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RESEARCH ARTICLE

Life cycle analysis of perfluorooctanoic acid (PFOA) and its salts in China

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Abstract China has been the largest producer and emitter of perfluorooctanoic acid and its salts (PFOA/PFO). However, the flows of PFOA/PFO from manufacture and application to the environment are indistinct, especially flows from waste treatment sites to the environment. Here, a life cycle analysis of PFOA/PFO is conducted in which all major flows of PFOA/PFO have been characterized for 2012. Processes related to uses and possible releases of PFOA/PFO include manufacture and use, waste management, and environmental storage. During manufacture and use, emission from application was the most important (117.0 t), regardless of whether it flowed first to waste treatment facilities or was directly released to the environment, followed by manufacture of PFOA/PFO (3.9 t), while flows from the service life and end of life of consumer products were the lowest (1.2 t). Among five waste treatment routes, flows through wastewater treatment plants (WWTPs) were the highest (10.6 t), which resulted in 12.8 t of PFOA/PFO being emitted into the environment. Masses of PFOA/PFO emission were estimated to be 96.3 t to the hydrosphere, 25.6 t to the atmosphere, and 3.2 t to soils. Therefore, control over reduction of PFOA/PFO should focus on application of reliable alternatives and emission reduction from WWTPs using effective treatment techniques.

Keywords PFOA · Life cycle analysis · Fluoropolymer · Manufacture · Application · Waste management

Introduction

Perfluoroalkyl substances (PFASs) are a group of chemicals that have been widely used in various industrial and consumer products, such as carpets, fiber, leather, packaging, paper, and textiles for more than 60 years (OECD 2013). PFASs have been widely detected in environmental media, wildlife, and humans, especially perfluorooctane sulfonate (PFOS) and perfluorooctnoic acid (PFOA) (Giesy and Kannan 2001; Giesy and Kannan 2002). Due to their persistence in the environment, hazardous effects, and potential accumulation in food webs, PFOS and related compounds have been listed in the *Stockholm Convention on Persistent Organic Pollutants (POPs)* in 2009 (Wang et al. 2009). Also, to reduce industrial PFOA emission and residual PFOA content in commercial products, the U.S. Environmental Protection Agency (USEPA) and the eight major fluoropolymer (FP) and fluorotelomer (FT) manufacturers initiated the voluntary *2010/2015 PFOA Stewardship Program* in 2006 (USEPA 2006). In the European Union (EU), PFOA and its ammonium salt (APFO) have been identified as persistent, bioaccumulative, and toxic (PBT) substances (Vierke et al.

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2012; ECHA 2013). For this reason, in 2015, the EU council decided to submit a proposal to the Secretariat of the Stockholm Convention suggesting that PFOA and its compounds should be added to Annex A of the Convention.

An inventory of sources and emissions of PFOA/PFO to the environment has been estimated for the globe and China (Wang et al. 2014; Li et al. 2015). Since the phase out of PFOA by the 3M Company in 2002, there has been a geographical shift of industrial sources of PFOA from primarily North America, Europe, and Japan to emerging Asian economies, and China has become the largest emitter of PFOA/PFO. To estimate emissions of PFOA/PFO, most assessments have focused on production and industrial applications, while little attention has been paid to treatment of wastes. However, during industrial production and application, most of the PFOA/PFO is released into waste treatment facilities and does not directly enter the ambient environment. Therefore, it is essential to develop a material flow analysis for PFOA/PFO from a life cycle perspective.

Beginning in the 1970s, life cycle analysis (LCA) was developed from analyzing use of energy to a more comprehensive assessment of movements of masses of materials (Guinee et al. 2011). LCAs for metals, such as anthropogenic nickel, iron, aluminum, and chromium, have been conducted widely (Johnson et al. 2006; Wang et al. 2007; Reck et al. 2008; Chen et al. 2010). However, LCAs for persistent organic pollutants (POPs) are relatively scarce, with a few being conducted for polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs) (Lee et al. 2015; Ventura et al. 2015). For PFOA and PFOS, LCAs have been conducted for Switzerland (FOEN 2009). Since there was no production of PFOA and related compounds in Switzerland, PFOA was mainly released during manufacturing of carpets, aqueous firefighting foams (AFFFs), and textiles. Wastewater was the dominant pathway for emission of PFOA to the environment.

Based on estimates of emissions for the globe and China, sources of PFOA/PFO were identified, mainly including synthesis of PFOA/PFO, production of FP, application of aqueous fluoropolymer dispersions (AFDs), industrial processes using perfluorooctane sulfonyl fluoride (POSF)- and FT-based substances, and domestic emission during use and disposal of FP-, POSF-, and FT-based substances (Prevedouros et al. 2006; Wang et al. 2014; Li et al. 2015). PFOA/PFO as ingredients are released from production of PFOA/PFO and FP, and application of AFDs, directly as impurities or transformation products from precursors during application of POSF- and FT-based substances. During production and application of relevant substances, some PFOA/PFO can directly enter the environment while some enter the environment via waste treatment facilities. Therefore, waste treatment facilities, such as industrial and domestic wastewater treatment plants (WWTPs) and landfill sites, are potential sources of PFOA/PFO to the wider environment (Kim et al. 2012; Taylor et al. 2014; Clarke et al. 2015). Therefore, the LCA for PFOA/PFO included three stages: production and use, waste management, and storage in the environment.

The LCA for PFOA/PFO offers new perspectives on emission reduction, alternative processes, manufacturing of products, treatment of wastes, and general management of the environment. This assessment has estimated emissions of PFOA/PFO at various stages of the whole life cycle, results of which identify current and future flows of PFOA/PFO in China. Such estimates can further help governmental departments make more detailed assessments of risks and implement realistic policies for controlling releases of PFOA/PFO at each stage of the life cycle. Furthermore, since China is a signatory to the Stockholm Convention, the results of this analysis will help the central government of China to meet its obligations under the treaty for any future changes in proposed use and management of PFOA/PFO. Finally, the assessment can serve as a model for other developing countries to apply LCA promulgating policies for their own specific situations.

Materials and methods

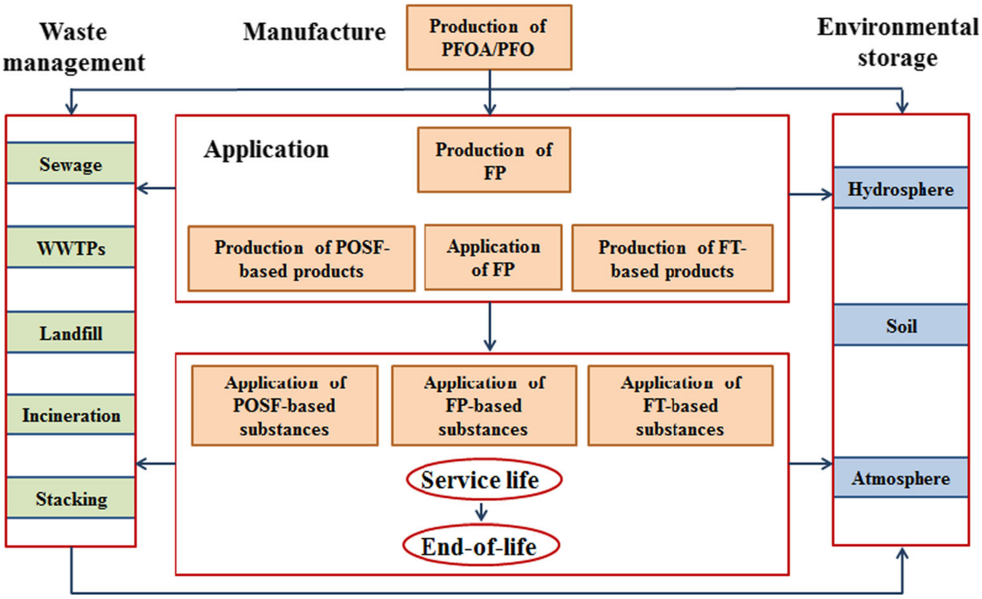
Framework of LCA

The LCA was based on relevant data for PFOA/PFO during 2012. System boundary ranged from production to environmental storage of PFOA/PFO and included three stages: production and use, waste management, and storage in environmental compartments (Fig. 1) (FOEN, 2009). Waste management included a sewage system, WWTPs, a landfill, incineration, and stacking, which was a little different with other countries due to a poor recovery system and simple stacking for solid waste in China. Environmental storage was estimated by information on releases from production, processing and use of relevant substances, and waste treatment.

Acquisition of data on production and use

According to the *Chinese National 12th Five-year Plan for Fluorine Chemical Industry*, in 2010, total production of perfluoroalkyl carboxylic acids (PFCAs) was 80 t, most of which was PFOA (CAFSI 2011). In 2012, approximately 90 t of PFOA was produced (Li et al. 2015). In industrial applications, PFOA/PFO and formulated products were used mainly as emulsifiers in suspension and emulsion polymerizations to produce FP (Prevedouros et al. 2006). According to the capacity for production of polytetrafluoroethylene (PTFE) in 2012 and forecast for production of FP in 2015, it was estimated that the volume of FP produced in 2012 would be 78,116 t (Table S1). There were three primary types of PTFE products, including granular (66.8%), fine powder (31.2%), and aqueous dispersion (2.0%) in China (Company LJ 2011).

Fig. 1 Sources of PFOA/PFO during the life cycle



Due to high-temperature melting or sintering treatment (usually >350 °C) during application of granular and fine powders, most of the PFOA/PFO is decomposed and only trace amounts (1–10 ppm) remain in plastics (Cope 2005). Therefore, releases via this process to the environment are likely to be negligible. However, AFDs are almost entirely applied to formulate coating products, during which 26% of PFOA/PFO residuals are released (FMG 2003).

On average, approximately 150 t of POSF-based substances is produced annually in China, among which approximately 50% was exported (Li et al. 2015). In addition, 50% of domestic consumption of POSF-based substances was mainly applied in three industrial sectors: metal plating as mist suppressants (20%), AFFFs as antifoaming surfactants (20%), and sulfluramid as a raw material (5%) (Zhang et al. 2012). Application in other industries, such as oil exploitation and semiconductor, was negligible.

