodo River, the highest concentrations of PFOS, PFOA, and PFHxA were 20.9 ng/L, 103.9 ng/L, and 41.1 ng/L, respectively. In river samples, the total concentration of 12 PFAS was 103.9 ng/L. The effects of PFAS on freshwater algae were also examined.

Goudarzi H, Miyashita C, Okada E, Kashino I, Chen CJ, Ito S, Araki A, Kobayashi S, Matsuura H, Kishi R (2017) Prenatal exposure to perfluoroalkyl acids and prevalence of infectious diseases up to 4years of age, Environ Int 104: 132-138

Prenatal exposure to PFAS and subsequent prevalence of infectious disease up to four years old was examined in 1558 mother-child pairs in the Hokkaido cohort. PFAS with high rates of detection were PFHxS, PFOS, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, and PFTrDA with PFOS showing the highest median exposure levels (4.92 ng/ml), followed by PFOA (2.01 ng/ml), PFUnDA (1.43 ng/ml), and PFNA (1.18 ng/ml). Note that these levels are significantly higher than drinking water restrictions in some US states. For example, a median PFOS level of 4.92 ng/ml is 4.92 ppb or 4,920 ppt compared to a drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US state of Vermont. In total, 1046 children had at least one infectious disease (67.1%). Diseases included otitis media, 649 (41.6%); pneumonia, 287 (18.4%); respiratory syncytial virus infection, 197 (12.6%); and varicella (chickenpox), 589 (37.8%). Prenatal exposures to PFHxS and PFOS through the mother, were associated with a higher risk of infectious diseases in early life in their children, suggestive an impact on immune system function observed in other studies.

Minatoya M, Itoh S, Miyashita C, Araki A, Sasaki S, Miura R, Goudarzi H, Iwasaki Y, Kishi R (2017) Association of prenatal exposure to perfluoroalkyl substances with cord blood adipokines and birth size: The Hokkaido Study on environment and children's health, Environ Res 156: 175-182

This study examined associations between prenatal exposure to PFOS and PFOA with cord blood adipokines and birth size. Adipokines are cell signaling proteins secreted by fat tissue. in A total of 168 women and their children participated in the study. Median levels of PFOS and PFOA in the mothers were 5.1 ng/ml and 1.4 ng/ml respectively. PFOS was positively associated with adiponectin level but negatively associated with the ratio of height to weight. PFOA was negatively associated with birth weight and had a small negative impact on the ratio of height to weight. The authors state that the data suggests that, “PFOS exposure may associate with disruption of fetal metabolic function” and “prenatal PFASs exposure may alter cord blood adiponectin levels and may decrease birth size.” Previous studies have indicated that reduced birth weight is a risk factor for a range of metabolic problems including high adult BMI, insulin resistance, increased visceral adiposity, and impaired glucose tolerance.

Kobayashi S, Azumi K, Goudarzi H, Araki A, Miyashita C, Kobayashi S, Itoh S, Sasaki S, Ishizuka M, Nakazawa H, Ikeno T, Kishi R (2017) Effects of prenatal perfluoroalkyl acid exposure on cord blood IGF2/H19 methylation and ponderal index: The Hokkaido Study, J Expo Sci Environ Epidemiol 27: 251-259

DNA methylation is an epigenetic process that plays a role in development and differentiation. This study examined the effect of PFOS and PFOA exposure on levels of insulin-like growth factor in cord blood and DNA methylation and its association with birth size. A total of 177 mother-infant pairs participated in the study. The results showed that PFOA was associated with a significant decrease in IGF2 methylation in cord blood. This could act indirectly to reduce the height to weight ratio (ponderal index). Previous studies have reported that reduced IGF2

methylation in cord blood and placental tissues is correlated with reduced fetal growth such as lower birth weight or small-for-gestational age.

Goudarzi H, Araki A, Itoh S, Sasaki S, Miyashita C, Mitsui T, Nakazawa H, Nonomura K, Kishi R (2017) The Association of Prenatal Exposure to Perfluorinated Chemicals with Glucocorticoid and Androgenic Hormones in Cord Blood Samples: The Hokkaido Study, Environ Health Perspect 125: 111-118

A total of 185 mother-infant pairs participated in this study to examine the relationship between prenatal exposure to PFOS and PFOA and cord blood levels of glucocorticoid and androgenic hormones such as DHEA. Median levels of PFOS and PFOA in the mothers were 5.1 ng/ml and 1.4 ng/ml respectively. The results showed that prenatal PFOS levels were inversely associated with cortisol and cortisone levels and positively associated with DHEA levels in cord blood. Prenatal exposure to PFOA was negatively associated with DHEA levels in cord blood. In addition, PFOS was negatively associated with the ratios of cortisol/DHEA and glucocorticoid / androgenic hormones but positively associated with the cortisol / cortisone ratio. In contrast, PFOA showed a positive nonsignificant association with cortisol to DHEA ratio, and the glucocorticoid to androgenic hormones ratios. The authors note that steroid hormone concentrations play a critical role in development. For example, glucocorticoids have a crucial role in lung and heart maturation in the fetus, and insufficient or excess amounts of these hormones have lifelong adverse effects on the cardiovascular system. The authors note that the results suggest that disruption of glucocorticoid and DHEA homeostasis at birth is associated with PFAS exposure, "and this may have adverse effects on the HPA axis and steroid hormone homeostasis in later life" and "in utero PFC [PFAS] exposure may be a public health concern."

Hori H (2016) International Regulatory Trend and Development of Decomposition Techniques for Environmentally Persistent Organofluorine Compounds, Oleoscience. 16:111 - 118

Organofluorine compounds have been widely used in industrial and consumer applications, owing to their specific characteristics such as high thermal and chemical stability. After it became clear that some of them, particularly, perfluorooctanesulfonate (PFOS) and perfluorooctanoic acid (PFOA), bioaccumulate in the environment, international legal measures regarding the production, use, and import and export of several organofluorine compounds are being implemented. Hence, it is desirable to develop effective methodologies for decomposing these compounds to environmentally harmless species under mild conditions. If they could be decomposed to F^- ions by means of environmentally benign techniques, the well-established protocol for the treatment of F^- ions could be used: Ca^{2+} is added to the system to form CaF_2, which is a raw material for hydrofluoric acid. Thus, the development of such techniques would allow for the recycling of fluorine resource, the global demand for which is increasing. We describe herein the international regulatory trend for organofluorine compounds. We also describe our developed methodologies for efficient decomposition of cutting-edge fluorinated materials (ionic liquid and polymers) to F^- ions.

Itoh S, Araki A, Mitsui T, Miyashita C, Goudarzi H, Sasaki S, Cho K, Nakazawa H, Iwasaki Y, Shinohara N, Nonomura K, Kishi R (2016) Association of perfluoroalkyl substances exposure in utero with reproductive hormone levels in cord blood in the Hokkaido Study on Environment and Children's Health, Environ Int 94: 51-59
This study explored associations between prenatal exposure to PFOS and PFOA and cord blood reproductive hormones. A total of 189 mother-infant pairs participated in the study and median levels of PFOS and PFOA in the mothers were 5.1 ng/ml and 1.4 ng/ml respectively. PFOS showed a positive association with levels of estradiol in male infants and negative association with the ratio of testosterone/estradiol and levels of progesterone and inhibin B (produced by the ovary). In contrast, among male infants, PFOA was positively associated with inhibin B levels. Among female infants, PFOS showed a negative association with progesterone and prolactin levels. The authors note that the results indicate that, "even low levels of PFOS and PFOA exposure can disrupt reproductive hormone imbalance in the fetus." In addition, they note that, "PFOS and PFOA, even at relatively low levels, has adverse effects on fetuses' synthesis of steroid hormones, their Leydig cell function, and their Sertoli cell function."

Kato S, Itoh S, Yuasa M, Baba T, Miyashita C, Sasaki S, Nakajima S, Uno A, Nakazawa H, Iwasaki Y, Okada E, Kishi R (2016) Association of perfluorinated chemical exposure in utero with maternal and infant thyroid hormone levels in the Sapporo cohort of Hokkaido Study on the Environment and Children's Health, Enivron Health Prev Med 21:334-344
This study examined the effect of PFOS sand PFOA on thyroid hormone levels important for development in 392 mother-infant pairs. Median maternal concentrations of PFOS and PFOA were relatively low at 5.7 and 1.2 ng/ml respectively. Maternal PFOS levels were associated with decreased thyroid stimulating hormone levels. PFOS exposure during pregnancy was also associated with higher levels of thyroid stimulating hormone in infants. The authors note that "such subclinical hyper- or hypothyroidism might lead to the imbalance of FT4 [free thyroxine], an important hormone for fetal neurodevelopment.

Goudarzi H, Nakajima S, Ikeno T, Sasaki S, Kobayashi S, Miyashita C, Ito S, Araki A, Nakazawa H, Kishi R (2016) Prenatal exposure to perfluorinated chemicals and neurodevelopment in early infancy: The Hokkaido Study, Sci Total Environ 541:1002-1010
This study examined associations between prenatal exposures to PFOS and PFOA and neurodevelopment of infants at 6 months (173) and 18 months (133). Median maternal concentrations of PFOS and PFOA were relatively low at 5.7 and 1.2 ng/ml respectively. PFOA exposure was associated with a significant decrease in neurodevelopment at 6 months in female infants, but not 18 months.

Kishi R, Nakajima T, Goudarzi H, Kobayashi S, Sasaki S, Okada E, Miyashita C, Itoh S, Araki A, Ikeno T, Iwasaki Y, Nakazawa H (2015) The Association of Prenatal Exposure to Perfluorinated Chemicals with Maternal Essential and Long-Chain Polyunsaturated Fatty Acids during Pregnancy and the Birth Weight of Their Offspring: The Hokkaido Study, Environ Health Perspect 123:1038-1045
This study estimated associations between PFOS and PFOA levels and maternal levels of fatty acids and triglycerides in 306 women and birth size of the offspring. Relatively low levels of

PFOS were associated with decreasing levels of triglyceride, palmitic acid, palmitoleic acid, oleic acid, linolenic acid, alpha linolenic acid, arachidonic acid and docosahexaenoic acid. The authors note that triglyceride and fatty acids are sources of energy for fetal development. Prenatal PFOS exposure was negatively associated with birth weight in female infants.

Eriksson U, Karrman A (2015) World-Wide Indoor Exposure to Polyfluoroalkyl Phosphate Esters (PAPs) and other PFASs in Household Dust, Eniviron Sci Technol 49: 14503-14511

PFAS precursor substances such as polyfluorinated phosphate esters (PAPs) were examined along with other PFAS in household dust samples from Canada, the Faroe Islands, Sweden, Greece, Spain, Nepal, Japan, and Australia. Mono-, di-, and triPAPs, including several diPAP homologues, were frequently detected in dust from all countries, revealing an ubiquitous spread in private households from diverse geographic areas, with significant differences between countries. Samples from Japan were collected in Kyoto. The median levels of monoPAPs and diPAPs ranged from 3.7 ng/g to 1 023 ng/g and 3.6 ng/g to 692 ng/g, respectively, with the lowest levels found in Nepal and the highest in Japan. The authors note that higher PAP dust levels in Canada and Japan are consistent with having fluorotelomer production, but since levels in Australia and Faroe Islands also had high PAP levels, that PAPs in dust may be more related to consumer products than fluorotelomer production. Dust from Japan also had the highest level of PFCAs (230 ng/g) and unlike other countries, PFNA was the predominating substance instead of PFOA.

Levels of PFAS in house dust from Japan

Substance	Mean Level (ng/g)	Maximum (ng/g)	Minimum (ng/g)
PFCA sum	239	355	118
PFSA sum	12	24	4
monoPAPs	1790	3738	<12
diPAPs	1415	4248	57
FTCA/FTUCA	0.6	2.2	<0.1
FOSA/FOSE	69	345	<0.1
6:2 FTSA	5.6	12	2
PFOSA	0.3	1.7	<1.4

PFCA – perfluoroalkyl carboxylates
PFSA – perfluoroalkyl sulfonates
PAP – polyfluoroalkyl phosphate esters
FTCA – saturated fluorotelomer carboxylic acids
FTUCA – unsaturated fluorotelomer carboxylic acids
FOSA – perfluoroalkane sulfonamides
FOSE – perfluoroalkane sulfonamidethanols
FTSA – fluorotelomer sulfonic acid
PFOSA – perfluorooctanesulfonamide

The study also estimated exposure due to ingestion of dust by adults and two-year-old children in high and low exposure scenarios. For the sum of PFAS, Japan had the highest exposure for adults (0.67 ng/kg body weight/day), 2-year-old children in a high exposure scenario (6.97 ng/kg body weight/day) and 2-year-old children in a low exposure scenario (0.29 ng/kg body weight/day). Japan also showed the highest estimated direct and indirect exposure (due to degradation of PAP substances) for adults and children for PFHxA, PFOA, PFDA, PFCAs, monoPAPs, and diPAPS.

Mochizuki E, Kobayashi H (2015) A Survey of Organofluorine Compounds in Environmental Water in Yamanashi Prefecture (Ⅱ), Annual Report, Yamanashi Institute for Public Health and Environment, 59: 44-46

In 2014, concentrations of PFAS in 8 water samples from several rivers were higher than those of other samples. In 2015, PFAS in 14 water samples were examined including the 8 samples mentioned above. The concentrations of PFAS in 2015 were almost same as those in 2014. Pollutant sources of PFBA might be from the sewage of an industrial park. Pollutant sources of PFOA and PFNA might be from total sewage.

