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Article

Highly Resolved Life Cycle Emissions of Per- and Polyfluoroalkyl Substances (PFAS) in China from 1985 to 2023

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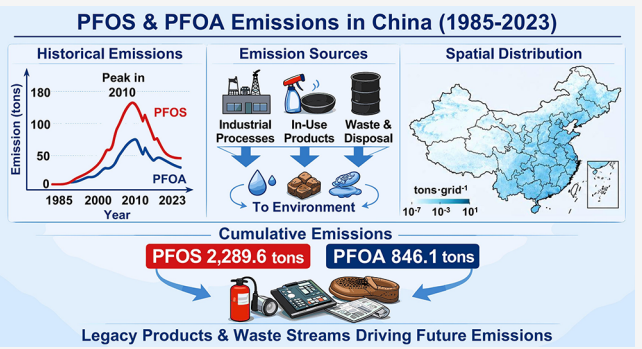
ABSTRACT: Per- and polyfluoroalkyl substances (PFAS), particularly perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), are persistent contaminants of global concern, yet their long-term and spatially resolved emissions in China remain poorly understood. Here we developed a life cycle emission inventory of PFOS and PFOA with high spatiotemporal resolution for mainland China from 1985 to 2023 using dynamic material flow analysis (DMFA). By integrating provincial activity data, trade statistics, and sector-specific emission factors, we quantified emissions from production, industrial applications, in-use products, and end-of-life management to multiple media. Cumulative emissions reached 2,289.6 tons for PFOS and 846.1 tons for PFOA. Early-2010s PFOS emissions were approximately double previous estimates, and our inventory filled gaps in emission hotspots that had been overlooked in earlier studies. Historical emissions exhibited an increase−peak−decline pattern, peaking around 2010 and then decreasing due to the Stockholm Convention and domestic regulations. High emissions were concentrated in eastern coastal and industrialized regions, with hotspots extending toward central and western China. With tightening controls on PFOS and PFOA, future emissions will increasingly be driven by legacy products and waste streams. This inventory provides a solid foundation for exposure assessment, hotspot identification, and PFAS emission management in China.



KEYWORDS: *PFAS, emission inventory, China, dynamic material flow analysis, source distribution*

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) have become emblematic "forever chemicals" in the global environment.[1,2] Perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), the two most extensively studied legacy compounds, are of particular concern because their fully fluorinated carbon chains confer exceptional resistance to environmental and biological degradation.[3] This resistance leads to long environmental half-lives, high mobility in aquatic systems, and bioaccumulation in food webs.[4,5] Accumulating toxicological and epidemiological evidence has linked PFOS and PFOA exposure to developmental toxicity, immune dysfunction, cardiometabolic effects, and cancer.[6,7] These concerns prompted the listing of PFOS and PFOA as persistent organic pollutants under the Stockholm Convention in 2009 and 2019, respectively,[8,9] as well as in China's List of New Pollutants for Priority Control (2023 Edition).[10]

China plays a vital role in the global PFOS and PFOA supply chain as both a major producer and consumer.[11,12] Domestic production of PFOS-, PFOA-, and precursor-based products began in the mid-1980s, expanded rapidly during the 2000s in response to phaseouts in other regions, and has declined in recent years under international and domestic controls.[13−15] PFOS and PFOA have been used in a wide range of industrial and consumer applications that exploit their surfactant and repellent properties, including metal-plating mist suppressants, aqueous film-forming foams, water- and oil-repellent textiles and leather, paper and packaging, surface coatings, and processing aids in fluoropolymer manufacturing.[16,17] Releases occur during production, downstream industrial processing, use of PFOS- and PFOA-containing products, and end-of-life management through wastewater discharges, air emissions, sludge, and solid-waste pathways.[18] Given the scale and diversity of these uses, PFOS and PFOA emissions are expected to be highly heterogeneous in both space and time, with hotspots governed by industrial clustering, product consumption patterns, and waste-management infrastructure.

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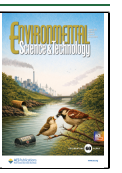