In China, FT-based substances are produced in two processes including complete domestic production and secondary formulation using imported long-chain perfluroalkyl iodide (PFOI). In 2012, domestic manufacturers began to produce FT-based substances, with a total output of 1500 t (Li et al. 2015). In addition, approximately 600 t of PFOI was imported annually to process FT-based substances (Chen 2010), with a production volume of 3000 t (Prevedouros et al. 2006). Almost all of this production was applied in processing of textiles and leather.

Identification of transfer coefficients (TC) and emission factors (EF)

In this LCA, TCs and EFs of PFOA/PFO were determined based on information in the literature, from controlled laboratory experiments, reports from the governments or organizations, and statistical data. Details of TCs and EFs are available in Supporting Materials. The transport volume to the next stage or environmental emission of PFOA/PFO was calculated as follows (Eq. (1)) (FOEN 2009):

$$\text{transport volume(environmental emission)of PFOA/PFO} \\ = \text{TC (EF)} \times \text{PFOA/PFO input} \tag{1}$$

Results and discussion

Flows in production and use

Releases of PFOA/PFO during application, which involved production of FP and processing of FP-, POSF-, and FT-based substances, were dominant (Figs. 2 and 3). Emission during production of PFOA/PFO was relatively higher than those from waste treatment facilities. Therefore, emission of PFOA/PFO from industrial production and application was greater than those from emissions during its service life and end of life, which was defined as domestic sources. Estimates of industrial sources (Fig. S1), including production of PFOA/PFO, production and processing of FP, use of AFDs, and uses of POSF/FT-based substances during industrial processes, were 3.9, 113.4, 0.06, and 3.8 t, respectively. These estimates of emissions for China are similar to those reported elsewhere for 2004 to 2012, except those from production and processing of FP, which was mainly due to high emission from domestic manufacture (Li et al. 2015). PFOA/PFO from industrial sources was released with waste streams, namely exhaust gases, wastewaters, and solid wastes. PFOA/PFO released from industrial sources to sewage, WWTPs, and solid

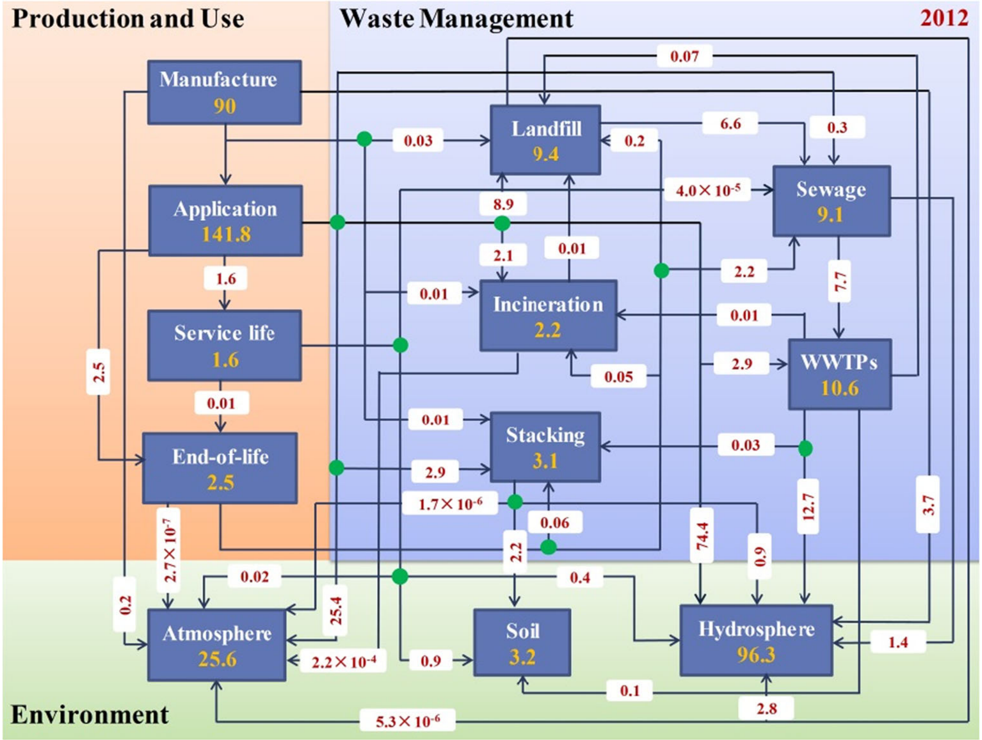
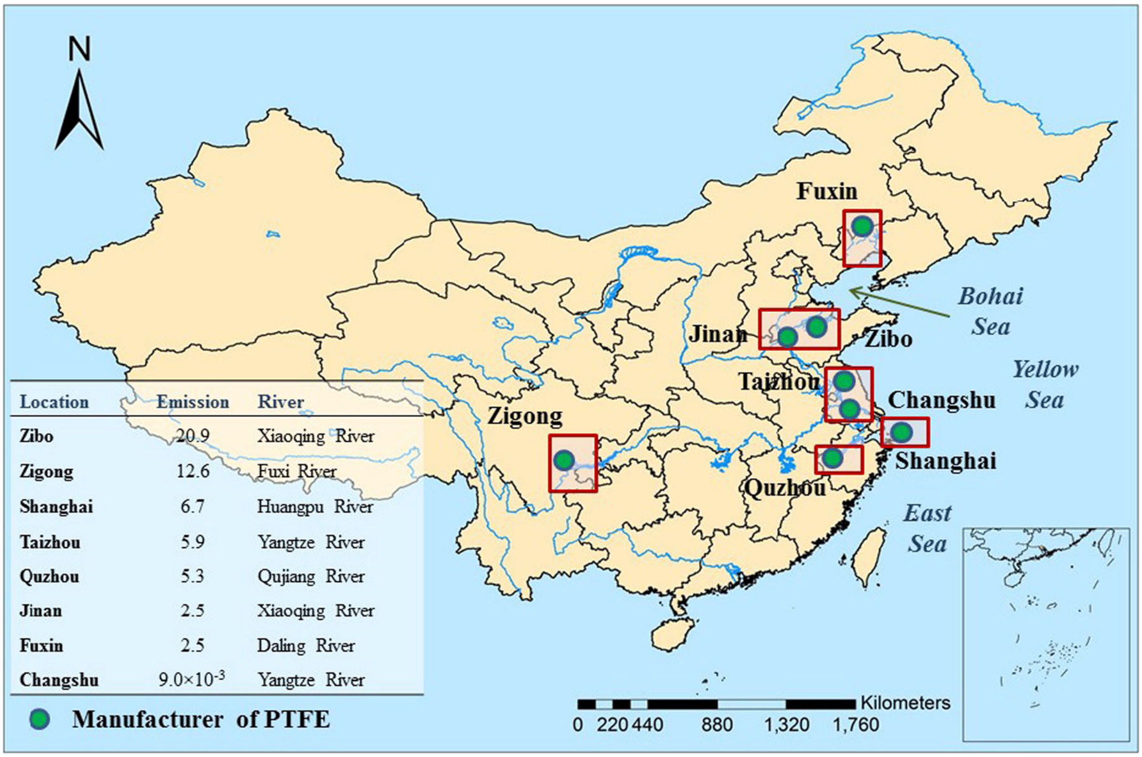


Fig. 2 Life cycle analysis of PFOA/PFO in 2012. Flows between processes are reported in tons

waste was calculated to be 0.3, 2.9, and 14.3 t, respectively. Most wastes were discharged into the hydrosphere and atmosphere, and emission was 78.1 and 25.6 t, respectively (Fig. S1). Such a large amount of direct discharge was caused by emission of wastewater from facilities for manufacture of PFOA/PFO and FP. This part of the wastewater did not enter into municipal WWTPs but would be treated in the WWTPs operated by the manufacturer. During production of



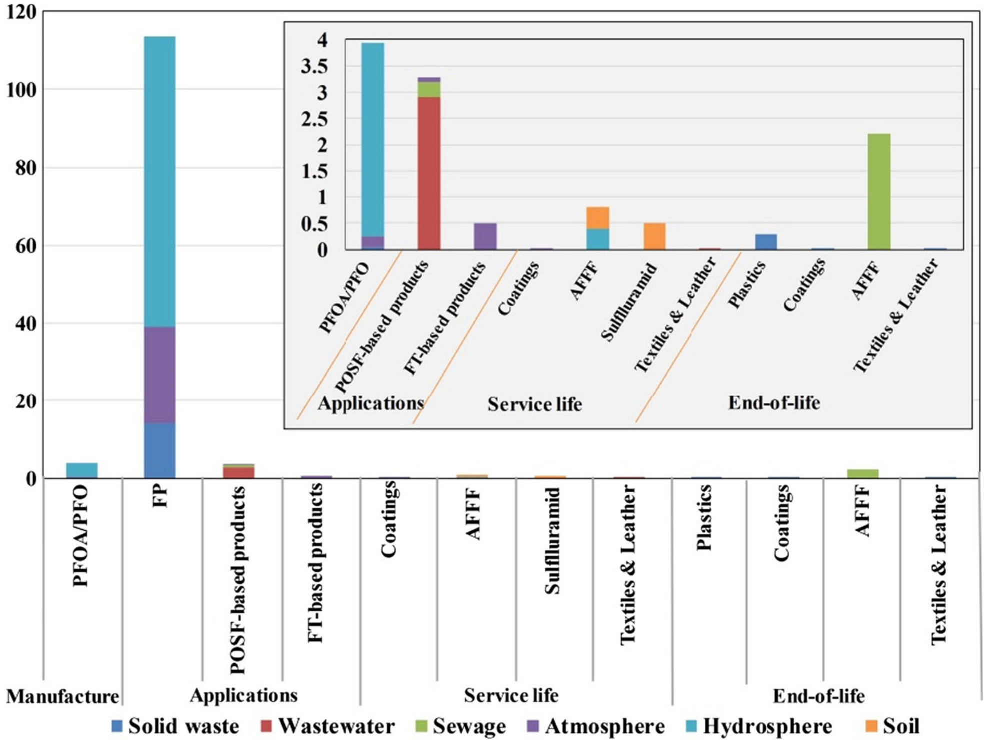
Location	Emission	River
Zibo	20.9	Xiaoqing River
Zigong	12.6	Fuxi River
Shanghai	6.7	Huangpu River
Taizhou	5.9	Yangtze River
Quzhou	5.3	Qujiang River
Jinan	2.5	Xiaoqing River
Fuxin	2.5	Daling River
Changshu	9.0×10^{-3}	Yangtze River