Nishino T, Kato M, Shimoma S, Kitano M (2015) Perfluorinated Compounds in the Groundwater in Tokyo and their Behavior in Soils during Leaching Experiments, Journal of Environmental Chemistry, 25: 149-160

We measured 13 perfluorinated compounds (PFAS) in the groundwater in Tokyo. PFAS with nine carbons or fewer were frequently detected. PFOS and PFOA were detected at high concentrations, despite discharge reduction measures taken since 2010. Chromatograms of PFOS showed variations in the peak area ratio between linear-PFOS and branched-PFOS in various samples. To investigate the cause, leaching experiments were conducted for the 13 PFAS and branched-PFOS isomers. The results show that branched-PFOS penetrated the soil faster than linear-PFOS, and shorter chain PFAS faster than longer-chain PFAS. Long chain PFAS with more than 10 carbons were expected to remain in soil.

Ye F, Zushi Y, Masunaga S (2015) Survey of perfluoroalkyl acids (PFAAs) and their precursors present in Japanese consumer products**, Chemosphere 127:262-268**

This study examined 32 PFAS in well-known brands of consumers products purchased at stores, home centers, and on-line. Categories of products included car wash / coating products, sprays for fabrics and textiles, insecticides, rust inhibitors, and paints. The concentration ranges for each PFAS group were: <LOQ – 25000 ± 10000 ng/g for precursors of PFOS, <LOQ– 5400 ± 2100 ng/g for PFCAs, <LOQ–59 ± 10 ng/g for PFSAs, <LOQ–24 ± 14 ng/g for precursors of PFCAs, and <LOQ – 18 ± 4.9 ng/g for C6–PFPA. The highest PFAS concentration was 25,000 ng/g for N-ethyl perfluorooctane sulfonamidoethanol (EtFOSE), a PFOS precursor, in a spray for textiles. Sprays for textiles and car wash / coating products were the categories that contained PFAS levels greater than 1000 ng/g. PFAS were detected in two out of three rust inhibitor products, including PFPeA. PFAS were not detected in paints or insecticides. The PFAS substance with the highest detection frequency was PFNA (16%). PFNA also had the second highest average concentration of 220 ng/g. The authors suggest that PFNA may have been substituted for PFOA in Japan.

Shirasaka H, Kadokami K (2014) Accumulation and Spatial Distribution of Perfluorinated Compounds in Crucian Carp (Carassius auratus (gibelio) langsdorfii) in Japan, Journal of Environmental Chemistry 24: 67-76

Perfluorinated compounds (PFCs) that are widely used as surfactants and coatings were determined in muscles of crucian carp (Carassius auratus (gibelio) langsdorfii) taken from 14 freshwater areas throughout Japan during 2003-2005. The sampling sites comprised 10 rivers and 4 ponds and were categorized into 4 groups based on local circumstances: large cities, small cities, agricultural areas and remote areas. PFAS were detected in all samples analyzed, and total concentrations ranged from 1.60 to 30.1 (average: 9.54, median: 8.80) ng/g wet wt. and 167 to 3496 (average: 914, median:645) ng/g lipid wt. The highest concentrations of PFCs were found in fish caught at sites in large cities. Fish in remote area's had low PFAS concentrations. Branched chain PFAS isomers were also detected, although the relative ratios of the straight chains to the branch isomers were different between sampling sites. To obtain maternal transfer rates for the PFAS substances, female fish were collected from the Murasaki River during the spawning season and their muscles and eggs were analyzed. The maternal transfer rate of PFAS was 9.1%, which is lower than those of hydrophobic substances such as dioxins, organochlorine pesticides and hexabromocyclododecanes. As a result, sexual differences between male and female fish were not found.

Tabara R, Yamaguchi T (2014) Deposition of Perfluorinated Chemicals during Winter in Hokkaido, Journal of Environmental Chemistry, 24: 1-17

A survey of deposition of perfluorinated chemicals (PFAS) during winter was performed in rural areas and hinterlands in Hokkaido (9 points). The depositions were calculated from the concentrations of PFAS in snow cover. The PFAS concentrations were 2.3 ~ 12 ng/kg. PFBA was the dominant compound, and PFOA and PFNA were also detected. The deposition amounts and amount rates of PFAS were 0.38 ~ 8.6 $\mu g/m^2$ and 5.7 ~ 95 ng/m^2/day, respectively. They were high in the area along the Sea of Japan. The concentrations of some of PFAS were higher than those in the snowfall in the metropolitan area. The result shows not only wet deposition but dry deposition and/or chemical reactions of precursors also contributed to the deposition during the winter.

Mochizuki E, Kobayashi H, Minai Y (2014) A Survey of Organofluorine Compounds in Environmental Water in Yamanashi Prefecture, Annual Report, Yamanashi Institute for Public Health and Environment, 58: 32-37

In 2014, the contamination of 16 PFAS in the water of 17 rivers and 5 lakes in Yamanashi Prefecture was measured. PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTrDA, PFTeDA, PFBS, PFHxS, PFHpS, PFOS, and PFDS were monitored. The water samples were collected from surface, underground, and underflow water. Several kinds of PFAS were detected in 15/28 samples of surface water, in 8/16 samples of underground water, and in 2/2 samples of underflow water. PFOS, PFBA, PFOA, and PFNA were detected especially frequently and their concentrations were higher than those of other PFAS. In 28 samples of surface water, the results of PFOS, PFOA, PFBA, and PFNA were as follows. PFOS was detected in 4 samples at levels ranging from 1.1-3.2 ng/L. PFOA was detected in 10 samples at levels ranging from 0.5-3.1 ng/L. PFBA was detected in 7 surface water samples at levels

ranging from 0.51-2.2 ng/L. PFNA was detected in 8 samples at levels ranging from 0.50-3.3 ng/L.

Nishino T, Kato M, Ueno K, Kitano M (2014) Perfluorinated Compounds in the Groundwater in Tokyo and their Behavior, Annual Report, Tokyo Metropolitan Research Institute for Environmental Protection, p.52-58

Thirteen perfluorinated compounds (PFAS) were measured in Tokyo groundwater. The samples were collected at 58 points in 2013. The highest concentration of PFOS was 150 ng/L in one sample from the Tama River. PFAS were also measured in one kind of sea fish (Lateolabrax japonicus) from the Tokyo Bay area. In 2003 and 2012, PFOS in sea fish was detected at levels of 6000 ng/kg and 1000 ng/kg, respectively. The concentration of PFOS in the fish was rather high but decreasing by year.

Kuroda K, Murakami M, Oguma K, Takada H, Takizawa S (2014) Investigating sources and pathways of perfluoroalkyl acids (PFAAs) in aquifers in Tokyo using multiple tracers, Sci Total Environ 488-489: 51-60

This study examined sources and pathways of PFAS pollution in 53 groundwater samples from aquifers in Tokyo and 5 samples of tap water. Thirty-two samples were from unconfined aquifers, 3 were from springs, and 18 were from confined aquifers. The study showed widespread PFAS pollution in groundwater with PFAS found in 96% of samples. The most prevalent PFAS were PFHxS, PFOS, PFHpA, PFOA, PFNA, and PFDA as they were found in more than 77% of samples. PFAS were even found in springs and confined aquifers.

Substance	Groundwater (ng/L)	Tap water (ng/L)
PFHxS	<0.25-56	0.68-2.7
PFHpS	<0.25-10	<0.25
PFOS	<0.25-990	1.7-11
PFDS	<0.25-4.3	<0.25
PFHpA	<0.25-61	1.0-4.6
PFOA	<0.25-1800	4.7-12
PFNA	<0.25-620	3.6-8.5
PFDA	<0.25-34	0.32-0.75
PFUnDA	<0.25-28	<0.25
PFDoDA	<0.25-7.6	<0.25
PFTrDA	<0.25-5.9	<0.25
PFTeDA	<0.25-1.6	<0.25
FOSA	<0.25-96	0.48-1.3

The highest levels in tap water exceed the drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US state of Vermont (38.8 ppt for these substances). Due to the high PFAS pollution found in groundwater, the authors warn that, “care should be taken if those groundwaters are directly used for drinking purposes, landscaping or replenishing rivers.”

Sewage was inferred to be a major PFAS source in some groundwater samples due to correlation with tracer compounds for sewage. However, some groundwater samples had a PFAS distribution more similar to that found in rivers and secondary effluents.

The authors note that the ubiquitous presence of PFAS in all types of aquifers, "showed the high vulnerability of Tokyo's aquifers to pollution by PFAAs [PFAS]."

Miyake Y, Kobayashi T, Kameya T, Managaki S, Amagai T, Masunaga S (2014) Comparison study on observed and estimated concentrations of perfluorooctane sulfonate using a fate model in Tokyo Bay of Japan, J Environ Sci Health A Tox Hazard Subst Envion Eng 49: 770-776
PFOS pollution in seawater (surface, intermediate, and bottom) and sediment samples from the Tokyo Bay were compared with a fate model used to estimate concentrations based in fluxes from the major rivers that flow into Tokyo Bay (Edo River, Naka River, Ara River, Sumida River, Tama River, and Tsurumi River). PFOS was found in all samples. Surface seawater levels ranged from 2.0 – 7.3 ng/L. Middle seawater levels ranged from 2.2 – 5.7 ng/L. Bottom seawater levels ranged from 1.5 – 3.4 ng/L. Sediment levels ranged from 0.3 – 0.9 ng/g. The highest levels were observed in samples from the mouth of the Naka, Ara, Sumida and Tama Rivers.

Takata M, Yamamoto S, Nishioka R, Takemine S, Tanaka S, Fujii S, Watanabe N (2013) Behavior of Perfluorinated Compounds Adsorbed in Exhausted Activated Carbon during Thermal Treatment and Combustion, Japan Society of Material Cycles and Waste Management: 24: 105 - 112
Activated carbon adsorbs perfluorinated compounds (PFAS) in an effluent treatment system at a final disposal site. We examined activated carbon for inorganic fluorine (F) trapped in the gas absorbent obtained by thermal treatment in air followed by combustion decomposition in oxygen flow. The F yields increased as the air temperature rose to 600℃. The combined yields from combustion pyrolysis in oxygen flow were 215 μg/g. However, yields decreased at temperatures of 700℃ or greater, probably because of alkaline substances such as calcium carbonate on the activated carbon surface holding F on the solid-phase because of the thermal treatment temperature. During the experiments, F measurements were conducted using activated carbon washed in distilled water or dilute sulfuric acid. Results show that with distilled water cleaning, F yields decreased with increasing heat, but with dilute sulfuric acid cleaning, measurements of 106-114 μg/g remained unchanged at 500-800℃. This portion of the total amount of PFAS trapped in activated carbon exhibits about 10 times the concentration of the result of possible PFAS identification (5-14 μg/g) sampled at the same point.

Tabara R (2013) The survey of organofluorinated chemicals in Hokkaido, Annual Report, Hokkaido Research Organization, Environmental and Geological Research Department, Institute of Environmental Sciences, 3: 41-51
The concentrations of perfluorinated chemicals (PFAS) in rivers in Hokkaido were determined. Remarkable contamination by PFAS substances was not found in the surveyed rivers. The detected compounds and their concentrations were different in each area. In the Ishikari River, the PFAS concentrations were higher than in the other rivers. Among the PFAS, high molecular perfluorocarboxylic acids were detected in especially high concentrations, but

perfluorosulfonates were rarely fire extinguisherdetected. In contrast, in the Chitose River and the Tonebetsu River, various PFAS including perfluorosulfonates were detected. PFOS and/or PFHxS were detected around airports, probably due to the use of s. PFAS contamination from industrial facilities was also detected in paper mill effluents. These compounds are used in surface treatment of paper products.

From 2010 to 2012, river water samples at 26 points were assayed for PFNS, PFOS, PFHpS, PFHxS, PFBS, PFUdA, PFDA, PFNA, PFOA, PFHpA, PFHxA, and PFPeA. Monitoring was performed in major two rivers (Ishikawa, Chitose) and tributaries, and minor rivers near two airports area (Asahikawa, Memanbetsu).

Motegi M, Nojiri K, Hosono S, Sugisaki M (2013) An Initial Survey of Perfluorooctanesulfonate (PFOS) and Perfluorooctanoic Acid (PFOA) in River Waters in Saitama Prefecture, Journal of Environmental Laboratories Association, 38: 16-22
The concentrations of PFOS and PFOA in 38 samples from of 35 rivers in Saitama Prefecture were determined, from 2006 to 2007. The geometric means of PFOS and PFOA were 15 ng/L and 7.7 ng/L, and their highest concentrations were 5100 ng/L and 500 ng/L, respectively.

Nishino T, Ueno T, Takahashi A, Takazawa Y, Shibata Y, Nakama S, Kitano M (2013) Perfluorinated Compounds in Tamagawa River Basin – Trend of Contamination and Mass Flow, Journal of Environmental Chemistry, 23:177-186
Thirteen kinds of perfluorinated compounds (PFAS) in the mainstream of the Tamagawa River and its inflows (tributaries, and effluents from sewage treatment plants) were analyzed, and the loads of PFOS, PFOA and other 4 compounds were evaluated. The concentrations of PFOS and PFOA were found to be much lower than those of 2005 since 2009. This result indicates that the Stockholm Convention on Persistent Organic Pollutants (POPs) and the 2010/2015 PFOA Stewardship Program are effective. The cumulative load of PFAS that was accumulated in the inflows sequentially from the Nagata Bridge, as the uppermost point in this study, closely resembled the measured load at each sampling point in Tamagawa River. These results indicate that PFAS were scarcely degraded or volatilized, during flow down the river. The ratio of PFAS with longer chain length such as PFUnDA and PFTrDA in sediment samples were much higher than those in water samples.

Okada E, Kashino I, Matsuura H, Sasaki S, Miyashita C, Yamamoto J, Ikeno T, Ito YM, Matsumura T, Tamakoshi A, Kishi R (2013) Temporal trends of perfluoroalkyl acids in plasma samples of pregnant women in Hokkaido, Japan, 2003-2011, Environ Int 60: 89-96
This study examined prenatal exposure to 11 PFAS substances in Hokkaido between 2003 and 2011 in 150 pregnant women. PFOA, PFNA, PFDA, and PFUnDA, and PFOS were found 100% of the women in 2003, 2005, 2007, and 2009. In 2011, significant detection frequencies of other PFAS were detected including PFHxA (20%), PFHpA (50%), PFDoDA (97%), PFTrDA (97%), PFTeDA (13%), and PFHxS (77%). Between 2003 and 2011, plasma concentrations of PFOS and PFOA decreased, but PFNA and PFDA concentrations increased by 4.7% and 2.4% per year respectively.

Yamaguchi M, Arisawa K, Uemura H, Katsuura-Kamano S, Takami H, Sawachika F, Nakamoto M, Juta T, Toda E, Mori K, Hasegawa M, Tano M, Shima M, Sumiyoshi Y, Morinaga K, Kodama K, Suzuki T, Nagai M, Satoh H (2013) Consumption of seafood, serum liver enzymes, and blood levels of PFOS and PFOA in the Japanese population, J Occup Health 55: 184-194

This study examined the connection between fish consumption and PFOS and PFA levels in the Japanese population. A total of 608 people participated in the study, distributed into five regional blocks: Hokkaido/Tohoku, Kanto/Koshin-estu, Tokai/Hokuriku/Kinki, Chugoku/Shikoku, and Kyushu/Okinawa. Participants were also distributed among three different types of areas: urban (269), farming villages (135), and fishing villages (204).

Median levels of PFOS and PFOA were 5.8 ng/ml and 2.1 ng/ml respectively. Men had significantly higher blood levels of PFOS than women. In both men and women, PFOS levels increased with age. PFOS levels were highest in residents of fishing villages, followed by urban areas and farming villages. PFOA did not vary among the different types of residential locations. PFOS levels were highest in the Tokai/Hokuriku/Kinki region. The authors cite studies showing PFAS pollution in river water from manufacturing in this region and levels of PFOS and PFOA in tap water more than five times higher in the Kinki region than in the Tohoku region.

The frequency of eating boiled fish in broth, sliced raw fish, and coastal fish showed significant positive correlations with PFOS, but not with PFOA. Blood levels of PFOS and PFOA were positively correlated with the serum levels of hepatic enzymes.

Sakurai T, Kobayashi J, Kinoshita K, Ito N, Serizawa S, Shiraishi H, Lee JH, Horiguchi T, Maki H, Mizukawa K, Imaizumi Y, Kawai T, Suzuki N (2013) Transfer kinetics of perfluorooctane sulfonate from water and sediment to a marine benthic fish, the marbled flounder (Pseudopleuronectes yokohamae), Environ Toxicol Chem 32: 2009-2017

The marbled flounder ((Pseudopleuronectes yokohamae) is a benthic fish and this study examined the transfer of PFOS from water, suspended sediment and bottom sediment to the fish. S1. Dissolved and particulate PFOS concentrations during the exposure period were relatively constant and averaged 74 ng/L and 18 ng/L, respectively. Most PFOS (37-66%) was found in the carcass i.e. body minus muscle and internal organs. PFOS in water, suspended sediment and bottom sediment all contributed to PFOS body burden in flounder.

Taniyasu S, Senthilkumar K, Yamazaki E, Yeung LW, Guruge KS, Kannan K, Yamashita N (2013) Perfluoroalkyl substances in the blood of wild rats and mice from 47 prefectures in Japan: use of samples from nationwide specimen bank, Arch Environ Contam Toxicol 65: 149-170

This study used rats as sentinels of human exposure because they are mammals and live in close proximity to humans. Blood samples (216) as well as surface water from habitats in 47 prefectures were examined. Thirteen PFAS were found in blood with the following detection rates: PFDS (28%), PFOS (100%), PFHxS (31%), PFOSA (83%), N-EtFOSAA (53%), PFTeDA (41%), PFDoDA (95%), PFUnDA (100%), PFDA (100%), PFNA (100%), PFOA (95%), PFBA (36%), and 7:3 FTCA (55%). Lower PFAS concentrations were found in Kyushu and Shikoku islands. In contrast, islands with greater industrial activity such as Honshu contained higher

levels and the highest levels were found in the Kanto and Kinki regions of Honshu. The Kanto region of Honshu is the largest industrial zone including electronics semi-conductor production which uses PFAS substance. Levels of PFOS, PFOSA, PFNA, and PFOA in rats were positively correlated with levels of these substances in humans and levels of PFOS and PFOA in rats were similar to those observed in humans.

Imai S, Kawanaka Y Tsuchiya Y, Yun SJ (2012) Concentrations and Composition Profiles of Perfluorinated Organic Compounds in Tap Water in Tokyo, Journal of Japan Society on Water Environment, 35: 57-64

The concentrations of 11 perfluoroalkyl carboxylic acids (PFCAs) and 5 perfluoroalkyl sulfonates (PFAS) in the tap water samples in Tokyo were determined using liquid chromatography-tandem mass spectrometry. The total concentrations of perfluoro organic compounds (PFCs; PFCAs + PFASs) in the tap water samples collected from 40 sampling points ranged from 0.72 to 95 ng/L, with a mean value of 19 ng/L. Twelve PFAS substances were detected in the tap water samples with the highest concentrations. The Tokyo tap water samples were divided into two main groups by cluster analysis on the basis of PFC composition profile, i.e., the Tama district and Tokyo wards. The compositions and concentrations of PFAS in the tap water samples from Tokyo wards were similar. On the other hand, there was variability among those in the tap water from the Tama district. This was likely because groundwater in addition to river water was used as raw water in the Tama district. The authors state that compared with the guideline values for PFCs and the health-based values for PFCs in the literature, the individual PFC concentrations found in the tap water samples in this study were low.

Nishino T, Ueno K, Takahashi A, Nakama S, Kitano M (2012) The Environmental Survey on Perfluorinated Compounds in the Tama River Basin, Annual Report, Tokyo Metropolitan Research Institute for Environmental Protection, p.3-8

Perfluorinated compounds (PFAS) in the mainstream of the Tama River and its inflows (tributaries, and effluents from sewage treatment plants) were analyzed, and the loads of PFOS, PFOA and 4 other compounds were evaluated. The concentrations of PFOS and PFOA were lower than in 2005 since 2009. This result indicates that the Stockholm Convention on Persistent Organic Pollutants (POPs) and the 2010/2015 PFOA Stewardship Program are effective. The loads of PFHxS, PFHxA and PFNA were almost comparable to those of PFOS and PFOA. At each point in the mainstream of the Tama River, the cumulative load of PFAS agreed with the measured load. It demonstrated that PFAS in the Tama River were brought by the inflows.
Water in the river: PFOS levels ranged from ND-12 ng/L (9 samples); PFOA levels ranged from trace-9.2 ng/L (9 samples).
Effluents from Sewage Treatment Plants: PFOS levels ranged from 2.4-28 ng/L (6 samples); PFOA levels ranged from 6.4-17 ng/L (6 samples).

Okada E, Sasaki S, Saijo Y, Washino N, Miyashita C, Kobayashi S, Konishi K Ito YM, Nakata A, Iwasaki Y, Saito K Nakazawa H, Kishi R (2012) Prenatal exposure to perfluorinated chemicals and relationship with allergies and infectious diseases in infants, Environ Res 112: 118-125

This study examined the relationship between prenatal exposure to PFAS and allergies and infectious diseases infants. PFOS and PFOA levels in 343 pregnant women and IgE in cord serum in 231infants were measured. In women, PFOS (1.3 – 16.2 ng/ml) was found in all

samples and PFOA in 93% of them (below detection limit – 5.3 ng/ml). Measured allergies and diseases included food allergy, 57 (16.6%); eczema, 37 (10.8%); wheezing, 33 (9.6%); otitis media, 61 (17.8%); chicken pox, 16 (4.7%); bronchitis, 9 (2.6%); RSV diseases, 7 (2.0%); rhinitis, 6 (1.7%); pneumonia, 6 (1.7%); skin infection, 5 (1.5%); and other virus infections (rotavirus, adenovirus, and cytomegalovirus), 15 (4.4%).

Levels of IgE in cord serum decreased significantly with high maternal PFOA concentration among female infants. No significant associations were observed between maternal PFOS or PFOA levels and food allergy, eczema, wheezing, and otitis media. The authors note the difficulty of obtaining diagnoses of allergic disease such as wheezing in infants from the birth to 18 months. The authors also note that the sample size was insufficient for identification of significant relationships between PFAS exposure and allergies and infectious diseases, given the low number of cases in the cohort with these conditions.

Uebori M, Shimizu T, Oyama K (2011) The environmental survey on contamination of perfluorinated compounds (PFCs) in Osaka Prefecture, Bull. Res. Inst. Env. Agr. Fish Osaka, 4: 1-8

A survey of the concentration of perfluorinated compounds (PFAS) in water such as rivers and sea, and ambient air of Osaka Prefecture has been conducted from 2007 to 2010. Fourteen PFAS were targeted. In river water, the average concentration of PFOA was 160 ng/L in July, 2007. In 2008, the average concentration was reduced by roughly one-half. In the Kanzaki River, the highest concentration of PFOA was 600 ng/L in 2007, and decreased to 80 ng/L in 2010. On the contrary, PFHxA as an alternative substance of PFOA increased. The increasing tendency of PFHxA concentration was observed in Osaka Bay. It indicated that contamination of PFHxA reached the sea from the Kanzaki River. In the atmospheric environment, PFOA showed the highest of concentration of PFAS in suspended particulate matter (SPM). The concentration level was in the range of 15 - 1800 pg/m^3, and varied daily. The concentration of PFNA correlated with that of SPM air pollutant was observed.

Takagi S, Adachi F, Miyano K, Koizumi Y, Tanaka H, Watanabe I, Tanabe S, Kannan K (2011) Fate of perfluorooctanesulfonate and perfluorooctanoate in drinking water treatment processes, Water Res 45: 3925-3932

This study investigated how drinking water treatment processes in Osaka affected levels of PFOS and PFOA. All samples of raw water contained PFOS (1.2 – 4.4 ng/L) and PFOA (10 – 42 ng/L). All sand filtered water samples contained PFOS (1.2 – 3.5 ng/L) and PFOA (12 – 34 ng/L). All ozonated water samples contained PFOS (1.1 – 3.4 ng/L) and PFOA (11 – 27 ng/L). The data indicates that sand filtration and ozonation did not remove PFOS or PFOA. PFOS was also found in 35 activated carbon filter samples (0.51 – 7.6 ng/L) and PFOA was found in 23 activated carbon samples (0.78 – 72 ng/L). The authors noted that older activated carbon was not effective at PFOS or PFOA removal. Since activated carbon at water treatment plants is commonly used for several years in Osaka, the authors note that current water treatment processes do not completely remove PFOS and PFOA. PFOS (1.3 – 3.7 ng/L) and PFOA (6.5 – 48 ng/L) were found in all finished water samples. In most water treatment plants, PFOS and PFOA levels were similar to or even higher than in raw water.

Nishikoori H, Murakami M, Sakai H, Oguma K, Takada H, Takizawa S (2011) Estimation of contribution from non-point sources to perfluorinated surfactants in a river by using boron as a wastewater tracer, Chemosphere 84: 1125-1132
River water samples were from the Iruma River, upstream of the intake of drinking water treatment plants in Tokyo, during dry weather and wet weather were used to estimate the contribution of non-point sources to PFAS pollution. The Iruma River joins the Ara River upstream of the water intake for Tokyo's drinking water.

PFHxA, PFOS, PFHpA, PFOA, PFNA, PFDA, PFUA, and PFDoDA were found in all samples. The maximum PFOA level during wet weather was 45.5 ng/L (ppt) – more than twice the drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US State of Vermont. The authors note that, "contamination of water by PFOA and PFNA during wet weather is a matter of concern."