Fig. 3 Emission from manufacture of PTFE to the hydrosphere (t) in 2012

PFOA/PFO, residues generally entered into the hydrosphere (3.7 t), accounting for 94.4% of total wastes. It was noted that due to high emission from domestic manufacture of FP, usage of PFOA/PFO exceeded domestic production. On one hand, usage and emission from production and processing of FP might be overestimated based on emission from only one factory. On the other hand, part of PFOA/PFO was imported from the international market (Mei 2008). During production of FP, 80% of PFOA/PFO was discharged with wastes, especially wastewater to the hydrosphere (74.4 t), followed by waste gases (24.8 t) and solid waste (14.2 t). PTFE accounted for 75.8% of the FP output (Table S1). There were eight parks or facilities producing PTFE. One park located in Zibo was the biggest manufacturer, and emission to the hydrosphere from this park reached 20.9 t, followed by emission (12.6 t) from another park in Zigong (Fig. 3 and Table S3). It was noted that emission to the hydrosphere from the park in Changshu was only 9.0 kg because of strict requirement from *2010/2015 PFOA Stewardship Program*, although its capacity reached 11,600 t. In addition, the amount of PFOA/PFO emitted to the hydrosphere from the manufacture of other FPs was 18.0 t. Emission to the hydrosphere from the manufacture of PTFE induced a high pollution load of PFOA/PFO, especially in Xiaoqing River (23.4 t) which would flow into Bohai Sea after dilution. When FP was used in commercial products, almost all of the PFOA/PFO was decomposed into FP powder, with approximately 26% of the PFOA/PFO emitted in AFDs. Due to very little application of AFDs, only 0.06 t of PFOA/PFO was emitted during use of FP. During production and application of POSF-based substances, 3.4 t of PFOA/PFO was discharged with wastes, mainly from application of POSF-based substances in metal plating (3.0 t). During production and application of FT-based substances, mainly textiles and leather, PFOA/PFO with a volume of 0.5 t was discharged to the atmosphere (Fig. 4).

After PFOA/PFO-related compounds were used to manufacture various consumer products, residual PFOA/PFO remained in the products. During their use and disposal, PFOA/PFO was released. PFOA/PFO residues were estimated with residual proportion between usage of PFOA/PFO and total production and application. Among applications of these three kinds of PFOA/PFO-related substances, residues of PFOA/PFO in POSF-based substances were greatest, including those in AFFF and sulfluramid, followed by those in plastics and coating, while residues of PFOA/PFO in textiles and leather were the lowest due to the release during application (Fig. S2). During the service life of these products (Fig. S2), 1.3 t of PFOA/PFO was discharged into waste treatment facilities and the environment. Due to use of coating products at high temperature, most of the PFOA/PFO was released to the atmosphere (0.02 t) (Sinclair et al. 2007; Schlummer et al. 2015). After use of AFFF, PFOA/PFO directly entered into the hydrosphere and soil (0.8 t). It was assumed that PFOA/PFO along with sulfluramid entered the soil (0.5 t). All of the PFOA/PFO from coatings, AFFF, and sulfluramid (1.3 t) were directly emitted into the environment. Most of the PFOA/PFO (0.1 t) from textiles and leather was emitted into the sewage system (4.0×10^{-5} t), followed by those emitted to the

Fig. 4 Flows of PFOA/PFO (t) in production and use



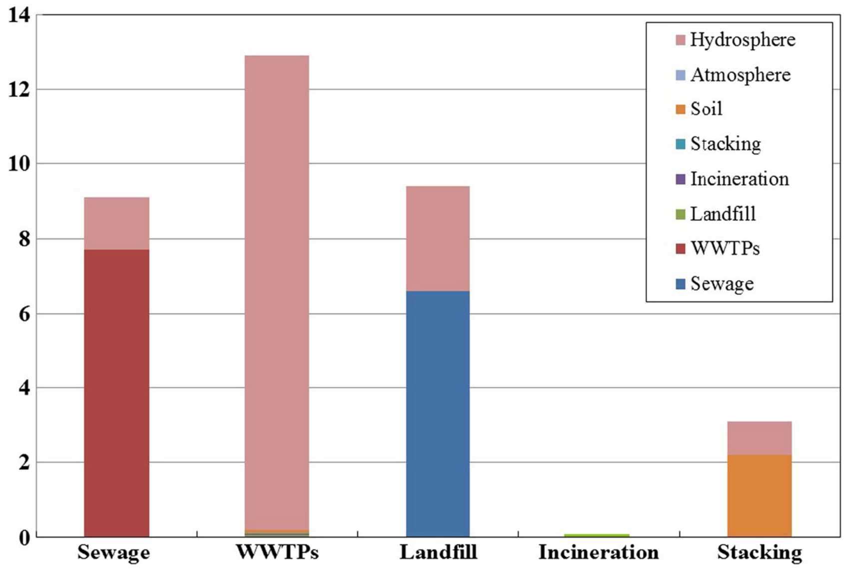
hydrosphere and soil (2.0×10^{-5} t). Considering friction resistance and corrosion resistance of FP-based products used in plastics, it was assumed that no PFOA/PFO entered the environment during use of plastics. Treatment of waste or expired products resulted in 2.5 t of PFOA/PFO being emitted into waste streams. PFOA/PFO from expired AFFF (2.2 t) entered sewage directly. For other products, including plastics, used coating products, textiles, and leather, PFOA/PFO (0.3 t) was released with solid waste (Fig. 4).

Flows in waste management In waste treatment facilities, most of the PFOA/PFO was generated during manufacture and application (Figs. 2 and 4). Waste management was primarily via wastewater and solid waste, and thus, most of the PFOA/PFO entered WWTPs, the sewage system, and landfill sites. Wastewater from industries was released to WWTPs, while those from domestic applications entered the sewage system and then passed through WWTPs. As a result, PFOA/PFO in sewage (2.5 t) and industrial processes (2.9 t) did not enter the environment directly, but rather entered WWTPs, where it received some form of treatment before entering the ambient environment. In WWTPs, pollutants can be released to the environment via treated effluent or sludge. Results of previous studies have found that conventional treatment was not effective in removing PFOA and that concentrations of PFOA even increased due to biodegradation of precursors (Guo et al. 2010), which resulted in greater emissions to the hydrosphere (12.7 t), while PFOA/PFO in sewage sludge was mainly transferred to solid waste treatment sites (0.1 t) or applied to agricultural soils (0.1 t) (Figs. 5 and S3).

Since there are no data concerning the disposal of industrial solid wastes, estimation of this portion of the flow was based on treatment of domestic solid waste. For treatment of solid waste and sewage sludge, incineration was found to be the primary disposal route in other countries, where most of the PFOA/PFO would likely be degraded. In Europe, majority of the sludge was disposed in landfills (~57%), followed by application as a fertilizer to land. Application of sludge in agriculture varied greatly among countries. For example, in Spain, 65% was disposed of in that manner, while in Germany, it was 30% (Gomez-Canela et al. 2012). Application of sewage sludge to land poses risks to soils. Results of a previous study showed that concentrations of all PFASs in soil increased linearly as a function of rates of loading of sludge (Sepulvado et al. 2011).

Results of more recent studies have demonstrated that landfills were sources of semivolatile PFASs to surrounding air and nonvolatile PFASs to landfill leachates (Ahrens et al. 2011; Huset et al. 2011; Benskin et al. 2012). Concentrations of PFOA in municipal landfill leachates in Germany ranged from <0.4 to 926 ng/L (Busch et al. 2010). Concentrations of PFOA in landfill leachate in the USA ranged from 177 to 2500 ng/L, with a mean of 820 ng/L (Clarke et al. 2015). When concentrations of PFASs were measured at landfill sites in five relatively developed cities in China, concentrations in leachate ranged from 7280 to 292,000 ng/L, and PFOA and perfluorobutane sulfonate (PFBS) were dominant. National leakage of PFASs to groundwater from landfill leachate was estimated to be 3110 kg/year, which indicated risks to groundwater (Yan et al. 2015). When wastes containing PFOA/PFO were placed into landfills, most of the PFOA/PFO entered the leachate, which resulted in 6.6 t of PFOA/PFO going into the sewage system and 2.8 t of PFOA/PFO flowing into the hydrosphere (Figs. 5 and S3). At stacking sites, it was assumed that leachate would be generated in quantities similar to landfill sites, even though the fate of leachates would have been different. Leachate from wastes at stacking sites would directly flow into soils (2.2 t) and be released to the hydrosphere (0.9 t), so stacking sites

Fig. 5 Flows of PFOA/PFO (t) in waste management



were considered to be important sources of emissions. At incineration sites, PFOA/PFO mostly degraded if the temperatures were sufficiently high. It was estimated that a relatively small amount (0.01 t) would be released and then enter into other pathways. Therefore, besides storage in WWTPs, landfills and stacking sites are also significant sources of PFOA/PFO.

Storage in the environment In China, the amount of PFOA/PFO stored in soils, the hydrosphere, and the atmosphere was estimated to be 3.2, 96.3, and 25.6 t, respectively (Fig. 2). PFOA/PFO existed predominantly in the hydrosphere and atmosphere, while little was stored in soils. For PFOA/PFO in China, the hydrosphere was also the largest sink. The only other assessment of flows of PFOA/PFO in the environment, conducted in Switzerland, suggested that the greatest proportion of PFOA/PFO (33.0 kg) was in the hydrosphere, with 0.2 kg in the atmosphere and 2.0 kg in soil (FOEN 2009). One significant difference between Switzerland and China was that there was no manufacture of PFOA/PFO and few applications of related products in Switzerland. In China, most of the PFOA/PFO in the atmosphere (25.4 t) was released from applications, namely manufacture and industrial use of FP and POSF- and FT-based products, accounting for 99.2% of total emission, and the others came from manufacture of PFOA/PFO. Most of the PFOA/PFO in the hydrosphere (74.4 t) also originated from applications, especially manufacture of FP, which was estimated to contribute 77.3% of the PFOA/PFO released to the hydrosphere, while WWTPs were an important source to the hydrosphere and contributed 13.2%, including emission of PFOA/PFO and transformation of precursors after treatment. Contribution from stacking sites to soil was the greatest (68.8%), followed by those from the service life of AFFFs and sulfluramid (28.1%) (Figs. 2 and S2). Contributions from industrial and domestic applications differed greatly. Most of the PFOA/PFO was released from industrial sources, which accounted for 97.0% of the total, most of which was released during production of FP. In China, landfill is the main method for treatment of solid wastes, and agricultural application is common for sewage sludge, which resulted in higher releases to soil compared to that in other countries (Wang 2009).

Inventories can be based on absolute masses, expressed as concentrations in each compartment or in relative terms based on proportions flowing along each pathway. Although emissions to environmental media were estimated, due to transport and transformation in these three media, relative concentrations of PFOA in the environment may be different. For example, PFASs in air might "flow" into soil and water via wet and dry deposition. Concentrations of PFASs have been measured in the environment in China, especially PFOA and PFOS (Wang et al. 2015). Compared with PFOS, which is already listed in Appendix B of the *Stockholm Convention*, current concentrations of PFOA were even higher in freshwater, especially in severely contaminated rivers, such as Daling River, Liaohe River, Hanjiang River, Yangtze River, and Huaihe River (Jin et al. 2009; Bao et al. 2011; Yang et al. 2011; Wang et al. 2013a; Yu et al. 2013). Concentrations of PFOA in lakes of China, such as Taihu Lake, Tangxun Lake, and East Lake, were similar to those reported for PFOS in the same lakes (Yang et al. 2011; Chen et al. 2012; Zhou et al. 2013). PFASs, especially PFOA and PFOS with concentrations ranging from tens of picograms to thousands of nanograms per liter of water, were widespread in the hydrosphere. Concentrations of PFOA in soil have been measured primarily in coastal areas of the Bohai Sea (Pan et al. 2011; Wang et al. 2012). Concentrations of PFOA and PFOS were the predominant PFASs in soils along the Bohai Sea, with concentrations of PFOA ranging from n.d. to 13.3 ng/g (Meng et al. 2015). Concentrations of PFOA in soils in the vicinity of Shanghai were relatively high, ranging from 3.3 to 47.5 ng/g (Li et al. 2010). While measurements of PFOA in air were few, one conducted in Shenzhen detected PFOA in all samples, with concentrations ranging from 1.5 to 15.0 pg/m^3 (Liu et al. 2015).

Alternatives or mitigation technologies for PFOA/PFO Through the LCA for production and use of PFOA/PFO and ultimate dispersion in the environment, it is possible to design more effective programs of monitoring and exposure assessment to determine which receptors would be most at risk. This information can also be used to mitigate risks by minimizing releases, including which applications of PFOA/PFO in various industries and which treatment of wastewater and solid waste resulted in the highest exposures (Fig. 6). Finally, the absolute and relative masses predicted to move along specific pathways allow for more effective mitigation of releases.

During production of PFOA/PFO, the most effective, currently available technology to reduce releases is vacuum distillation. While this would reduce releases, the reclaimed PFOA could be reused for production of organic fluoride, such as PTFE, which is an improved method compared to primary production using ECF (Tang et al. 2009). There are several alternative ways to minimize releases of PFOA during the application of PFOA/PFO. Owing to the presence of biological residues and slow degradation of C8 products, the development of alternatives has been focused on two technologies: use of low-carbon fluoride products and alternatives with heteroatoms, such as oxygen or chlorine (Lu et al. 2011). Currently, the 3M Company has implemented use of PFBS, which is a shorter-chain replacement for PFOS (3M Company 2007). Dupont uses telomerization to produce perfluorinated alkyl monomers, mainly C6-based products to replace C8 products (Lu et al. 2011). Perfluoropolyethers (PFPEs) have also been considered to be effective alternatives. For example, Dupont uses PFPEs or their salts as emulsifiers

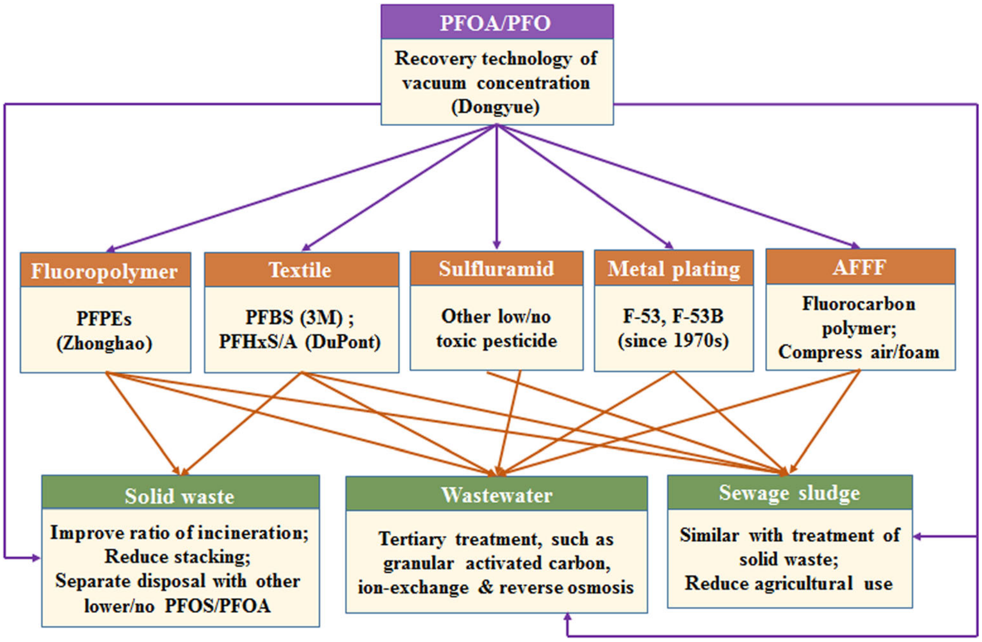


Fig. 6 Alternatives or mitigation techniques for PFOA/PFO. The *purple box* represents techniques used during manufacture of PFOA/PFO; *orange boxes* represent alternatives or mitigation techniques during application of PFOA/PFO; *green boxes* represent mitigation techniques during waste treatment; in *brackets* is the representative enterprise

during production of PTFE and Solvay Solexis uses PFPEs in food packaging materials. The skeleton of PFPEs has a double energy structure connected by an ether bond. And the average relative molecular weight is 1500. Therefore, PFPEs are absorbed less efficiently and have a shorter half-life compared to PFOA/PFOS, while they are not degraded into PFOA/PFOS (Gu et al. 2009). However, some reports have indicated the global warming potential of PFPEs due to the presence of C–O and C–F bonds (Young et al. 2006). With increased production and application of PFPEs, they may present a risk to global warming. In addition, it was noted that the manufacture of FP was the biggest source, and the difference of unit emission from manufacturers in Changshu Hi-tech Fluorochemical Industrial Park and other Chinese manufactures was very large. On one hand, manufacturers in Changshu applied a different processing technology and took waste recovery measures. On the other hand, this industrial park had established a circular economy industrial chain, while most of the other fluorochemical industrial parks only gathered together fluorochemical enterprises (Chen 2009). Therefore, the state should guide enterprises to change technology and set up waste recovery facilities, while enterprises should make full use of the industrial parks to reduce waste emissions. In the firefighting industry, fluorinated surfactants produced using the telomerization process were demonstrated to be effective alternatives to PFOS and PFOA. However, their risks to the environment due to widespread use in AFFF have not yet been assessed. Firefighting technologies using compressed air/foam have also been considered feasible (Tian et al. 2009). At the same time, considering that treatment of expired AFFF was the biggest source during use and disposal of consumer products, the state should establish a perfect recycling system and better treatment method. For metal plating, polyfluorinated ether sulfonates (PFAESs) have been used as alternatives for PFOS. A 6:2 chlorinated ether sulfonate (6:2 Cl-PFAES, $C_8ClF_{16}O_4S^-$) with the commercial name F-53B has been used in China since the 1970s (Ruan et al. 2015). F-53B has moderate toxicity based on the Globally Harmonized System (GHS) criteria for classification of chemicals and is as persistent as PFOS. Concentrations of F-53B were similar to those of PFOS in the ambient environment around metal-plating factories (Wang et al. 2013b). Although F-53B has been used for many years, the impact has yet to be fully assessed so increasing their application in replacement of PFOS should be advocated with caution.

Apart from technologies for reducing emissions or developing alternatives for use in applications, some mitigating measures can also be implemented during waste treatment. WWTPs were the most important receiving and discharging sites for PFOA/PFO due to poor removal efficiency by conventional wastewater treatment techniques, especially WWTPs in manufacturing sites for PFOA/PFO and FP. For example, the concentration of PFOA increased about seven times from influent (532 ng/L) to effluent (3630 ng/L) in the WWTPs of Jiangsu Hi-tech Fluorochemical Industry Park (Jin et al. 2015). Tertiary treatment techniques, such as granular activated carbon, ion-exchange, and reverse osmosis, have been proved to take some effect for its removal (Vecitis et al. 2009). Granular activated carbon was proved to effectively remove 90% of PFOA from effluent (USEPA 2004). However, it was also reported that the removal efficiency was strongly dependent upon the concentrations of pollutants, background organic, metal concentration, and available degradation time (Vecitis et al. 2009). If an effective removal method was generally applied in WWTPs of manufacturing sites and could remove half of the PFOA/PFO, the total emission to the environment could be reduced by about 31.2%. Incineration is the most effective treatment to remove PFOA/PFO in solid waste or sewage sludge. As long as the minimum temperature is maintained, this method is effective and results in complete mineralization of PFOA/PFO. For China, reduction in the use of wastewater for irrigation and sludge for agricultural use was the most cost-effective measure that could be taken to reduce accumulation of PFOA in soils.