Zushi Y, Ye F, Motegi M, Nojiri K, Hosono S, Suzuki T, Kosugi Y, Yaguchi K, Masunaga S (2011) Spatially detailed survey on pollution by multiple perfluorinated compounds in the Tokyo Bay basin of Japan, Environ Sci Technol 45: 2887-2893
The Tokyo Bay basin contains a population of 29 million, an area of 8000 km^2, and is highly industrialized and urbanized. This study examined pollution from 35 substances in water samples, including in rivers feeding into the bay and effluent samples from sewage treatment plants. PFOS and PFNA were dominant PFAS substances in the Tokyo / Kanagwa area; PFOA in Chiba; and PFNA in the Saitama area. In the entire Tokyo Bay basin, PFNA was the most prevalent PFAS substance. The authors note that this might indicate a shift in PFAS substance use due to restrictions on PFOS and the voluntary phase-out of PFOA. The data shows that levels of PFHxA, PFHpA, PFOA and PFNA were 10 – 100-fold higher near production plants than in other areas. PFCA levels in coastal water (491 ng/L) and plant effluent (6024 ng/L) were described as "extraordinarily high." Estimated loadings from the six major rivers into Tokyo Bay were 96.6 kg/year for PFOA and 139.6 kg/year for PFOS. The authors conclude that, "the introduction of PFC [PFAS] regulations on the use/production/emission has not reduced PFC pollution."

Raj Shivakoti B, Tanaka S, Fujii S, Hong Lien NP, Nozoe M, Kunacheva C, Okamoto R, Seneviratne ST, Tanaka H (2011) Perfluorinated compounds (PFCs) in Yodo River system, Japan, Water Sci Technol 63: 115-123
The Yodo River basin is a major economic and industrial zone of Japan with an area of 8,240 km^2 which includes Lake Biwa (largest lake in Japan) and the Ai River sub-basin. This study examined PFAS pollution in the Yodo River system, including wastewater treatment plant effluents. PFAS substances were found in sites – and in some them exceeding 1000 ng/L (ppt). Dominant PFAS substances were PFOS, PFHxS, PFOA and PFNA. The authors note that, "the river could be a continuous pathway of PFCs [PFAS] exposure to the people of Kansai region."

Guruge KS, Yeung LW, Li P, Taniyasu S, Yamashita N, Nakamura N (2011) Fluorinated alkyl compounds including long chain carboxylic acids in wild bird livers from Japan, Chemosphere 83: 379-384
This study examined PFAS pollution in the livers of 10 wild bird species from Miyagi and Tochigi prefectures. Eight of the birds are considered to be top predators. Species included swans (Cygnus Cygnus), common kestrel (Falco tinnunculus), northern goshawk (Accipiter gentilis), Japanese sparrowhawk (Accipiter gularis), Eurasian sparrowhawk (Accipiter nisus), ural owl (Strix uralensis), brown hawk owl (Ninox scutulata), mallard (Anas platyrhynchos), great egret (Ardea alba), and a cattle egret (Bubulcus ibis).

The dominant PFAS was PFOS which ranged from <0.25 - 210 ng/g wet weight and was found in all birds. Other PFAS found in all birds were PFUnDA, PFDA, PFNA, and 7:3 FTCA. PFTeDA and PFDoDA were found in 9 of the 10 birds, PFOA was found in 8 of them and PFHxDA in 7 of them.

In swans, dominant PFAS were PFOS (72%), 7:3 FTCA (66%), and PFNA (61%) and they were the least-contaminated birds in the study. In Japanese sparrowhawk 7:3 FTCA was another commonly detected PFAS substance. Mallards contained the lowest PFAS levels, reflecting a diet of grains and insects, compared with the other analyzed species which are top predators. The highest PFOS concentrations were found in the Japanese sparrowhawk, which was 210 ng/g wet weight. The authors note that PFAS levels observed in the study were below toxicological threshold values, however these values do not consider mixture effects, timing of exposure, and the age of the standards is not clear.

Benskin JP, Yeung LW, Yamashita N, Taniyasu S, Lam PK, Martin JW (2010) Perfluorinated acid isomer profiling in water and quantitative assessment of manufacturing source, Environ Sci Technol 44: 9049-9054
This study developed an isomer profiling method for PFAS substances to assess contributions from electrochemical (ECF) and telomer manufacturing processes. ECF manufacturing accounted for 94% of total PFOA in the Japan Sea site 1 (4.86 ng/L). Smaller contributions from ECF were observed in Tokyo Bay (33% - 1.76 ng/L), Tomakomai Bay site 1 (75% - 14.8 ng/L), and Japan Sea site 2 (74% - 2.93 ng/L). The findings are consistent with what is known about PFAS manufacturing in Japan. Small-scale ECF ammonium PFOA manufacturing took place in Japan from 1947 – 2002. In 1975, at least one site in Japan began manufacturing PFOA via direct oxidation of PFOI. Manufacturing of specific branched PFCA substances (presumably by telomerization) has reportedly also taken place in Japan since the mid-1970s.

Shivakoti BR, Tanaka S, Fujii S, Kunacheva C, Boontanon SK, Musirat C, Seneviratne ST, Tanaka H (2010) Occurrences and behavior of perfluorinated compounds (PFCs) in several wastewater treatment plants (WWTPs) in Japan and Thailand, J Environ Monit 12: 1255-1264
This study examined PFAS in wastewater treatment plants due to concerns that they can be a major pollution point source due to discharge of wastewater effluents, septic discharges or application of sludge to farmland. Eight plants were examined from the Kansai region in Japan. All of them receive a mixture of domestic and industrial wastewater and serve a population

ranging from 51,000 – 870,000. The plants drain into the Yodo River system – a drinking water source for people in the Kansai region.

Some PFAS substances were found in all samples, sometimes at very high levels of more than 1000 ng/L (ppt). The highest PFAS level among all plants examined from Japan and Thailand was from a plant in Japan (391 – 575 ng/L in liquid phase samples and 50 – 2343 ng/L in suspended solid samples). Data from different parts of plant operations showed that the plants failed to remove PFAS. The most detected PFAS substances in Japan were PFOA and PFNA. Daily discharges of PFAS from individual plants in Japan are in the range 0.35–55.9 g/d in Japan. The estimated total daily discharge of PFAS mass from plants in Japan is 125 g/d. This ultimately flows into the Yodo River, which serves as a drinking water source.

Techniques such as aerobic and anaerobic biological treatment, sand filtration, chlorination, ozonation, and activated carbon were all found to be ineffective at removing PFAS substances.

Sakurai T, Serizawa S, Isobe T, Kobayashi J, Kodama K, Kume G, Lee JH, Maki H, Imaizumi Y, Suzuki N, Horiguchi T (2010) Spatial, phase, and temporal distributions of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in Tokyo Bay, Japan, Environ Sci Technol 44: 4110-4115

This study investigated the spatial distribution, partitioning and time trends of PFOS and PFA in the water column and bottom sediment of Tokyo Bay in 480 water and 60 sediment samples. The median concentrations in sediment were 0.61 (PFOS) and 0.20 (PFOA) ng/g-dry weight. Study of levels vs. water salinity indicated that freshwater inputs into the Bay were the source of PFAS substances – particularly in the northern part of the Bay.

Zushi Y, Tamada M, Kanai Y, Masunaga S (2010) Time trends of perfluorinated compounds from the sediment core of Tokyo Bay, Japan (1950s-2004), Environ Pollut 158:756-763

Sediment core samples from Tokyo Bay were measured to reveal time trends of PFAS deposition between the 1950s and 2004. The levels of most PFAS substances increased with time. Measurements show that levels of PFOA, PFNA, PFOS, N-EtFOSAA, and N-MeFOSAA were high during the late 1950s – 1960s. PFOS decreased gradually in the early 1990s and N-EtFOSAA and N-MeFOSAA decreased rapidly in the late 1990s. PFOA increased rapidly from the late 1990s to 2004 (60% increase). The authors hypothesize that this could be due to the shift from PFOSF-based products toward telomer-based products. The study identified six trends:

- Rapid increase after the early 1970s for PFUnDA, PFDoDA, PFTrDA
- Gradual increase from the 1950s for PFNA and PFDA
- Unexpected peak in the later 1950s – 1960s and rapid increase after the late 1990s for PFOA
- Gradual increase from the 1950s and gradual decrease after the early 1990s for PFOS
- Unexpected peak in the late 1990s for precursors of PFOS such as N-EtFOSAA and N-MeFO-SAA. The authors note that this might represent releases from firefighting foams or discharge from manufacturing facilities.
- No distinct trend: FTCAs, FTUCAs, and 1H,1H,2H,2H-tetrahydroperfluorooctanesulfonate (THPFOS)

Ahrens L, Yamashita N, Yeung LW, Taniyasu S, Horii Y, Lam PK, Ebinghaus R (2009) Partitioning behavior of per- and polyfluoroalkyl compounds between pore water and sediment in two sediment cores from Tokyo Bay, Japan, Environ Sci Technol 43: 6969-6975

Sediment is a reservoir of pollutants and this study examined the portioning behavior of PFAS between pore water (water between grains of sediment) and sediment in the Tokyo Bay. Eleven PFAS substances were found in pore water including PFHxS, PFOS, TH-PFOS, N-EtFOSAA, PFBA, PFPeA, PFHxS, PFHpA, PFOA, PFNA, and PFDA. Previously reported surface water levels of PFHxS, PFOS, PFOA, PFNA were approximately 10-fold higher than the levels observed in this study for pore water. Shorter chain PFCAs were found exclusively in pore water. Statistically significant increased levels of PFAS in Tokyo Bay were found for PFOS (1956-2008), PFNA (1990-2008), and PFUnDA (1990-2008).

Harino H, Iwasaki N, Arai T, Ohji M, Miyazaki N (2009) Occurrence of antifouling biocides and fluorinated alkyl compounds in sediment core from deep sea: Suruga Bay, Tosa Bay, and Nankai tough, Japan, Arch Environ Contam Toxicol 57: 661-669

PFOS and PFOA were found in sediment cores from Suruga Bay (many surrounding industries) and Tosa Bay (low human activity and fertile fishing ground). Samples from Suruga Bay were taken at water depths of 800 m and 850 m. Samples from Tosa Bay were taken from 50, 100, and 200 meters. In the sample from Suruga Bay, PFOS was high in the subsurface sediment, then decreased. PFOA levels did not change throughout the sediment core. In Tosa Bay, high levels of PFOS were found in the subsurface sediment toward shallow water indicating scattered sources on land. PFOA levels were near detection limit. The authors note that the finding of PFAS in the deep-sea environment could be due to the coastal waters and "garbage such as plastic products, waste cans, and gum products are often observed in the deep-sea environment, and perfluoro organic compounds are considered to be eluted in this garbage."

Harada KH, Koizumi A (2009) Environmental and biological monitoring of persistent fluorinated compounds in Japan and their toxicities, Environ Health Prev Med 14: 7-19

This is a review article summarizing monitoring studies in Japan and the toxicities of PFAS substances. The review of monitoring studies shows the following:

- PFOA levels in Kansai surface water were much higher than other regions, due to a single PFOA source along the Ai River.
- PFOS levels are relatively high in the Kanto and Kansai regions.
- Sewage treatment plant effluents show high concentrations of PFOS and PFOA.
- High levels of PFOS were in wastewater around airports.
- PFOA levels in drinking water in Kansai are "extremely high, especially Osaka where the water supply is mainly derived from the Yodo River."
- PFOS levels in Kanto tap water were higher than in other regions.
- Concentrations of PFOA within urban atmospheric particles were 50-fold higher than those of PFOS. The amounts of PFOA and PFOS in the respirable fraction (1.1–11.4 um) ranged from 58.3 to 89.8% of the total amounts.

- The levels of PFOS and PFOA were significantly higher in the urban atmosphere of Oyamazaki than in the suburban atmospheres of Morioka and Fukuchiyama.
- Across Japan, there was a tendency for PFOA to be the predominant contaminant of outdoor air, particularly in Osaka.
- The concentrations in indoor dust in Japan ranged from18 to 3,700 ng/g dust for PFOA and from 7 to 2,500 ng/g dust for PFOS.
- Airborne levels of several FTOHs were significantly higher in the Kyoto-Osaka area than in other areas. This may be a source of airborne PFOA due to degradation of FTOHs.
- The estimated exposure through food was predominant for both PFOA and PFOS, followed by tap water and indoor dust.
- Residents belonging to the Kansai region (Kyoto, Osaka, and Hyogo) exhibited significantly higher serum PFOA levels.
- Serum PFOS levels in the Kansai region were significantly higher than those in the Tohoku and Chubu regions (Akita, Miyagi, Gifu, and Fukui) and comparable to those in Yamaguchi, Kochi, and Okinawa.
- Serum concentrations increased 3-fold for PFOS and 14-fold for PFOA between 1977 and 2003 in Yokote in Miyagi prefecture.
- The PFOA concentrations in Kyoto increased by 4.4-fold from 1983 to 1999.

The authors note that, "identification of the sources and appropriate control of the release of PFCs [PFAS] urgently require discussion."