Uncertainty analysis Uncertainty in the LCA for PFOA originates primarily from the limited data available for some of the required parameters such as production and consumption, as well as determination of transfer coefficients and emission factors. Transfer coefficients and emission factors were mainly determined through experimental data, with consideration of the practical situation in China. This, of course, made the assessment specific to processes and storage in environmental media for China. Since there were no related statistical or empirical experimental data on treatment of industrial solid waste, it was assumed that industrial solid waste was treated the same as domestic solid waste. In addition, there may be a little deviation for residuals of PFOA/PFO in commercial products due to limited data sources. The biggest uncertainty was estimation accuracy for emission from manufacture of FP. This analysis was only based on one manufacturer in Zibo, Shandong, and its emission was estimated by concentrations of PFOA in outlet and flux. If this analysis was based on reports of Li et al. and Wang et al., the emission from the FP manufacture was 25.0 and 40.0 t, respectively, though they were made with similar estimation methods (Wang et al. 2014; Li et al. 2015).

Conclusions

- Emission of PFOA/PFO from industrial sources (production and processing of industrial products) was much higher than those from domestic sources (use and treatment of consumer products), among which the former was 121.2 t and the latter was 3.8 t.
- During production and processing of industrial products, 113.4 t of PFOA/PFO came from production of FP, among which 74.4 t of PFOA/PFO after treatment of industrial WWTPs flowed into rivers nearby these manufacturers. The load of Xiaoqing River was the biggest. During use and treatment of consumer products, PFOA/PFO in expired AFFF was the biggest contributor with an amount of 2.2 t.
- In waste management, more PFOA/PFO flowed into wastewater with an amount of 19.7 t, including those in the sewage system and municipal WWTPs. And others entered into solid waste treatment plants with an amount of 14.7 t, among which 9.4 t of PFOA/PFO existed in landfills. After waste treatment, 2.3 and 17.8 t of PFOA/PFO were emitted into soil and the hydrosphere.
- Masses of PFOA/PFO in 2012 were estimated to be 96.3 t to the hydrosphere, 25.6 t to the atmosphere, and 3.2 t to soils.

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Supporting Information for

Life cycle analysis of perfluorooctanoic acid (PFOA) and its salts in China

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S1. Production of fluoropolymer (FP)

Production capacity and actual output of FP in 2010 were approximately 80,000 t and 60,000 t, respectively (CAFSI 2011). Actual output was approximately 75% of the production capacity. Based on production capacity of 78,900 t for polytetrafluoroethylene (PTFE) (ChinaIOL 2012), it was estimated that 59,175 t of PTFE was produced in 2012. In addition, based on the same proportion of PTFE in 2012 and 2015 and import/export volumes, approximately a total of 78,116 t FP was produced in 2012 (Table S1).

Table S1 Production, import, export and consumption volume (t) of FP in 2012

	Production	Import [a]	Export [a]	Consumption	Consumption forecast (2015)[b]
PTFE	59,175[c]	4,200	21,000	42,375	56,794
Other FP	18,941	8,100	8,400	18,641	24,984
Total	78,116	12,300	29,400	61,016	81,778

a. China customs statistical yearbook in 2013;

b. National 12th five-year plan for fluorine chemical industry;

c. 75% production capacity.

S2. Transfer coefficients (TC) and emission factors (EF) during production of PFOA/PFO

Currently, electrochemical fluorination is still using primary technology to produce PFOA, although telomerization has been the predominant method in North America and Europe since 2002. The purity of PFOA products was relatively high

(94.0-95.8%), and the major impurity was PFOS (2.06-3.09%) (Jiang et al. 2015). It also proved that isomer structure of PFOA products in China was similar with that in 3M Company. Therefore, the same transfer coefficients and emission factors were used. PFOA/PFO from production and industrial applications would be released with wastewater, gas and solid waste, and then enter WWTPs, air and disposal sites for solid waste. It was noted that wastewater in PFOA manufacturing factories was treated in inner WWTPs, and then discharged into rivers. The process in WWTPs was unknown, so only the process from manufacture to emission to the hydrosphere was considered. According to a report from 3M Company, a total of 885, 20,412 and 227 kg of PFOA/PFO were released to air, water and solid waste with the production of 498,960 kg PFOA/PFO (3M Company 2000). Assuming that the same production technique was used, the corresponding emission factor for solid waste and releases to water, air were 0.0005 and 0.04, 0.002, respectively (Table S2).

Table S2 TC and EF during production of PFOA/PFO

	TC solid waste	EF hydrosphere	EF atmosphere
Best estimate	0.0005	0.04	0.002

S3. TC and EF during industrial applications

FP manufacture. PFOA from production of FP firstly entered WWTPs in fluoride factories, and then was discharged into rivers after treatment. There were no studies on PFOA/PFO from internal WWTPs in fluoride factories. The process of PFOA/PFO from FP manufacture to rivers, however, was clear and the corresponding EF was available. It was estimated that annual emission of PFOA to river from fluorochemical industry park in Zibo, Shandong, was 58.0 t for 2013 based on concentrations in environmental samples (Wang et al. 2016). Currently, the main products of this park include PTFE (capacity, 44,300 t), polyvinylidene fluoride (PVDF of capacity, 8,400 t), fluorinated ethylene propylene (FEP of capacity, 5,500 t), fluororubber (FKM of capacity, 10,000 t) and PFOA (capacity, 30 t) (Dongyue Federation 2015). It was assumed that actual output of all products was approximately 75% of the production capacity. Therefore, approximately 0.9 t of PFOA/PFO from PFOA manufacture was discharged into Xiaoqing River based on EF to water (0.041). Approximately 57.1 t of PFOA/PFO was released from FP manufacture, with actual output of 51,150 t. Based on the same emission factor, emission of PFOA/PFO from other manufacture factories were also estimated (Table S3). It was noted that within the total capacity of 78,900 t of PTFE, 11,600 t was from non-Chinese manufacturers in Changshu, Jiangsu (including DuPont, Solvay, Daikin, etc.) (Table S3). For these PTFE manufactures in Changshu, Jiangsu, it was reported that concentration of PFOA in effluents from WWTPs was 3,630 ng/L (Jin et al. 2015). The total emission of wastewater of Jiangsu Hi-tech Fluorochemical Industrial Park was 2.5×10^9 L (Jiangsu Hi-tech Fluorochemical Industry Park 2016), so annual emission of PFOA was 9.0 kg. It was far lower than those from Dongyue, Shandong, and likely due to the restriction and requirement of *2010/2015 PFOA Stewardship Program* (USEPA 2006). Assuming the same emission factor to the hydrosphere for PTFE and other FP, it was estimated that emission of PFOA/PFO from other FP manufacture was 18.0 t.

According to an investigation by the Fluoropolymers Manufacturers Group (FMG), approximately 64% PFOA/PFO were released with wastes, including 14% to air, 42% to wastewater and 8% to solid waste, while the remaining residues would be destroyed, re-processed and remained in products (Wang et al. 2014). Volumes emitted via various routes were similar to those estimated previously (Prevedouros et al. 2006). Since no technologies to reduce emissions have been applied in China, it has been estimated that as much as 80% of PFOA-based emulsifiers eventually enter the environment (Wang et al. 2014). Assuming a constant distribution ratio in the environment, emission factors to solid waste, the hydrosphere and the atmosphere were calculated to be 0.10, 0.53 and 0.17, respectively (Table S4).

Table S3 Production capacity of PTFE and emission to the hydrosphere (t) in 2012

Location	Capacity	Emission	River
Zibo, Shandong	25,000	20.9	Xiaoqing River
Zigong, Sichuan	15,000	12.6	Fuxi River
Shanghai	8,000	6.7	Huangpu River
Taizhou, Jiangsu	7,000	5.9	Yangtze River
Quzhou, Zhejiang	6,300	5.3	Qujiang River
Jinan, Shandong	3,000	2.5	Xiaoqing River
Fuxin, Liaoning	3,000	2.5	Daling River
Changshu, Jiangsu	11,600	$9.0*10^{-3}$	Yangtze River
Total	78,900	56.4	

Table S4 TC and EF during FP manufacture

	TC solid waste	EF hydrosphere	EF atmosphere
Best estimate	0.10	0.53	0.17

Use of aqueous fluoropolymer dispersions (AFDs). PTFE products are of three types, namely granular molding powders, paste extrusion powders (fine powders) and AFDs, which account for 72.8%, 19.7% and 2.0% of overall consumption in China, respectively (Company LJ 2011). Considering that powder products would usually undergo high-temperature melting or sintering treatment to produce plastics and only a little PFOA/PFO remained in products (1~10 ppm) (Cope 2005), releases to the environment during use of powders were deemed to be negligible. Therefore, only release from use of AFDs was estimated. AFDs are mixed with fine-granular FP resins dispersed in solution that are typically of 60% solid content. In China, almost all AFDs are used for low-temperature (<300℃) thermal spraying, dipping, and impregnation to coat metals, asbestos, glass-fiber woven and fabric surface (Wang 2006). In PTFE aqueous dispersions, it has been estimated that there is 125 ppm of PFOA/PFO residuals in coating products used in China (Li et al. 2015). According to a previous mass balance analysis, an average of 62% of PFOA/PFO residual decomposed, with the remaining 5%, 5% and 16% of residuals released to water, soil and air, respectively (FMG 2003). Corresponding emission factors to WWTPs, solid waste and hydrosphere were estimated to be 0.05, 0.05 and 0.16, respectively (Table S5). The environmental release from use of AFDs was calculated (Equation S1) (Li et al. 2015).

Environmental releases = Consumption volume of AFDs × PFOA/PFO content in commercial AFD products × TC (EF) (S1)

Table S5 TC and EF during use of AFDs

	TC WWTPs	TC solid waste	EF hydrosphere
Best estimate	0.05	0.05	0.16

Production of perfluorooctane sulfonyl fluoride (POSF)-based substances. On average ~150 t of POSF-based substances have been produced annually in China (Li et al. 2015). The estimated emissions into water and air during production were 0.55~3.5 t and 1.0~1.4 t, while the production of PFOS equivalents was approximately 220~240 t in China. The same emission factors were applied for direct sources, namely 0.0024~0.015 to water and 0.0045~0.0060 to air (Xie et al. 2013). In the LCA, results of which are presented here, emission factors of 0.009 to wastewater and 0.0053 to the atmosphere were used. It was reported that purity of PFOS products in Chinese market was only 76.7~80.6%, and PFOA contributed more than 10% to PFOS products (Jiang et al. 2015). Therefore, it was assumed that the contents of PFOA in POSF-based substances were about 10%. In terms of indirect sources of POSF-equivalents released during production of POSF-based substances, excluding POSF, PFOS and its salts, the emission factors of precursors were set as 1×10^{-5} for air and 0.0003 for water (Wang et al. 2014). Based on assumptions and calculations made previously (Li et al. 2015), the transformation rate of POSF-based precursors was

0.050~0.268. A value of 0.16 was used.