Murakami M, Kuroda K, Sato N, Fukushi T, Takizawa S, Takada H (2009) Groundwater pollution by perfluorinated surfactants in Tokyo, Environ Sci Technol 43: 3480-3486

This study examined PFAS pollution in 16 groundwater and spring samples. PFAS were found in all samples and PFOS (0.28-133 ng/L), PFHpA (<0.1-20 ng/L), PFOA (0.47-60 ng/L), and PFNA (0.1-94 ng/L) were the predominant substances. PFOS was more abundant in groundwater than in rivers, and this could not be explained by wastewater or street runoff. One possibility is that sewer pipe leakage is pathway for groundwater pollution. The authors note that 13% of sewers in Tokyo (77% in Chuo Ward, 84% in Chiyoda Ward, 91% in Taito Ward) have exceeded their estimated service life. In Tokyo, wastewater and surface run-off are considered to be dominant sources of PFAS in water. The authors note that, "groundwater contamination by PFOS will continue unless the precursors are regulated." In addition, they observe that, ", since manufacturers are switching to shorter-chain PFCAs with <7 fluorinated carbons, these compounds could also contaminate groundwater."

Washino N, Saijo Y, Sasaki S, Kato S, Ban S, Konishi K, Ito R, Nakata A, Iwasaki Y, Saito K, Nakazawa H, Kishi R (2009) Correlations between prenatal exposure to perfluorinated chemicals and reduced fetal growth, Environ Health Perspect 117: 660-667

This examined possible associations between PFOS and PFOA in the blood of 428 pregnant women and birth weight and birth size in their infants. PFOS (1.2 – 16.2 ng/ml) was found in every woman and PFOA (ND – 5.3 ng/ml) in 92.8% of them. The study showed that prenatal exposure to relatively low levels of PFOS was negatively correlated with birth weight – even

though PFOS levels in serum were significantly lower than levels causing developmental effects in animal studies (at that time).

Tao L, Ma J, Kunisue T, Libelo EL, Tanabe S, Kannan K (2008) Perfluorinated compounds in human breast milk from several Asian countries, and in infant formula and dairy milk from the United States, Environ Sci Technol 42 :8597-8602
This study examined breast milk for PFAS in samples from Cambodia, India, Indonesia, Japan, Malaysia, Philippines, and Vietnam. In addition, infant formula from five US manufacturers and 11 brands of dairy milk were tested.

PFOS was found in 100% of the samples from Japan. PFOA and PFHxS were found in 92% of the samples; PFNA in 13% of them; and PFHpA in 25% of them. In fact, Japan showed the highest level of detection for PFOA, PFHxS (shared with Philippines) and PFNA (shared with Cambodia) and the second highest level of detection of PFHpA (after Cambodia).

Breast milk samples from Japan were collected from 24 women in Ehime Prefecture. The results showed significant PFAS levels for PFOS, PFOA, PFHxS, PFNA, and PFHpA. Overall, average PFOS levels in Japanese breast milk averaged 232 ppt – more than 10 times higher than the drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US State of Vermont. Average levels of PFOS in Japanese breast milk were the highest in all countries examined. The highest level of PFOS in Japanese breast milk (523 ppt) was more than 26 times higher than this drinking water health advisory limit.

PFAS levels in Japanese breast milk from Ehime Prefecture
n = 25 Overall mean PFOS level = 232 ppt

Substance	Range (ppt)	Fraction of samples containing (%)
PFOS	140 - 523	100
PFOA	<42.5 – 170	92
PFHxS	<1.66 – 18.2	92
PFNA	<8.82-23.9	13
PFBS	<1.11	0
PFHpA	<4.45-13.9	025

The study found that PFAS levels in US infant formula and dairy milk were approximately 10-fold lower than levels in Asian breast milk. This resulted in a daily intake of PFOS by Asian infants that was 7 – 12 times higher than the dietary intakes previously reported for adults in Canada, Germany and Spain.

Murakami M, Imamura E, Shinohara H, Kiri K, Muramatsu Y, Harrada A, Takada H (2008) Occurrence and sources of perfluorinated surfactants in rivers in Japan, Environ Sci Technol 42: 6566-6572
This study examined nine PFAS substances in 18 rivers and 5 wastewater secondary effluent samples. Two samples were selected from each of nine regions from these districts: Hokkaido,

Tohoku, Kanto, Hokuriku, Chubu, Kinki, Chugoku, Shikoku, and Kyushu. PFAS substances included PFOS, FOSA, PFHpA, PFOA< PFNA, PFDA, PFUA, PFDDA, and PFTDA.

PFAS were found in all rivers and more than half of them had levels higher than 40 ppt – twice the drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined in the US State of Vermont. Predominant PFAS were PFOS, PFHpA, PFOA, and PFNA. PFOS, PFHpA, and PFNA in rivers were strongly correlated with population density, indicating an urban source. PFOA levels were relatively high even in rivers with relatively small contributions of urban effluent (Tokorogawa River, Narusegawa River, Tamagawa River st. 1, Arakawa River, Abegawa River, Yuragawa River, and the Asahikawa River.) Correlation studies indicated that sewage effluent was the likely source of PFOS, PFHpA and PFNA. The total fluxes of sewage-derived PFAS were estimated to be: PFOS (3.6 t/year), PFHpA (2.6 t/year), PFOA (5.6 t/year), and PFNA (2.6 t/year). The authors note that these fluxes, especially for PFNA were remarkably high in Japan compared to rivers in Europe.

Murakami M, Takada H (2008) Perfluorinated surfactants (PFSs) in size-fractionated street dust in Tokyo, Chemosphere 74: 1172-1177
PFAS substances were measured in street dust collected from five heavily trafficked sites in Tokyo (Kamiuma, Narimasu, Nakagawa, Senju, and Tabashi-honcho). All sites had a traffic density of more than 38,000 vehicles per day. PFOS, PFOA, PFNA, PFDA, and PFUA were found in all samples. PFOS and PFOA levels ranged from <0.2 to 11 ng/g and 1.2 to 11 ng/g Respectively – higher than the levels in sediments in the Ariake Sea. PFAS levels were higher in heavily trafficked fine dust than residential dust, suggesting that traffic as a source of PFAS in dust. The authors suggest that PFAS in street dust could contribute to marine pollution.

Nakayama K, Iwata H, Tao L, Kannan K, Imoto M, Kim EY, Tashiro K, Tanabe S (2008) Potential effects of perfluorinated compounds in common cormorants from Lake Biwa, Japan: an implication from the hepatic gene expression profiles by microarray, Environ Toxicol Chem 26: 2378-2386
PFAS levels in the livers of wild cormorants from Lake Biwa were examined for impacts on gene expression. Cormorants are a fish-eating bird and a top predator in the Lake Biwa ecosystem. PFOS levels in liver ranged from 30 – 120 ng/g wet weight and PFOSA ranged from 7 – 22 ng/g wet weight. PFNA ranged from <5 – 43 ng/g and PFHxS and PFOA were not found. PFOS levels were significantly higher in male birds. The authors hypothesize that this might be due to maternal transfer of PFAS to eggs. Correlations indicated that male birds likely metabolized PFOSA into PFOS. PFAS levels also correlated with changes in expression of a wide variety of genes including those for antioxidant enzymes and molecular chaperones. Since molecular chaperones appeared to be down regulated by PFAS, the authors suggest that a result would be "accumulation of abnormal proteins or functional loss of proteins depending on molecular chaperones."

Takagi S, Adachi F, Miyano K, Koizumi Y, Tanaka H, Mimura M, Watanabe I, Tanabe S, Kannan K (2008) Perfluorooctanesulfonate and perfluorooctanoate in raw and treated tap water from Osaka, Japan, Chemosphere 72: 1409-1412
This study investigated PFAS pollution in raw and treated tap water from 14 water purification plants in Osaka. The raw water originated from lakes, rivers, groundwater, or subsoil water.

PFOS was found in all raw water samples at levels ranging from 0.26 – 22 ng/L (ppt). PFOA was found in all raw water samples at levels ranging from 5.2 - 92 ng/L (ppt). PFOS and PFOA also contaminated all tap water samples. PFOS was found at levels ranging from 0.20 – 22 ng/L (ppt). PFOA was found at levels ranging from 2.3 – 84 ng/L (ppt). The highest levels of PFOS and PFOA in tap water exceeded drinking water limits set in some US states. For example, Vermont sets a drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined. In many water treatment plants, the PFAS removal rate was less than 50%. Frequent changes of activated carbon yielded higher removal rates. At some plants, the effluent had even higher PFAS levels than the influent.

Guruge KS, Manage PM, Yamanaka N, Miyazaki S, Taniyasu S, Yamashita N (2008) Species-specific concentrations of perfluoroalkyl contaminants in farm and pet animals in Japan, Chemosphere 73: S210 – 215

This study investigated PFAS contamination in cattle (Japanese black, Holstein, F1), pigs, chickens, goats, horses, and dogs. PFOS was found in the blood of all animals at mean levels of: chicken (5.8 ng/ml), cattle (3.0 ng/ml), goat (2.4 ng/ml), horse (0.71 ng/ml), dog (25 ng/ml), and pig (0.37 ng/ml). Levels in dogs are similar to levels found in humans. PFOS was also found in the livers of all animals at mean levels of: chicken (67 ng/g), pigs (54 ng/g) and cattle (34 ng/g). Lower levels of PFHxS, PFDoDA, PFUnDA, PFDA, PFNA and PFOA were found in all animals. The levels of PFHxS, PFNA, and PFOA were highest in dogs – which live among humans and can have contact with carpets, floor polish vacuum cleaner dust and other possible PFAS-containing sources. The authors note that animal-derived products are widely used as protein concentrates for animal feed and that this might result in accumulation of PFAS in farm animals.

Zushi Y, Takeda T, Masunaga S (2008) Existence of nonpoint source of perfluorinated compounds and their loads in the Tsurumi River basin, Japan, Chemosphere 71:1566-1573

River water and sewage treatment plant effluent were sampled along the Tsurumi River to reveal sources of PFAS. The Tsurumi River flows through Yokohama and into Tokyo Bay. PFOS (17.1 – 612.2 ng/L), PFHxA (3.2 – 7.1 ng/L), PFHpA (4.9 – 7.7 ng/L), PFOA (11.2 – 19.8), PFNA (17.0 – 37.9 ng/L), and PFDA (2.1 – 4.3 ng/L) were all found in the river samples. At sewage treatment plants, PFOS (78.7-689.9 ng/L), PFHxA (3.5 – 9.4 ng/L), PFHpA (5.5 – 7.2 ng/L), PFOA (17.8 – 24.9), PFNA (27.5 – 41.8), and PFDA (3.0 – 4.5 ng/L) were found. The loads of various sewage treatment plants were: PFOS (0.36 - 15.10 g/hour), PFHxA (0.03 – 0.76 g/hour), PFHpA (0.03 – 0.37 g/hour), PFOA (0.11 – 1.14 g/hour), PFNA (0.21 – 1.33 g/hour), and PFDA (0.06 – 0.18 g/hour). The authors note that PFAS loads were higher during rainy weather, but rainwater itself should not contain significant PFAS levels. The authors hypothesize that the results indicate that significant portion of PFAS in the river could come from nonpoint sources such as PFAS-containing products.

Senthilkumar K, Ohi E, Wajwan K, Takasuga T, Kannan K (2007) Perfluorinated compounds in river water, river sediment, market fish, and wildlife samples from Japan, Bull Environ Contam Toxicol 79: 427-431

This study examined PFAS in river water (Kamogawa, Tenjingawa, Katsuragawa), sediment (Osaka), fish (from retail markets and trout from a fish farm in the Kanto region, sandfish from

the Ishikawa area, jack mackerel from Kyushu, sardine from Seto Inland Sea), cormorants, raccoon dog, large-bill crow, and eagle.

PFOS (<5.2 – 10 ng/L) and PFOA (7.9 – 110) were found in all river water samples. Other analyzed PFAS were not found in water but PFOSA and PFDoA were found in sediment along with PFOS and PFOA. PFOS, PFOA, PFOSA, and PFDoA were all found in fish livers. PFHxS, PFOS, PFOS, PFOSA, and PFDoA were found in cormorants, racoon dog, eagle, and large-bill crow. Cormorants showed the highest levels followed by eagle, raccoon dog and large-billed crow.

Nozoe M, Fujii S, Tanaka S, Tanaka H, Yamashita N (2006) Investigation of PFOS and PFOA in a Wastewater Treatment Plant, Environmental Engineering Research, 43:105-111

Few researchers have reported on influent or effluent load and the behavior of PFOS and PFOA, persistent fluorinated organic compounds, in wastewater treatment plants. In this study, water quality and quantity were investigated in a wastewater treatment plant (population: 570,000), in September 2005 and January 2006. The main conclusions are as follows: 1) More than 80% of PFOS and PFOA in an aeration tank were adsorbed to activated sludge and were likely accumulated in a wastewater treatment plant, 2) PFOS decreased 84% in an ultra-advanced treatment by ozonation and biological activated carbon and 3) the flux of PFOS and PFOA were 1.5 and 10.0 g/day in influent and 5.6 and 12.4 g/ day in effluent (flow: 185,000 m^3/day), respectively.

Nakata H, Kannan K, Nasu T, Cho HS, Sinclair E, Takemurai A (2006) Perfluorinated contaminants in sediments and aquatic organisms collected from shallow water and tidal flat areas of the Ariake Sea, Japan: environmental fate of perfluorooctane sulfonate in aquatic ecosystems, Environ Sci Technol 40: 4916-4921

This study investigated PFAS pollution in Ariake Sea tidal flats and shallow water and in aquatic organisms such as lugworm, mussel, crab, clam, oyster, mudskipper fish, filefish, bream, flounder, shark, finless porpoise, gull, and mallard duck. PFOS and PFOA were detected in most of the samples analyzed, but levels varied between shallow water and tidal flat organisms. PFNA, PFHS, and PFOSA were either undetected or present at low levels. In shallow water organisms, PFOS was the dominant pollutant followed by PFOA, PFNA, PFOSA, and PFHxS. In tidal flat organisms, PFOA was the dominant pollutant and levels were high in oysters, mussels, clams, and crabs.

High average PFOS levels were present in mallards (375 ng/g wet weight liver), gulls (301 ng/g wet weight liver) and finless porpoises (278 ng/g wet weight liver). Finless porpoises also contained significant levels of PFNA (8.3 – 44 ng/g wet weight) and PFOSA (<1.5 – 10 ng/g wet weight). Mean PFOS concentrations in shallow water fish ranged from 0.84-65 ng/g wet weight and the highest concentration was found in right eye flounder (2.5 – 171 ng/g wet weight in liver). PFOA was found in shallow water species, at concentrations ranging from <3.0 to 15 ng/g.

The authors note that elevated concentrations in higher trophic level organisms suggest the biomagnification of this compound through the marine food chain.

Hori H, Nagaoka Y, Yamamoto A, Sano T, Yamashita N, Taniyasu S, Kusuna S, Osaka I, Arakawa R (2006) Efficient decomposition of environmentally persistent perfluorooctanesulfonate and related fluorochemicals using zerovalent iron in subcritical water, Environ Sci Technol 40:1049-1054

The development of techniques to destroy PFAS substances is very important for dealing with current stockpiles and contaminated sites. This study investigated methods surrounding use of subcritical water to destroy PFOS (water held by pressure at a temperature higher than its 100C boiling point). A PFAS-containing antireflective coating agent used in Japanese semiconductor manufacturing was the test subject.

At 350C for six hours in subcritical water, most PFOS remained. Aluminum did not enhance decomposition. However, other metals did enhance decomposition with iron being most effective. Treatment in subcritical water with 9.6 mmol iron at 350C resulted in no PFOS being detected in the reaction solution. However, recovered iron powder showed carbon-fluorine bonds present in the iron surface, indicating strong adsorption. Fifteen hours of treatment in subcritical water at 350C achieved complete destruction. The reaction also created hydrogen gas.

The method was also applied to shorter chain PFAS with 3.61-4.67% remaining after the reaction.

Morikawa A, Kamei N, Harada K, Inoue K, Yoshinaga T, Saito N, Koizumi A (2006) The bioconcentration factor of perfluorooctane sulfonate is significantly larger than that of perfluorooctanoate in wild turtles (Trachemys scripta elegans and Chinemys reevesii): an Ai river ecological study in Japan, Ecotoxicol Environ Saf 65: 14-21

This study investigated bioaccumulation of PFOS and PFOA in river turtles from the Ai River in Settsu City. Serum PFOS levels ranged from 2.4 to 486 mg/L, while water levels ranged from 2.9 to 37 ng/L. The calculated geometric mean bioconcentration factor of PFOS was 10,964, indicating bioaccumulation. Serum PFOA levels varied from 0.2 to 870 mg/L and water levels ranged from 16.7 – 87,100 ng/L. The calculated bioconcentration factor of PFOA was 3.2 indicating slight bioconcentration.

Inoue K, Okada F, Ito R, Kato S, Sasaki S, Nakajima S, Uno A, Saijo Y, Sata F, Yoshimura Y, Kishi R, Nakazawa H (2004) Perfluorooctane sulfonate (PFOS) and related perfluorinated compounds in human maternal and cord blood samples: assessment of PFOS exposure in a susceptible population during pregnancy, Environ Health Perspect 112: 1204-1207

This study investigated PFAS in 15 women and in cord blood from their infants. PFOS (4.9 – 17.6 ng/ml) was found in all women and PFOA (0.5 – 2.3 ng/ml) in 3 of them. In cord blood, PFOS was found in all infants (1.6 – 5.3 ng/ml). PFOA was not found. The authors note that, "To the best of our knowledge, this is the first report of PFOS and its related compounds in pregnant Japanese women and fetal cord blood."

Saito N, Harada K, Inoue K, Sasaki K, Yoshinaga T, Koizumi A (2004) Perfluorooctanoate and perfluorooctane sulfonate concentrations in surface water in Japan, J Occup Health 46: 49 - 59
This study measured PFOS and PFOA in rivers, coastal, water and tap. The river and coastal areas included the Iwate River, Osaka River, Iwate Se and Osaka Sea. Tap water samples were from Kyoto, Osaka, Amagasaki, Kobe, Morioka, Sendai, and Yokote.

PFOA (0.33 – 456.41) ng/ml) and PFOS (0.24 – 37.32) were found in all river and coastal water samples. High levels were found in the Ina River (Kinki District – 456.41 ng/L PFOA, 37.32 ng/L PFOS) and the Koshien Coast near Kinki District (447.74 ng/L PFOA and 27.69 ng/L PFOS). The collection site along the Ina River was downstream from the Osaka International Airport and the authors note that it is likely that fluorinated firefighting foams led to the contamination. The highest levels were found in the Osaka near a discharge site where a level of 67,000 ng/L PFOA was measured. The authors estimated that this site discharges 18 kg of PFOA daily.

PFOA was found in all samples of tap water (0.12 – 40 ng/L) and PFOS was found in four out of six samples (0.2 – 12 ng/L). PFOA (40 ng/L) and PFOS levels (12 ng/L) were highest in samples from Osaka (Hanshin Area and city of Osaka). More than one million people drink water contaminated with these levels of PFAS.

Moriwaki H, Takatah Y, Arakawa R (2003) Concentrations of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in vacuum cleaner dust collected in Japanese homes, J Environ Monit 4:753-757
This study investigated PFOS and PFOA in household dust from 16 homes. PFOS (15 – 2500 ng/g) and PFOA (69 – 3700 ng/g) were found in all samples. The authors note that "small children spend their life in the space near the floor, and the compounds have been used in floor wax and carpets."

Saito N, Sasaki K, Nakatome K, Harada K, Yoshinaga T, Koizumi A (2003) Perfluorooctane sulfonate concentrations in surface water in Japan, Arch Environ Contam Toxicol 45: 149-148
This study investigated PFOS levels in surface water from many rivers and coastal areas.

PFOA was found in all samples of river water at levels ranging from 0.3 – 135 ng/L. Rivers included: Kusiro River, Kameda River, Takase River, Oirae River, Mabuchi River, Iwaki River, Kitakami River, Mogami River, Sakura River, Lake Kasumigaura, Ara River, Tsurumi River, Sagami River, Agano River, Shinano River, Seki River, Jinzu River, Shou River, Fuji River, Abe River, Ooi River, Kiku River, Tenryuu River, Yahagi River, Shounai River, Kiso River, Nagara River, Ibi River, Yasu River, Suzuka River, Jumozu River, Kusida River, Miya River, Kino River, Yamato River, Tenjinn River, Kii River, Gouno River, Yoshii River, Asahi River, Takanasi River, Ashida River, Oota River, Sanami River, Yamakuni River, Onnga River, Yabe River, Chikugo River, Kase River, Matsuura River, Motoake River, Yoshino River, Doki River, Sigenobu River, Hiji River, Mononobe River, Niyodo River, Shimanto River, Abukuma River,

Matu River, Suriage River, Siroishi River, Tone River, Watarse River, Kido River, and Kogai River.

Increases in PFOS levels along rivers could be connected to sewer or industrial waste water discharge sites. The authors note that the Ara, Tama, and Yodo Rivers all of which are polluted with PFOS are the drinking water sources for more than eight million people.

PFOS was found in all samples of coastal areas at levels ranging from 0.2 – 25.2 ng/L. Coastal areas included: Sea of Kushiro, Mutsu Bay, Hachinohe Bay, Jyodoga Coast, Miyako Bay, Kamaishi Bay, Honjyo Marina, Souma Bay, Nagasaki Bay, Chiba-Funahashi Bay, Yamashita Bay, Pacific Ocean, Nagoya Bay, Koshien Bay, Motoujina Shima coast, and Hakata Bay.

Taniyasu S, Kannan K, Horii Y, Hanari N, Yamashita N (2003) A survey of perfluorooctane sulfonate and related perfluorinated organic compounds in water, fish, birds, and humans from Japan, Environ Sci Technol 37: 2634-2639
This study examined 22 surface seawater samples from Ishikari Bay and Lake Shikotsu (Hokkaido), Tokyo Bay, Osaka Bay, Hiroshima Bay, Ariake Bay, and Kin Bay (Okinawa) along with freshwater samples from Lake Biwa. In addition, 48 blood samples and 30 liver samples were collected from 23 species of fish collected from Tokyo Bay, Osaka Bay, Hiroshima Bay, Ariake Bay, Kin Bay (Okinawa), and Lake Biwa. Blood and liver samples were also collected from carrion crow (*Corvus corone corone*), mallard (*Anas platyrhynchos*), and pintail duck (*Anas acuta*) around Tokyo Bay. Samples were also collected from rabbits and domestic ducks. Finally, human blood and serum were collected from 10 volunteers.

PFOS was found in surface water from Tokyo Bay (8 – 59 ng/L), Osaka Bay (<4 – 21 ng/L), Lake Biwa (<4 – 7.4 ng/L) and Ariake Bay (<9 – 11 ng/L). PFOS was found in all blood (1 – 834 ng/ml) and liver samples (3 – 7900 ng/g wet weight) from fish. The highest levels, 834 ng/mL in blood and 7900 ng/g in liver, were respectively found in bluegill (*Lepomis macrochirus*) from Lake Biwa and ornate jobfish (*Pristipomoides argyrogrammicus*) from Kin Bay (Okinawa). The authors note that an electric power plant and a US military base are located at Kin Bay and that use of PFOS in fire-fighting operations on military bases may provide a possible source of PFOS in Kin Bay.

Birds (carrion crow, mallard, and pintail duck) and pets (rabbit and domestic duck) contained detectable levels of PFOS in liver and blood

In humans, PFOS was found at levels ranging from 2.4 – 14 ng/ml.

Press reports on PFAS

Okinawa Times, January 11, 2019
健康被害に危機感　米軍と県は解決急務　嘉手納基地内で高濃度 PFOS
Crisis to human health – US Force and Prefectural Government needs to solve immediately
High concentration of PFOS in Kadena Base

Okinawa Times, January 11, 2019
嘉手納基地の PFOS 汚染、米基準の最大 1 億倍　水源へ流出　高い可能性
PFOS contamination in Kadena Base, high possibility 100 mil. times of US standards outflowed into water source
Survey by US Force in June 2014 showed that 90,000 ppt PFOS was contained in the fire extinguisher located a reservoir, 200 m away from the town office. US Force's survey on fire sprinklers in eleven buildings in February 2016 found that PFOS concentration was 9.5 million to 9.5 billion ppt. US Force seems likely to be a cause of water source and drinking water contamination around Kadena Base but US Force still denies the facts and not allow Japanese government to access the base.

Okinawa Times, December 10, 2018
普天間飛行場で汚染指摘の有害物質、横田基地でも漏出　井戸から高濃度の PFOS
Hazardous substance pointed in Futenma found in Yokota base, high concentration PFOS from wells
Based on the records obtained by information disclosure in the US, at least 3136L of fire-distinguisher spilled out during 2010-2017 in Yokota Air Base. Wells in the base contained 35 ppt of PFOS and PFOA in 2016.

Okinawa Times, November 24, 2018
湧き水から高濃度の化学物質　「普天間」周辺　泡消火剤の影響示唆
High concentration of chemical substance from springs around Futenma Air Base suggest influence by foam extinguisher
The prefectural government survey in Aug-Sep showed highest concentration of PFOS since 2016. 2000ng/L in Kyuna, 650ng/L in Mendakarihiga and etc.

Okinawa Times, October 30, 2018
沖縄県、汚染原因確認求める　普天間の有害物質　防衛局通し米軍に
Okinawa Prefecture requested US Force to confirm the source of contamination by PFOS/PFOA

Okinawa Times, August 28, 2018
沖縄県の面会要請、米軍が拒否　普天間飛行場周辺で汚染物質検出
US Force rejected meeting request by Okinawa Prefectural Government, detection of PFOS around Futenma Air Base

Okinawa Times, May 18, 2018
浄水場汚染、沖縄防衛局が補償へ　「基地が起因かは微妙だが」
Okinawa Defense Bureau compensate contamination for a filtering plant, "not sure whether the US base is a source"
Okinawa Governmental Enterprise Bureau requested Okinawa Defense Bureau of Ministry of Defense 170 million yen to compensate for cost to replace activated carbon to eliminate PFOS at Chatan Filtering Plant. The Defense Bureau will arrange compensation without determining the source of contamination clearly.

PFAS regulations

A small number of PFAS are partially regulated under five principal laws.

Chemical Substances Control Law (CSCL)

PFOS or its salts, and PFOSF - Class I Specified Chemical Substance since April 2010
(manufacture, import and use are virtually prohibited)
PFOA, PFHxA, PFHxS - General Chemical Substances
(notification for manufacture/import of substances more than 1 ton/year)
Please see the section "PFAS production, use, and waste management in Japan" for more details.