Industrial use of POSF-based substances. According to an industrial investigation by the China Association of Fluorine and Silicone Industry (CAFSI), ~50% of currently produced POSF-based substances were exported and the remaining 50% were used in the domestic market, mainly including metal plating, aqueous fire-fighting foams and sulfluramid (CAFSI 2013). Historically, larger proportions of POSF-based substances were used in surface treatments for textiles, but this application ceased after 2009/2010 due to import restrictions applied by other countries. Because they were found to be few, applications of POSF-based substances in other sectors, such as semiconductor industry, were not estimated in the present study.

Ⅰ: ~20% of POSF-based substances were used as mist suppressants during plating of metals. In this application, active ingredients were mainly PFOS and its salts, which are not precursors of PFOA/PFO (Mei 2008). Therefore, only direct releases of PFOA/PFO impurities from the discharge of POSF-based mist suppressants were estimated. According to estimation on emissions of PFOS from metal plating, approximately 34 t of PFOS, almost 100% was discharged during 30~40 t PFOS using for metal plating (Zhang et al. 2012). Metal plating plants are mainly distributed in east coastal cities, especially in Pearl River Delta and Wenzhou of Zhenjiang Province. In the developed cities in east China, new or reconstructed metal plating plants are required to increase the reuse rate of metal plating wastewater to 50%~90% (Li 2011). However, metal plating wastewater is only treated by using traditional methods and directly discharged after meeting the standards in most plants. In a few plants, treated wastewater is only used to flush toilet or as landscape water, and finally gets into sewage system. Based on a conservative estimate, 90% PFOA/PFO in metal plating sector is discharged into wastewater and 10% into sewage by reuse.

Ⅱ: ~20% of POSF-based substances were used as fluorocarbon foamers during the production of AFFFs. Active ingredients were mainly non-ionic or amphoteric fluoroalkylamide derivatives, among which not all components could degrade to PFOA/PFO (Mei 2008). For simple estimation, direct sources were based on estimates made previously, where 2% and 0.1% of POSF-based products were calculated to be released to wastewater and air, respectively (Xie et al. 2013).

Ⅲ: 5% of POSF-based substances were used as raw materials to produce sulfluramid. The active gradient was *n*-ethyl perfluorooctane sulfonamide (Et-FOSA), and its gross loss was no more than 0.1% based on previous investigation (Li et al. 2015). Here, a value of 0.1% loss was assumed to be discharged to wastewater.

Production of fluorotelomer (FT)-based substances. At present, domestic production of FT-based substances includes two routes, namely de novo production and secondary processing, among which long-chain perfluroalkyl iodide (PFOI) (telomer A) as a raw material imported from aboard is applied during secondary processing. Based on previous assessments and calculations, approximately 600 t of telomer A were annually imported (Chen 2010), which could be used to produce approximately 3,000 t of FT-based substances (Prevedouros et al. 2006). According to an industrial investigation by CAFSI, domestic de novo production of FT-based substances was 1,500 t from two local manufactures (Li et al. 2015). For *de novo* production, an emission factor (in tons released FTOH-equivalents per tons of produced FT-based products) was estimated to be 2.5×10^{-5} for air (Wang et al. 2014). Concentrations of PFOA/PFO in FT-based products ranged from <1 to 100 ppm (TRP) and 10 ppm was used. Rates of transformation of FT-based precursors to form indirect sources were calculated to be 0.059%~0.59% (Li et al. 2015). For secondary processing, the emission factor was set to be half of that of *de novo* production (Table S6).

Industrial use of FT-based substances. In China, most FT-based substances were applied as finishing agents for surface treatment of textiles or leather. Polymeric FT-based substances are synthesized from FT-based monomers by polymerization, during which unpolymerized FT-based monomers remain as residuals and PFOA/PFO impurities appear in the ultimate polymeric FT-based substances. It has been estimated that contents of precursors and PFOA/PFO impurities were 3.8% and 0.36 ppm in the final polymeric FT-based products, respectively (Danish EPA 2008). During finishing of textiles or leather, most of the FT-based substances are affixed to the treated materials after thermal impregnation and drying, and almost 100% of FTOH residuals and 95% of PFOA/PFO impurities were released to air (Buck et al.

2005). The environmental releases from production and use of POSF/FT-based substances were calculated (Equation S2) (Li et al. 2015). And the corresponding TC and EF were listed in Table S6.

Environmental releases = Amount of POSF/FT-based substances × [TC (EF)×PFOA/PFO content + TC (EF) of precursors × (PFOA/PFO content + transformation rate)] (S2)

Table S6 TC and EF during industrial applications

		Direct releases			Indirect releases		
	PFOA/PFO content	TC WWTPs	TC Sewage	EF atmosphere	TC WWTPs	EF atmosphere	Transformation Rate
Production [a]	10%	0.009		0.005	0.0003	1.0×10^{-5}	0.16
Metal plating		0.9	0.1				
AFFFs		0.02		0.001			
Sulfluramid					0.001		0.16
Production [b]	10 ppm			0.0003			
Tex.& Lea. [c]	0.36 ppm			0.95			
	3.8% [d]			1.0			0.003

a. production of POSF-based products;

b. production of FT-based products;

c. textiles & Leather;

d. content of precursors.

S4. PFOA/PFO residues in commercial products

FP-based products. It was noted that only a little PFOA/PFO, 1~10 ppm, remained in plastics after high-temperature treatment (Cope 2005). However, considering that PFOA/PFO used in plastics (98%) were huge compared to those used as AFDs, there was approximately 0.3 tons of PFOA/PFO remaining in plastics, which might be discharged during use and treatment of plastics. For PFOA/PFO in coatings, approximately 12% PFOA/PFO entered into final products (FMG 2003), namely 0.03 t of PFOA/PFO remaining in coatings.

POSF-based products. In metal plating, almost all PFOA/PFO was discharged into environment during treatment, so the contents of PFOA/PAO in final electroplating products were negligible. During production of AFFFs, approximately 2% of PFOA/PFO were discharged (Xie et al. 2013), so it was estimated that 98% (2.9 t) existed in products without considering degradation of precursors. Similar with that for AFFFs, almost all PFOA/PFO (0.8 t) entered into final sulfluramid due to minimal losses during production (Li et al. 2015).

FT-based products. During finishing of textiles and leather, it was estimated that 95% PFOA/PFO and 100% FTOHs were discharged to the atmosphere (Buck et al. 2005). Therefore, only 5% of PFOA/PFO (8.1×10^{-5} t) remained in final textiles and leather.

S5. TC and EF during service life of products

Plastics. Plastics were mostly used in electric appliance, chemical industry, aviation and machinery. Because of FP stability, including anti acid, alkali, organic solvent, high temperature resistance, and friction resistance, it was assumed that loss of PFOA/PFO during use of such plastics was negligible.

Coating. Although only a small fraction of PFOA/PFO in AFDs enter into coated products, PFOA/PFO is still released to the atmosphere during use of coated products. One study reported that mass of PFOA in nonstick coating was 59 to 1237 pg/cm, with a release of 11-503 pg/cm^2 during an experiment carried at 250℃ for 20 min (Sinclair et al. 2007). And it also showed that release of PFOA decreased with multiple uses. Another study investigated release of PFOA from different nonstick cookware. The results showed that releases varied greatly, and those from pans were relatively higher and those from waffle irons were relatively lower (Schlummer et al. 2015). This may be caused by different residual PFOA in cookware or different operation temperature. However, there were no related information on residual PFOA in eliminated coating products. Considering different types of cookware and reduced release during constant use, an emission factor to atmosphere of 0.7 was used during service life (Table S7).

Table S7 TC and EF during service life of coating products

	TC product	EF atmosphere
Best estimate	0.3	0.7

AFFF. Considering actual situation in China, residual liquid was not treated and all released into environment after use of fire-fighting extinguishers. Based on the assumption of the Environment Agency that there is no containment of foams applied to fight fires, 50% is estimated to go into the hydrosphere and 50% into soil (Brooke et al. 2004). According to investigation by Fire Department of Ministry of Public Security of China, cumulative production of AFFFs was 24,224 t and inventory was 18,259 t from 2001 to 2008 (Yu et al. 2010). Therefore, usage of AFFFs during this period was 5,965 t. The fraction of the AFFF stock annually used by fire-fighting services is 24.6%.

Table S8 TC and EF during service life of AFFF

	EF hydrosphere	EF soil
Best estimate	0.5	0.5

Sulfluramid. It was known that sulfluramid was mainly used to control termites and other crawling insects (Goosey and Harrad 2011). According to average validity in China, approximately 35% pesticide acted on crops and most (65%) flowed into soil (Table S9), when pesticide was used on agriculture (MOA 2015).

Table S9 TC and EF during service life of sulfluramid

	EF soil
Best estimate	0.65

Textiles & Leather. Based on the results of an investigation by 3M Company, it was concluded that cleaning garments with over a 2 year life span is expected to lose 73% of surface treatments containing PFOA related substances (UNEP 2006). Loss from textiles and leather mainly included two processes, namely washing and wear outside, which induced emission of PFOA/PFO into sewage system and environment. If it was wore or used outside, PFOA/PFO was mainly released with particles. According to estimation in UK by its Environment Agency, approximately 75% was released to soil and 25% to hydrosphere along with particles. While other report showed that 25% lost during outside wear (Brooke et al. 2004). Assuming similar process of leather and textiles, transfer coefficient to sewage system was estimated as 0.48, and emission factors were 0.19 to soil and 0.06 to hydrosphere, respectively (Table S10).