PRTR

PFOS or its salts, and PFOSF - Class I Specified Chemical Substance
(subject to reporting if manufacturing over 0.5 ton/year)
PFOA - APFO (Cas:3825-26-1) is Class II Specified Chemical Substance
(subject to SDS system, but not PRTR system)

Water Supply Standard

PFOS and PFOA are added as "monitoring substances" for water supply standard in 2009 but no limit was set by the government. No maximum levels for PFOS, PFOA or any other PFAS substance are set for tap water in Japan.

Waste Management Law

Wastes containing fluorine and its compounds are subject to the Waste Management Law.
"Technical Documents on Treatment of Wastes containing PFOS (Revised March 2011)[15]" provides the following as targets for treatment.

1. Decomposition rate of PFOS and its salt - more than 99.999%
2. Concentration of PFOS and its salt
   i. Drainage 2μg/L
   ii. Residue 3mg/kg
3. Concentration of hydrogen fluoride
   i. Exhaust gas 5mg/m^3N
   ii. Drainage 8mg/L for public water area other than sea area
   15mg/L for sea area

The Export Trade Control Order and the Import Trade Control Order

POPS wastes for substances listed in the Stockholm Convention (applies to PFOS) are subject to the requirement of import or export approval.

PFAS regulations in other countries

Most PFAS are not regulated, but PFOA and PFOS have come under regulatory scrutiny, particularly in the US where a large number of contaminated drinking water sites have been

[15] https://www.env.go.jp/recycle/misc/pfos/tptc.pdf

identified. In 2016, the US established a federal health advisory limit in drinking water of 70 ppt (parts per trillion) for PFOA and PFOS combined. This advisory limit is not enforceable but is used as a guideline. A recent US government review by the Agency for Toxic Substances and Disease Registry has proposed tightening exposures which would translate to drinking water limits of 7 ppt for PFOS and 11 ppt for PFOA.

In the absence of federal regulations, individual US states (California Colorado, Minnesota, Michigan, New Jersey, New Mexico, Texas, Vermont, and Washington) have moved forward to regulate PFAS in drinking water, firefighting foam, personal protective equipment and wastes. Another 11 states are considering or have already proposed similar regulatory actions. Information about individual state proposals can be obtained here.

In 2018, state regulators in California set interim notification limits of 13 ppt for PFOS and 14 ppt for PFOA in drinking water. Regulators noted that both substances were listed by the state as developmental toxicants and that the National Toxicology Program concluded that both substances are "presumed to be an immune hazard to humans." Colorado uses a 70 ppt combined limit of PFOS and PFOA as a groundwater quality standard. Colorado also regulates PFOS and PFOA as hazardous waste. Massachusetts sets a 70 ppt limit for PFOA, PFOS, PFHxS, PFNA and PFHpA combined. Michigan uses the federal 70 ppt combined PFOS and PFOA standard as a limit for drinking water. The Minnesota Department of health recommends the following guidance values: 2000 ppt for PFBS, 27 ppt for PFHxS, 27 ppt for PFOS, 7000 ppt for PFBA, and 35 ppt for PFOA. New Jersey added PFNA to its hazardous substances list and set a limit for PFNA of 13 ppt in drinking water. New Jersey proposed limits of 14 ppt for PFOA and 13 ppt for PFOS. Vermont sets a drinking water health advisory limit of 20 ppt for PFOA, PFOS, PFHxS, PFHpA and PFNA combined. In 2018, Washington banned PFAS in firefighting foams and personal protective equipment and began a rulemaking process to established drinking water limits. The New York Department of Health has proposed 10 ppt for PFOS and 10 ppt for PFOA. The proposal considered the fact that people already have exposure to these substances from other sources.

Recommendations for Stockholm Convention COP9

1. PFOA should be listed in Annex A with no specific exemptions. If exemptions are granted, they should be for specific products and the listing should require labeling new products that contain PFOA so that Parties can fulfill requirements under Article 6 as done previously for HBCD (SC-6/13).

2. Due to the costly, highly polluting nature of firefighting foams, and the availability of cost-effective, technically feasible non-fluorinated alternatives, no specific exemptions should be adopted for either PFOS or PFOA production and/or use in firefighting foams.

3. Specific exemptions or acceptable purposes for the following 11 uses of PFOS should be ended: photo-imaging, photo-resist and anti-reflective coatings for semiconductors; etching agent for compound semiconductors and ceramic filters; aviation hydraulic fluid; certain medical devices; photo masks in semiconductor and LCD industries; hard metal

plating; decorative metal plating; electric and electronic parts for some color printers and color copy machines; insecticides for control of red imported fire ants and termites; and chemically-driven oil production.

4. The following 3 acceptable purposes should be converted into specific exemptions: metal plating (hard metal plating only in closed loop systems); firefighting foams; insect bait for control of leaf-cutting ants from *Atta* spp. and *Acromyrmex* spp. Sulfluramid should be named in the listing and its use sharply limited to cultivation of specific crops.

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Annex 1. PFAS toxicity

The Stockholm Convention expert committee (please see Annex 3) evaluated the toxicity characteristics of PFOS in 2007 and PFOA in 2017. Since then, more scientific information has emerged for both these substances along with some of the shorter-chain PFAS aggressively promoted by the industry as substitutes.

Recent research shows the harmful impacts of PFAS

Recent studies have linked PFAS substances to a variety of human health effects: cardiovascular disease, markers of asthma, damage to semen quality, ovarian insufficiency, altered glucose metabolism, lower testosterone levels in male adolescents, association with shorter birth length in girls, elevated blood pressure, abnormal menstruation, lower birth weight in infants, possible increased risk of female infertility due to endometriosis, and decreased lung function in children with asthma.

The chemical industry promoted perfluorohexane sulfonate (PFHxS) as a substitute for PFOS. In 2018, the Stockholm Convention expert committee concluded that it "warrants global action." PFHXS is found in 2 – 4 month-old infants and associated with damage to semen quality. The Stockholm Convention expert committee found that PFHxS has been detected in human blood and breast milk in many regions, and is together with perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and perfluorononanoic acid (PFNA) one of the most frequently detected and predominant PFASs in human blood. The Committee noted that the fetus is exposed to PFHxS via umbilical cord blood and that animal studies show impacts on reproduction, liver function, thyroid hormone levels, and lipid and lipoprotein metabolism.

Studies showing the toxicity, environmental fate, and occurrence of PFAS in current use include:

Perfluorobutanoic acid (PFBA)

- Effects on thyroid and developmental delays in offspring exposed during pregnancy
- Similar toxicity to liver as PFOA
- Associated with damage to semen quality
- Found in home-produced eggs
- Found in the Arctic
- Efficiently translocated into plants
- Taken up by corn
- Found in fruits and vegetables
- Contaminates fish
- Found in humans in a community with known drinking water contamination
- Found in consumer products

Perfluorobutane sulfonate (PFBS)

- Associated with damage to semen quality
- Disrupts pancreas formation in zebrafish
- Associated with cardiovascular disease in humans
- Associated with markers of asthma in humans

- Increases fatty tissue formation in laboratory studies
- Impairs visual function in fish
- Damages thyroid function in fish in subsequent generations
- Induces reproductive toxicity in animal studies
- Found in 2 – 4 month-old infants
- Found in humans in community with known drinking water contamination
- Found in children
- Found in the Arctic
- Found in consumer products

Perfluorohexanoic acid (PFHxA)

- Similar toxicity to liver as PFOA
- Associated with damage to semen quality
- Negatively associated with testosterone levels in adolescent humans
- Alters zebrafish behavior
- Modulates immune response in vitro
- Contaminated drinking water linked to human body burden
- Alters amphibian embryogenesis
- Exposes the human fetus vis presence in amniotic fluid
- Found in human milk
- Found in house dust
- Found in US wildlife preserves
- Found in the Arctic
- Contaminates fish
- Found in Indo-Pacific humpback dolphins and finless porpoises
- Efficiently translocated into plants
- Resistant to sewage treatment
- Found in US wastewater treatment plants

Perfluoroheptanoic acid (PFHpA)

- Alters amphibian embryogenesis
- Exposes the human fetus via presence in amniotic fluid
- Found in human milk
- Manufacturing sites, military fire training, and wastewater treatment plants are predictors of pollution
- Use in airport firefighting foams pollutes groundwater, lakes, soils, and fish
- Found in remote mountain snow
- Bioaccumulates in plankton
- Contaminates fish
- Efficiently translocated into plants

PFAS in people

Numerous studies show PFAS contamination in people. For example, in one study of 299 infants, PFOS was found in the blood of 297 of them and PFOA was found in all of them.

The Stockholm Convention conducts global monitoring of substances listed in the treaty as part of its effectiveness evaluation. The most recent data is from a series of regional monitoring reports published in 2015.

In Africa, the treaty monitoring study noted that PFOS was detected in mothers' milk from all 11 countries that submitted samples with levels varying from 1 – 34 ppt. The report notes that, *"Assuming that there is no industrial production of PFOS in the region, exposure of humans to PFOS and related chemicals might probably come from different kinds of waste, releases from industrial applications in firefighting and the various consumer products."*

The monitoring report for the Asia-Pacific region notes that only a few countries reported data. The report shows PFOS in air in Fiji, Hong Kong, Japan and in blood including maternal plasma in Japan. PFOS was also measured in marine areas in China, Hong Kong, Japan, Macao and rivers and lakes in Philippines, South Korea, and Thailand.

In Central and Eastern Europe, the Stockholm Convention monitoring report notes that data on water monitoring are scarce and data for the presence of PFOS in human tissues is even more limited.

Stockholm Convention monitoring in Latin America and the Caribbean showed that only Uruguay reported data on PFOS in air and the report notes that at this time (2015) there was no formal monitoring program in the region for determination of PFOS.

In Western Europe and Other States, monitoring data also includes the Arctic where PFOS and PFOA in air were measured. The report notes that phaseouts of PFOS and PFOA are reflected in declining concentrations but that fluorinated substitutes show increasing levels in Arctic air. The study also reveals that of all the measured POPs, PFOS was the predominant substance in human plasma, with the highest level of 470 ppt reported in an Inuit resident of the Arctic.

Recent scientific studies show the widespread presence PFAS in humans. Data include the following:

- Perfluorohexane sulfonate (PFHxS), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnDA), and perfluorotridecanoate (PFTrDA) in human milk in Sweden
- PFOS, PFOA, PFNA, PFDA, PFUnA and PFHxS in maternal sera, placentas, and fetuses.
- PFOS, PFOA, PFHxS, and PFNA in New Zealand adults
- PFOS, PFDoDA, PFUnDA and PFTrDA in pregnant Japanese women
- PFOS, PFOA, PFHxS in >94% of community residents with drinking water contaminated by a former US Air Force base.

- 10 long-chain PFAS in California women.
- PFOS< PFOA< PFHxS, PFNA, PFUnDA, PFHpS found in maternal plasma in Norway.
- PFAS in amniotic fluid in Denmark.
- Prenatal exposure to PFOS, PFHxS, PFHpS, PFNA, and PDFA in Denmark.
- Prenatal exposure to PFBS, PFHxS, PFUA in China.
- Six PFAS in middle-aged US women.
- PFNA, PFDA, PFUnDA, PFHxS, PFOA, and PFOS in more than 99% of sampled pregnant Swedish women.
- PFAS in maternal and cord blood in mothers exposed to the US World Trade Center disaster during pregnancy.
- PFOA, PFOS, PFNA, PFHxS in cord blood of Slovak infants.
- PFOS, PFOS and 6:2 CL-PFESA in cerebrospinal fluid in China indicating ability to cross the blood-CSF barrier.
- PFOS, PFOA, PFNA, and PFHxS in children.
- PFOA, PFOS< PFNA, and PFHxS in pregnant US women.
- PFOS< PFOA< PFHxS and PFNA in maternal serum in the UK.
- PFOA, PFOS, and PFHxS in Chinese women.
- PFOA and PFNA in US children.
- PFAS in Alaska Natives.
- PFHxS, PFOA< PFOA, PFNA, PFDA, PFUdA, PFDoA, and PFTrDA in >85% of sampled pregnant women in China.
- PFAS in pregnant Chinese women.

Manufacturers knew PFAS were harmful

Recently obtained documents indicate that the original manufacturers of PFOS and PFOA knew about the harmful characteristics of both substances decades ago.

A lawsuit filed by the US State of Minnesota against 3M produced internal company documents that demonstrated that the company knew PFOS and PFOA were accumulating in people for more than 40 years. 3M had previously withheld required documents from US regulators which resulted in a USD$1.5 million fine in 2006. In 1975, university researchers found a fluorinated substance in human blood and 3M confirmed that it was PFOS. Subsequent company testing found PFOS levels in 3M personnel at levels 50 – 1000 times higher than normal levels. In 1978, tests on monkeys feed PFOS resulted in all the animals dying and those given PFOA developed lesions on their spleen, lymph nodes, and bone marrow, all relevant to a functioning immune system. By 1989, the company knew that PFOS suppressed the immune system, caused tumors in animals, and that rates of cancers of the digestive organs and prostate were elevated in its own workers. The company proceeded to produce the substance anyway.

Internal company documents reveal that DuPont knew decades ago that PFOA affected the livers of dogs and humans, encouraged the growth of testicular tumors in rats, and appeared to result in endocrine disorders and kidney cancer in workers. In 1978, the company documented immunotoxicity and other adverse effects in tests on monkeys exposed to PFOA and PFOS. By 1984, DuPont knew that PFOA was toxic, didn't break down, accumulated in blood, transferred

from mothers to the fetus, and polluted drinking water supplies. DuPont decided to keep producing it anyway as it became incorporated into a multitude of products and processes. The company's real attitude about the consequences of PFOA production is revealed in its internal documents as "the material 3M sells us that we poop to the river and into drinking water."

DuPont was fully aware of PFOA's hazards, but a study of the company's decision-making processes noted that DuPont made a calculated, rational decision to pollute anyway. The authors estimate that for DuPont, "it was value-maximizing to pollute if the probability of getting caught was less than 19%." In reality the probability was much less than that and now communities and governments bear the burden of that private sector decision.

Annex 2. The high cost of PFAS cleanup

PFAS manufacturing and use in a multitude of products such as firefighting foams has resulted in widespread pollution – especially in water due to the solubility of PFAS substances. PFAS-contaminated sites have been identified in Australia, Canada, China, Germany, Italy, Japan, Netherlands, New Zealand, South Korea, Sweden, and the US, including a large number of military bases that contribute to 172 PFAS contamination sites in 40 states. In 2018, the US State of Minnesota entered into an agreement with 3M for the company to pay the state USD$850 million for costs associated with cleanup of PFAS including PFHxS due to manufacturing and releases by the company.

Clean up of PFAS pollution is difficult and costly. According to the Polluter Pays Principle, and sound economic policy, these types of external costs should not be borne by taxpayers, the state or national treasury, or by any other third party. Rather, these costs should be internalized within producer industries to avoid market distortion. As noted by UN Environment in 2012, "The vast majority of human health costs linked to chemicals production, consumption and disposal are not borne by chemicals producers, or shared down the value-chain. Uncompensated harms to human health and the environment are market failures that need correction."

Examples of estimated and actual cleanup costs for PFAS pollution include:

- Recent US government agency estimates for the cost PFAS clean-ups and associated monitoring due to use of firefighting foams at US military bases are more than USD$2 billion. There are also expensive clean up costs and estimates in a variety of US states including Alaska, New Jersey, New York (see also here and here), Vermont, Virginia, and Washington.
- The World Bank estimates that if just 20% of fluorinated firefighting foam in China is used for training or fire extinguishing, remediation costs would exceed USD$800 million.
- Remediation of PFAS-containing firefighting foam at the Düsseldorf Airport in Germany will take years or even decades. Cleanup costs cited by the European Chemicals Agency exceed €100 million. There are additional documented remediation costs due to PFAS pollution in Germany – see here, here, and here.
- Clean up due to use of 3M's "Light Water" firefighting foam containing PFOS and PFHxS at 18 military bases in Australia is estimated to cost hundreds of millions of dollars. The cleanup of just a single firefighting training college in Australia is estimated to cost AUS$80 million.
- To clean up groundwater polluted by PFAS around firefighting areas in Norway costs €3.5-5.5 million per training site.
- Firefighting training sites are the main sources of PFAS pollution in Sweden leading to €1 million in annual costs for charcoal filtering of water in Uppsala and a new water supply in Ronne costing €3 million. Extrapolated estimates for advanced cleaning of all waste water treatment plants in Sweden would only moderately remove fluorinated compounds but still cost USD$230 million per year.
- New Zealand has budgeted NZE$1 million to investigate cleanup of PFAS associated with firefighting foam use by military bases.

Annex 3. PFAS and the Stockholm Convention

The Stockholm Convention objective is to protect human health and the environment from persistent organic pollutants. Persistent organic pollutants (POPs) are a class of highly hazardous chemical pollutants that are recognized as a serious, global threat to human health and to ecosystems. Substances can be added to the Stockholm Convention after evaluation and recommendation by the POPs Review Committee (POPRC). Japan became a Party to the treaty in 2002.

PFOS

Governments added PFOS to the treaty list at the 4th Conference of the Parties in 2009 and subsequently adopted a series of guidance documents on PFOS alternatives.

Japan did not register any specific exemptions for PFOS. However, Japan did register the following acceptable purposes: Photo-imaging, photo-resistant and anti-reflective coatings for semiconductors, etching agent for compound semi-conductors and ceramic filters, and certain medical devices.

When PFOS was listed in Annex B of the treaty in 2009, a very large number of loopholes accompanied its listing that permitted continued production and use. At COP9 in April/May 2019, Parties will determine if these loopholes are still needed or if some can be ended. The decision will focus on 6 time-limited ones (specific exemptions) and 8 time-unlimited ones (known as acceptable purposes). The POPRC recommended the following changes to the PFOS listing in the Convention:

End loopholes for 11 PFOS uses: photo-imaging, photo-resist and anti-reflective coatings for semiconductors; etching agent for compound semiconductors and ceramic filters; aviation hydraulic fluid; certain medical devices; photo masks in semiconductor and LCD industries; hard metal plating; decorative metal plating; electric and electronic parts for some color printers and color copy machines; insecticides for control of red imported fire ants and termites; and chemically-driven oil production.

Convert two time-unlimited exemptions to time-limited exemptions: metal plating (hard metal plating only in closed loop systems) and firefighting foams. This gets the clock running on ending these uses in five years. On the firefighting foams, the Committee recommended stopping production and only allowing use for class B fires (ones involving solvents, oil etc.) and only in installed systems. The Committee also noted that, *"a transition to the use of short-chain per- and polyfluoroalkyl substances (PFASs) for dispersive applications such as fire-fighting foams is not a suitable option from an environmental and human health point of view…"* This is extremely important since the fluorinated alternatives are persistent, toxic and readily pollute drinking water.

Continue time-unlimited exemption for one use: insect bait for control of leaf-cutting ants from *Atta* spp. and *Acromyrmex* spp. This vaguely-worded listing actually refers to a pesticide called sulfluramid that degrades to PFOS. The POPRC recommended naming sulfluramid in the treaty under the PFOS listing and narrowing its use to agriculture.

IPEN recommendations for PFOS

Specific exemptions or acceptable purposes for the following 12 uses of PFOS should be ended: photo-imaging, photo-resist and anti-reflective coatings for semiconductors; etching agent for compound semiconductors and ceramic filters; aviation hydraulic fluid; certain medical devices; firefighting foams, photo masks in semiconductor and LCD industries; hard metal plating; decorative metal plating; electric and electronic parts for some color printers and color copy machines; insecticides for control of red imported fire ants and termites; and chemically-driven oil production. If a specific exemption is allowed for use in firefighting foams, the POPRC recommendations should be adopted.

The following 2 acceptable purposes should be converted into specific exemptions: metal plating (hard metal plating only in closed loop systems); and insect bait for control of leaf-cutting ants from *Atta* spp. and *Acromyrmex* spp. Sulfluramid should be named in the PFOS listing and its use sharply limited to cultivation of specific crops.

Due to the costly, highly polluting nature of firefighting foams, and the availability of technically feasible, high-performing non-fluorinated alternatives, no exemption should be granted for this use.

PFOA

PFOA is extremely persistent and does not degrade under relevant environmental conditions. It bioaccumulates in air-breathing land and marine mammals, including humans. PFOA is found in water, snow, air, sediment and biota at remote locations including the Arctic. In 2017, the Stockholm Convention POPs Review Committee noted the link between PFOA and serious illnesses in humans, including diagnosed high cholesterol, ulcerative colitis, thyroid disease, testicular cancer, kidney cancer and pregnancy-induced hypertension. PFOA is transferred to the fetus through the placenta and to infants via breast milk. PFOA-related compounds such as fluorotelomer alcohols, fluoropolymers and fluorotelomer-based polymers must be included in actions designed to eliminate PFOA releases since they can degrade to PFOA.

In 2018, the POPRC recommended that governments list PFOA and related substances in Annex A of the Stockholm Convention for global elimination.

Ten time-limited exemptions accompany the PFOA listing recommendation, however, many of these are not justified.

Proposed PFOA Exemption	**Comment**
5 years 3 exemptions connected to semiconductor manufacturing (equipment or plant infrastructure, legacy equipment, photo-lithography, etch process)	Alternatives without PFOS or PFOA are available for photolithography and etch processes. For example, IBM eliminated both in 2010. The other proposals are not sufficiently defined.

Photographic coatings applied to films	Obsolete use of PFOA replaced by digital imaging, including in developing and transition countries.
Textiles for oil and water repellency for workers	Proposal relies on industry claims and does not state what specific products the exemption would cover or how worker protection can be achieved without relying on a toxic chemical-impregnated textile.
Invasive medical devices	Alternative medical devices made without PFOA have passed all regulatory requirements, are available on the market, and in use.
Implantable medical devices	Alternative medical devices made without PFOA have passed all regulatory requirements, are available on the market, and in use.
Firefighting foams	Cost-effective non-fluorinated alternatives are in use at major airports and military installations and perform as well as PFAS-containing foams.
10 years For manufacture of semiconductor or related electronic devices; refurbishment parts containing fluoropolymers and/or fluoroelastomers with PFOA for legacy equipment or legacy refurbishment parts	See above for manufacturing. Legacy equipment proposal is not specific and include thousands of unnamed parts. Retrofitting with parts that do not contain PFOA should be utilized, instead of continuing PFOA production and use.
Until 2036 To use PFOI (a PFOA-related substance) to make PFOB for producing pharmaceutical products *"with a review of continued need for exemptions."*	In 2015, more than 100 governments agreed that environmentally persistent pharmaceutical products are an emerging policy issue of global concern in the SAICM process. A global exemption should not be adopted on behalf of a single company (Daikin) and exemptions for environmentally persistent pharmaceutical products should not be recommended.

IPEN recommendations for PFOA

PFOA should be listed in Annex A with no specific exemptions. If exemptions are granted, they should be for specific products and the listing should require labeling new products that contain PFOA so that Parties can fulfill requirements under Article 6 as done previously for HBCD (SC-6/13). In addition, due to the costly, highly polluting nature of firefighting foams, and the availability of technically feasible, high-performing non-fluorinated alternatives, no exemption should be granted for this use.

PFHxS

PFHxS and related compounds are persistent in water, soil and sediment and unlikely to undergo degradation in the environment including hydrolysis, aqueous photolysis or under anaerobic conditions. PFHxS biomagnification factors (BMF) greater than 1 have been observed in food chains including Arctic bird/fish, Arctic polar bear/ringed seal, dolphin/fish, and fish/zoo plankton among others, indicating bioaccumulation. PFHxS has the longest half-life in humans determined for any PFAS. PFHxS undergoes long-range transport and is found in Arctic air, sediment, snow, ice, soil, sediment and biota (including humans) and in Antarctic biota and snow. *In vivo* and epidemiological studies show that PFHxS negatively affects liver function, thyroid, and the developing immune system resulting in reduced effects of vaccines and higher incidences of infections and asthma in children. A significant association between PFHxS exposure and breast cancer has been found in Greenlandic Inuit women. PFHxS is widely found in breast milk and is one of the most frequently detected and predominant PFAS in human blood, including maternal and infant cord blood. In September 2018, the POPRC determined that PFHxS "warrants global action" and moved the substance to the third and final evaluation during 2018 – 2019.

PFAS use in firefighting foams

There are many uses of PFAS, but one of the most highly polluting is in firefighting foams. This pollution occurs where the foam is used and quickly contaminates water and moves. Airports and military bases are common sources of PFAS pollution.

PFOS and PFOA were the original components in firefighting foams, but after regulatory pressure in the US, many companies switched to shorter-chain substances such as PFHxS, PFBA, PFBS, PFHxA, and PFHpA. These substances also are persistent and have hazardous properties. Some are found in the Arctic, suggesting ability to undergo long-range transport. Recently, IPEN assembled a group of fire safety experts who produced a detailed report on issues involving firefighting foams and the technical feasibility of fluorine-free firefighting foams. Safer cost competitive non-fluorinated alternatives to PFAS in firefighting foams have been adopted by major airports, including Auckland, Copenhagen, Dubai, Dortmund, Stuttgart, London Heathrow, Manchester, and all 27 major airports in Australia.

In September 2018, the POPRC recommended severe restrictions on the use of PFOS and PFOA in firefighting foams. In addition, the Committee also made an extremely important recommendation **not** to use the fluorinated alternatives to PFOA and PFOS, *"due to their*

persistency and mobility as well as potential negative environmental, health and socioeconomic impacts."

The recommended restrictions on firefighting foams containing PFOA, PFOA-related substances, or PFOS include:

- No production.
- Use for 5 years only for liquid fuel vapor suppression and liquid fuel fires (Class B fires) already in installed systems.
- No import or export, except for environmentally-sound disposal.
- No use for training or testing purposes.
- By 2022, restrict use to sites where all releases can be contained.
- Ensure that all firewater, wastewater, run-off, foam and other wastes are managed in accordance with the treaty.

IPEN recommendations on PFAS use firefighting foams

Due to the costly, highly polluting nature of firefighting foams, and the availability of technically feasible, high-performing alternatives, no exemption should be granted for this use. IPEN supports the POPRC recommendation that fluorinated alternatives to PFOA and PFOS should not be used.