Table S10 TC and EF during service life of textiles & Leather

	TC sewage	EF soil	EF hydrosphere
Best estimate	0.48	0.19	0.06

S6. TC and EF at end-of-life of products

Disposal of solid waste. In 2012, the total amount of urban domestic refuse was 163.95 million tons, and harmless treatment was 130.89 million tons in China, of which treatment of sanitary landfill, incineration and compost were 100.63, 25.99 and 4.26 million tons, respectively. Other 20.1% was only stacking and simple landfill (Table S11) (CAEPI 2014). Currently, compost in China is still a simple operation at landfills. At present, urban solid waste in China has been increasing at an annual rate of 10%, and production per capita is approximately 450~500 kg (Zhang et al. 2011).

Table S11 Disposal routes of solid waste

	Incineration	Landfill	Stacking
Proportion	15.9%	64.0%	20.1%

Plastics. Due to negligible loss of PFOA/PFO during service life of plastics, it was assumed that all PFOA/PFO remained and entered into end-of-life. It was reported that a total amount of used plastics reached 200 million tons in 2011 and rate of recovery was below 10%, among which most were plastic box and packaging (Tang et al. 2013). Therefore, it was considered that such used plastics containing PFOA/PFO were treated as solid waste (Table S11).

Coatings. According to the Annual Report on Comprehensive Utilization of Resources (NDRC 2012), the amount of recycled copper, lead and aluminum accounted for 50%, 23% and 42% of each total yield, respectively. Considering that cookware were mainly recycled as metals, a total utilization ratio of 30% was used (Table S12). It was considered that high temperature treatment was operated during reuse. So most PFOA would degrade and a small portion was released to the atmosphere, where the emission factor was determined as same as those in incineration. The remaining 70% metals would be treated as solid waste.

Table S12 TC and EF at end-of-life of coating products

	TC solid waste	EF atmosphere
Best estimate	0.7	0.00003

AFFF. In addition to release during actual fighting of fires, PFOA/PFO is discharged due to disposal of expired AFFF. For example, in the Netherlands, expired foam from mobile AFFF (~ 5 year life span) tends to be dumped directly into sewers (Armitage et al. 2009). It was assumed that AFFF in petrochemical enterprises, gasoline stations, fire training bases and military bases were all in validity and expired AFFF would be disposed on schedule. It was calculated that approximately 75.4% of AFFFs were need to treated and this part would all be discharged into sewers (Table S13).

Table S13 TC and EF at end-of-life of AFFF

	EF sewage
Best estimate	1.0

Textiles & leather. Generation and discharge of solid waste by the textile industry were 6.9 and 1.1 million tons. In 2011, the total volume of waste textiles was approximately 26 million tons, of which only 2.3 million tons of waste textile was recycled (NDRC 2012). Other waste textiles were disposed of as municipal solid waste. At present, domestic recycling of waste textiles is based on recovery from small traders or donations, centralized recovery in the residential area, recovery at big sites and reuse. After simple physical treatment, waste textiles are mainly used to produce fiberfill, felt, mops and similar products. All of these at end-of-life products are then treated as solid wastes no matter for recycled or non-recycled textiles (Table S11).

S7. TC and EF in sewage system

Two types of losses from sewer system are possible, namely sewer overflows and exfiltration. The former is caused by strong or long-lasting precipitation events, and the latter refers to loss from leaking pipes. Currently, most cities don't achieve rainwater and sewerage diversion. Once storms occur, rainfall exceeds the capacity of sewage system, which finally leads to untreated sewage into hydrosphere. Combined the occurrence time of rainstorm and volume of sewage overflow, 1% load is assumed for sewer overflows (FOEN 2009).

Most sewage pipelines were conducted in the 1960s or 1970s. Until 2010, the total length of sewage pipelines was 370,000 km in China. Most pipelines are made of concrete, which can result in exfiltration of sewage into groundwater once crack. According to the standards for allowing exfiltration, 48 m^3/d/km is made of D800 mm reinforced concrete pipe (Wang and Yu 2003). Based on an assumption that all sewer pipelines are D800 mm reinforced concrete pipe, 17.8 million m^3 sewage is predicted to be released underground every day. In 2010, total discharge of sewage was 61.7 billion tons in China (MEP 2010). While the exact density of sewage varies and is unknown, using the density of pure water, exfiltration accounts for approximately 10.5% of total sewage. According to the worst-case scenario, it was assumed that 85% of PFOA/PFO entered WWTPs and 15% was directly into hydrosphere (Table S14).

Table S14 TC and EF in sewage system

	TC WWTPs	EF hydrosphere
Best estimate	0.85	0.15

S8. TC and EF in wastewater treatment plants (WWTPs)

In China, the total volume of wastewater discharged in 2012 was 68.5 billion tons, including 22.2 billion tons of industrial wastewater and 46.3 billion tons of household wastewater (NBS 2013). WWTPs are unable to reduce concentrations of PFOA/PFO in effluent significantly compared to concentrations in influent. In several studies, even an increase in concentrations or mass flows from the influent to the effluent has been found and attributed to degradation of precursor substances (Table S15). Data from different WWTPs, however, vary significantly.

Table S15 Concentrations and flows of PFOA in WWTPs in China

	Site	Capacity	Type	Technology	Influent	Effluent	Sludge	References
Concentration		10^4 m^3/d			ng/L	ng/L	ng/g	
	Beijing 1	100	D	CAS	9.2	13.6		(Pan et al. 2011a)
	Beijing 2	20	D	OD	2.7	10.7		
	Beijing 3	40	D	A2O	36.0	19.0		
	Beijing 4	10	D	MBR	1.1	1.4		
	Beijing 5	8	D	SBR	0.9	0.9		
	Beijing 6	60	D	A2O	12.0	12.5		
	Beijing 7	4	D	CO	0.09	0.3		
	Beijing 1	100	D	CAS	3.6	n.d.		(Zhang et al. 2013)
	Beijing 2	10	D	MBR	4.3	n.d.		
	Beijing 3	4	D	CO	1.5	2.6		

Beijing 4	20	D	OD	5.3	8.5
Beijing 5	40	D	A2O	2.9	3.9
Beijing 6	8	D	SBR	4.6	6.3
Beijing 7	90	D	A2O	6.7	6.4
Bengbu 1	10	D	A2O+OD	3.3	5.1
Hefei 1	30	D	OD	45.7	34.4
Hefei 2	10	D	OD	3.1	6
Hefei 3	18	D	OD	4.4	5.5
Guangzhou 1	64	D	Modified A2O	13.8	7.3
Guangzhou 2	20	D	Modified A2O	9.6	14.7
Guangzhou 3	15	D	A2O	15.3	15.3
Foshan 1	20	D	A2O	8.6	9.5
Wuhan 1	15	D	Modified A2O	6.2	6.4

	Wuhan 2	15	D	A2O	6.9	6.1	
	Dalian 1	8	D	LINPOR	8.9	15.5	
	Dalian 2	7	D	A2O	5.3	5.6	
	Shanghai 1	8	D	CAS	7.9	34.1	
	Shanghai 2	6	D	PASF	90.6	106.6	
	Shanghai 3	1.7	Both	CAS	58.2	67.8	
	Ningbo 1	16	D	A2O	10.9	10.2	
	Ningbo 2	6	D	OD	12.2	15.3	
	Nanjing 1	64	Both	A/O	5.6	13.5	
	Nanjing 2	30	D	CAS	7	8.4	
	Wuxi 1	30	D	MBBR	15.3	23.6	
	Wuxi 2	15	D	Modified A2O	10.1	26.1	
	Shenyang 1	38	70% D/30% I	BAF	34.5	28.0	(Sun et al. 2011)

	Shenyang 2	20	D	SCAS	71.1	41.1		
	Shenyang 3	40	60% D/40% I	CAS+A/O	26.6	32.5		
	Shenyang 4	2	D	CW+FBB	26.2	18.4		
	Taian 1	5×10^4 t/d	D	AGC+A2O	2.47	0.87		(Wang et al. 2011)
	Taian 2	8×10^4 t/d	Main I	OD	3.30	2.24		
	Beijing 1		D	A2O+MBR	6.6	6.8		(Fan 2010)
	Beijing 2		D	A2O	49.6	26.4		
	Beijing 3		D	OD	94.3	16.4		
Mass flow (g/d)	Dalian 1	6	D	CAS	1.0	0.6	n.d.	(Chen et al. 2012)
	Dalian 2	12	D	BF	1.2	1.2	0.02	
	Dalian 3	8	D	CAS+BF	3.5	0.3	n.d.	
	Dalian 4	1	30% D/70% I	A2O	188	100	0.23	
	Shanghai 1	7.5	D	CAS	6.2	6.2	0.12	

Shanghai 2	200	D	A2O	224	1,062	1.9
Shanghai 3	170	D	CF	325	202	1.0
Shanghai 4	9.5	30% D/70% I	A2O	6,288	15,657	4.0
Guangzhou 1	11	D	A2O	1.0	1.0	0.04
Guangzhou 2	3	D	CAS	0.9	0.5	0.01
Guangzhou 3	20	D	A2O	0.8	1.3	0.05
Guangzhou 4	30	D	A2O	0.8	0.8	0.09

D: Domestic wastewater; I: Industrial wastewater; CAS: Conventional activated sludge process; BF: Biofilm process; A2O: Anaerobic/anoxic/oxic process; CF: Chemical flocculation; MBR: Membrane bio-reactor; OD: Oxidation ditch; PASF: Removing phosphorus and nitrogen combined activated sludge and filter technology; SBR: Sequencing batch reactor; MBBR: Moving bed biofilm react; CO: Contacting-oxidation; SCAS: Suspended carrier activated sewage; AGC: Aerated grit chambers; BAF: Biological aerated filter; CW+FBB: Constructed wetland+ Floating bio- bed.

Statistical result: For EF, 56 values were contained. The mean and median values were 1.28 and 1.03, respectively. A best estimated emission factor of 1.2 was used for the model. For TC, 12 values were contained. The mean and median values were 0.02 and 0.01, respectively.

Mean and median values of EF were 1.28 and 1.03, respectively. A best estimate emission factor to the hydrosphere of 1.2 was used in the model. The mean value for calculation of transfer coefficient to sewage sludge was 0.02. Due to limited data, which was taken from a single study, there is uncertainty in this transfer coefficient.

Disposal of sewage sludge. Due to political and economic incentives, application in agricultural land and garden land (48.3%), used as fertilizer is still the main method of disposal, followed by landfill (34.5%) (Wang 2009). For this reason, PFOA/PFO would be expected to be transported from sludge to soil. Compared with massive incineration in other countries, in China the proportion of incineration was only 3.5%. The remaining 13.8% was untreated, which was assumed to remain at stacking sites (Table S16). Transfer coefficients to soil, incineration and landfill were calculated by multiplying the transfer coefficient to sewage sludge by the disposal ratio of sludge (Table S17).

Table S16 Disposal routes of sewage sludge

	Incineration	Landfill	Stacking	Soil
Proportion	3.5%	34.5%	13.8%	48.3%

Table S17 TC and EF in WWTPs

	TC incineration	TC landfill	TC stacking	EF hydrosphere	EF soil
Best estimate	0.0007	0.007	0.003	1.2	0.01

S9. TC and EF in incineration plants

PFOA/PFO could not be detected after incineration of solid waste, such as paper, textile and carpet. These results were interpreted due to effective removal or destruction of PFOA/PFO during incineration (Taylor et al. 2014). The actual temperature in China could reach 850℃, so PFOA/PFO could be effectively removed (Zhang 2015). The atmospheric emission factor and transfer coefficient to landfill were 0.0001 and 0.005, respectively (FOEN 2009), which was derived as the half of maximum EFs and TCs in simulation experiment on laboratory scale or actual monitoring in incineration plants (Table S18).

Table S18 TC and EF in incineration plants

	TC landfill	EF atmosphere
Best estimate	0.005	0.0001

S10. TC and EF in landfills

Atmospheric emission. It was reported that the annual emission of polychlorinated biphenyls (PCBs) to atmosphere was 95 kg when the total storage was 170,685 t in sludge (Yi et al. 2008). Therefore, the EF was 5.6×10^{-7}, and this value was also used in estimation of PFOA/PFO because of similar properties.

Landfill leachates. Landfills are classified as three types, including simple landfill, controlled landfill and sanitary landfill. Simple landfills refer to those without any environmental protection measures. Controlled landfills refer to those which could not fully meet the environmental standards. Sanitary landfills have complete engineering measures and could fully comply with the standards. The proportions of three types of landfills were 50%, 30% and 20%, respectively (Baidu 2015).

It was assumed that PFOA/PFO in landfill sites would eventually enter into leachate and air. The maximum percentage of landfill leachates collected and introduced to sewage system reaches 90% for sanitary landfill (E20 Platform 2015). Considering complex composition in leachate and different treatment technologies, the overall percentage collected to sewage was estimated to be 70% (Table S19).

Table S19 TC and EF in landfills

	TC sewage	EF atmosphere	EF hydrosphere
Best estimate	0.7	5.6×10^{-7}	0.3

S11. TC and EF in stacking sites

Leachate also emerges from stacking sites compared with that in landfill plants. The difference is that the leachate can’t be effectively collected and then it is released into the sewage system. It is assumed that 70% leachate in stacking sites enters soils, which is considered similar to that part flowing into sewerage system in landfill plants. The remaining 30% is assumed to enter into hydrosphere with runoff (Table S20).

Table S20 TC and EF in stacking sites

	EF soil	EF atmosphere	EF hydrosphere
Best estimate	0.7	5.6×10^{-7}	0.3

Table S21 Detection of PFOA and PFOS in environmental media in China

Media	Location	Sampling time	PFOA	PFOS	Reference
Water (ng/L)	Daling River	2009	27.2-668 (169.0)	0.28-0.54 (0.4)	Bao et al. 2011
	Liaohe River	2009	n.d.[a]-27.9 (10.9)	n.d.-6.6 (0.3)	Yang et al. 2011
	Hanjiang River	2010	n.d.-256 (81.0)	n.d.-88.9 (51.8)	Wang et al. 2013
	Yangtze River	2003	0.2-297.5 (16.2)	0.2-37.8 (6.7)	Jin et al. 2009
	Huaihe River	2011	6.2-47 (18.0)	1.4-25 (4.7)	Yu et al. 2013
	Taihu Lake	2009	10.6-36.7 (21.7)	3.6-394 (26.5)	Yang et al. 2011
	Tangxun Lake	--[b]	70.5-1390 (372)	73.4-1650 (357)	Zhou et al. 2013
	East Lake	2009	15.8-158 (55.0)	9.1-132 (60.4)	Chen et al. 2012
Soil (ng/g)	Haihe Watershed	2008	n.d.-0.51 (0.2)	0.02-2.36 (0.2)	Pan et al. 2011
	Tianjin Binhai New Area	2008	n.d.-0.93 (0.2)	n.d.-9.4 (1.8)	Wang et al. 2012
	Coastal area of Bohai Sea	--	n.d.-13.3 (0.5)	n.d.-9.4 (0.1)	Meng et al. 2015

	Shanghai	2007	3.3-47.5 (35.3)	8.6-10.4 (9.5)	Li et al. 2010
Air (pg/m^3)	Shenzhen	2011	1.5-15 (5.4)	n.d.-4.3 (3.1)	Liu et al. 2015

Concentrations of PFOA and PFOS were presented with range of min. to max., and mean concentration was presented in brackets;

a: not detected;

b: not available.

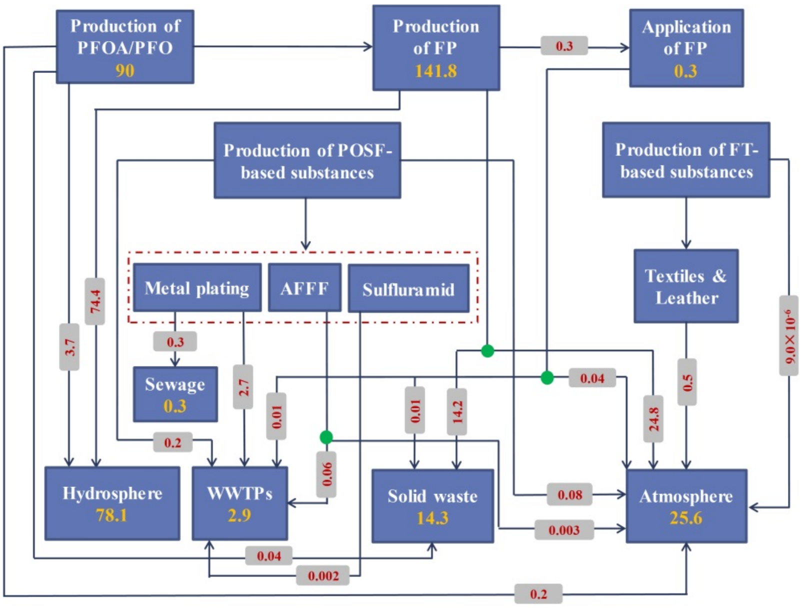

Fig. S1 Flows of PFOA/PFO (t) during production and use by industries

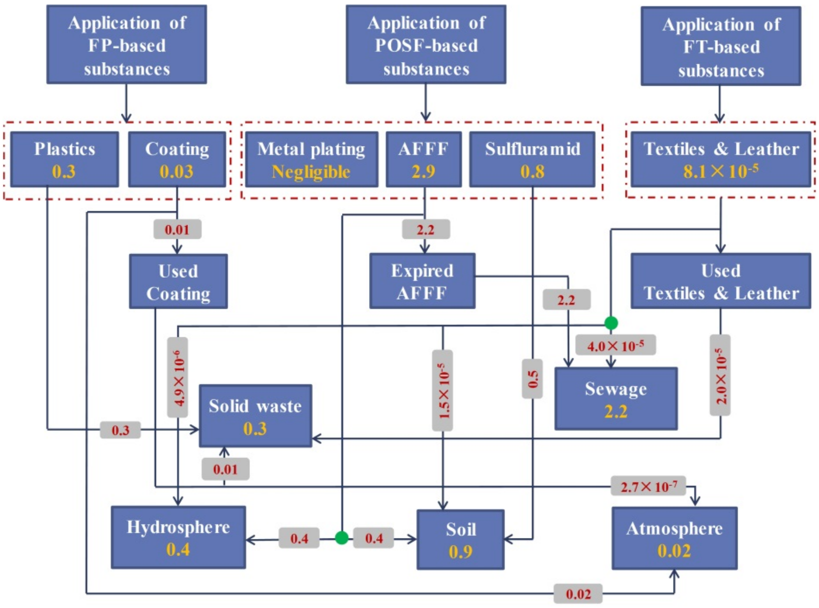

Fig. S2 Flows of PFOA/PFO (t) in domestic use

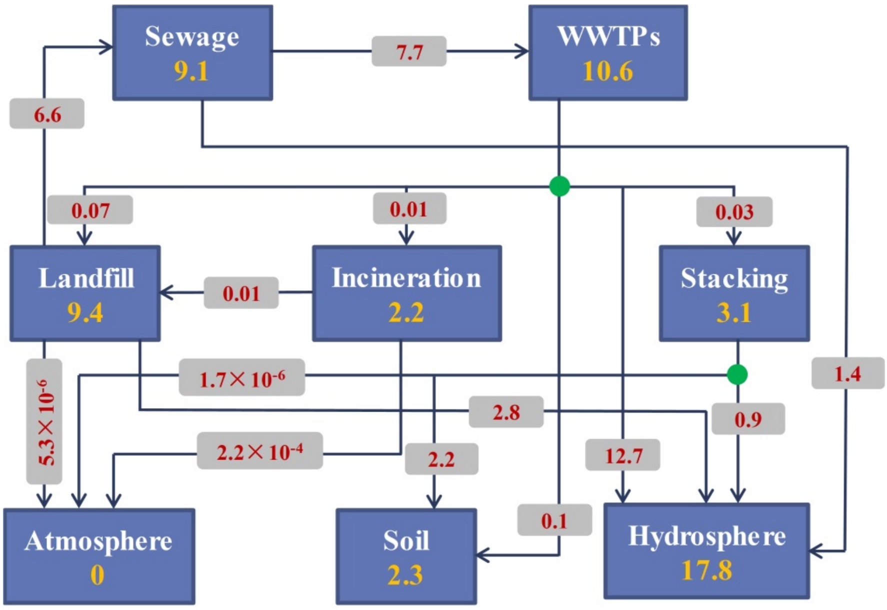

Fig. S3 Flows of PFOA/PFO (t) in waste management

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