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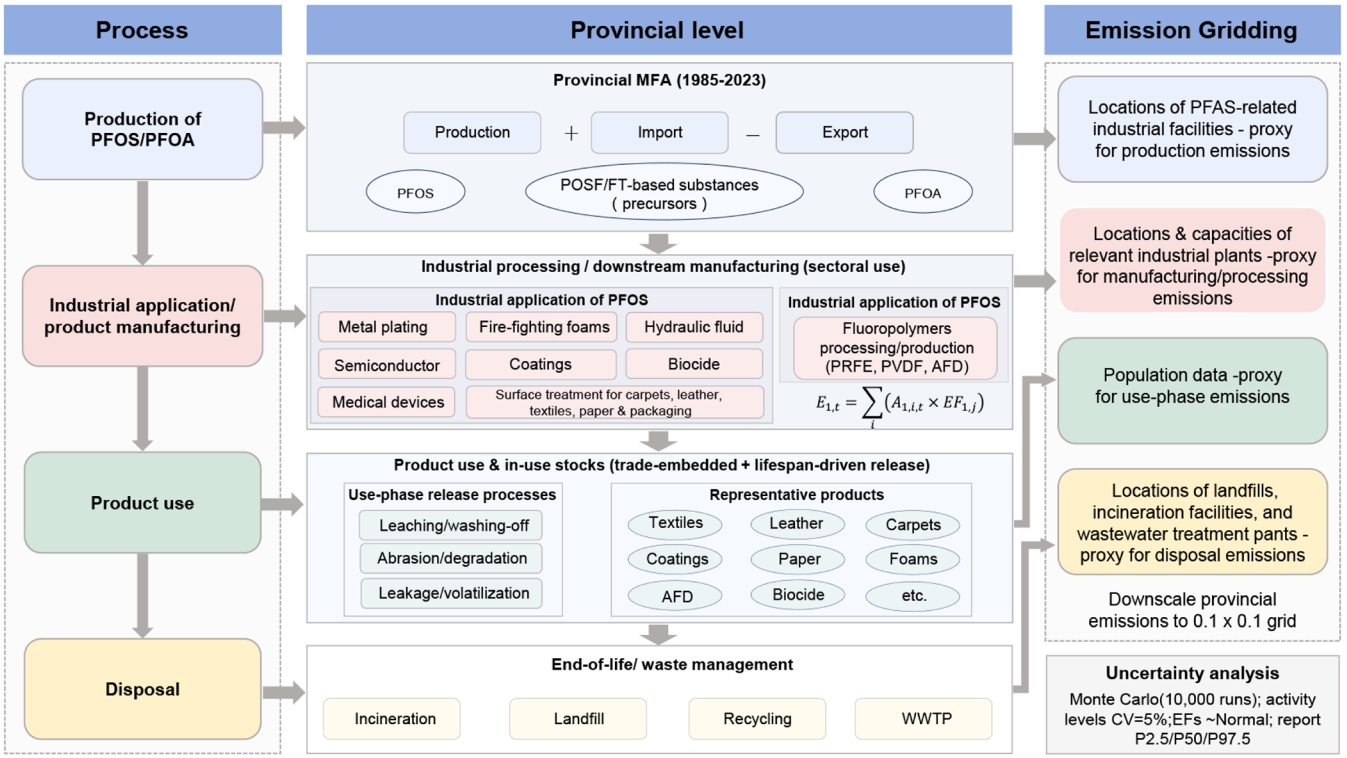

Figure 1. Flowchart showing the steps and procedures for establishing China's gridded PFOS and PFOA emission inventory.

An emission inventory that tracks releases across life cycle stages is essential for policymakers to implement effective measures to reduce PFAS concentrations in environmental media and to identify source−receptor relationships. While several studies have quantified PFOS and PFOA emissions at global and national scales,[11,15,18−20] providing valuable insights into production histories, dominant source sectors, and broad temporal trends, existing inventories exhibit four substantial limitations. First, most inventories are restricted to specific periods, often centered around 2010, or focus on selected sectors or regions,[15,19] and therefore fail to provide a continuous long-term emission trajectory that captures both industrial expansion and subsequent regulatory control.[15,18] Second, spatial resolution is generally coarse, with many inventories reporting only national totals or provincial estimates and lacking gridded data at resolutions compatible with atmospheric and hydrological models or with the spatial scales relevant to environmental monitoring and exposure assessment.[16,20] Third, emissions are often treated in aggregated form, without standardized differentiation by life cycle stage, application sector, or environmental compartment. This lack of harmonization constrains the identification of dominant emission pathways and limits the ability to anticipate future changes as production declines while legacy stocks and waste streams persist. Finally, most existing assessments do not explicitly integrate precursor emissions and their potential conversion to PFOS or PFOA, nor do they systematically examine how emission trajectories respond to evolving regulatory frameworks. A recent study on polytetrafluoroethylene (PTFE)-related nonpolymeric PFAS in China provided valuable emission estimates and noted a shift to emerging alternatives due to regulations,[20] but it only addressed PTFE-related PFAS and did not provide a detailed spatial emission inventory for environmental modeling and exposure assessment.

To address these knowledge gaps, this study developed a long-term (1985−2023), high-resolution (0.1° × 0.1°) emission inventory for PFOS and PFOA in mainland China by combining a dynamic material flow analysis (DMFA) framework[21−23] with provincial activity data, trade statistics, sector-specific emission factors, and spatial information on industrial and waste management infrastructure (Tables S1−S5). The inventory was used to assess how PFOS and PFOA emissions across life cycle stages and application sectors have responded to regulation under the Stockholm Convention and China's List of New Pollutants for Priority Control and to project future emissions from in-use stocks under current policy frameworks. This comprehensive inventory provides a life cycle-consistent and spatially explicit baseline for research on PFOS and PFOA emissions in China and supports environmental fate and exposure modeling, hotspot identification, and the development and evaluation of PFAS emission control strategies.

2. MATERIALS AND METHODS

2.1. Emission Framework

Figure 1 presents a flowchart illustrating the framework used to develop the long-term, high-resolution emission inventory of PFOS and PFOA in mainland China. First, we collected data on annual provincial production, imports and exports, and sectoral applications of PFOS and PFOA (Tables S1−S4) and tracked their accumulation and release through in-use stocks and end-of-life product flows within a DMFA framework. Second, we quantified multimedia emissions to air, water, and soil across key life cycle stages, including production of PFOS and PFOA, downstream industrial application, product use, and waste management, using stage- and sector-specific emission factors derived from the literature. Finally, provincial emissions were spatially allocated to grid cells using proxy data to establish a

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multimedia gridded emission inventory for PFOS and PFOA in China at a resolution of 0.1° × 0.1° from 1985 to 2023.

2.2. Emission Sources and Activity Data

In China, the large-scale production and use of PFOS, PFOA, and their related compounds began in the mid-1980s using the well-established electrochemical fluorination technology.[11,24] These substances have been widely used across multiple industries and are broadly distributed throughout product supply chains.[25,26] The historical use of PFOS and its related compounds in China has been documented across several applications, including fire-fighting foams, metal plating, and surface treatment agents for materials such as carpets, leather, textiles, upholstery, paper, and packaging.[11,19] They have also been extensively applied in coatings and coating additives, as well as in industrial and household cleaning products and biocides.[27] PFOA and its related compounds have been predominantly used as processing aids in the manufacture of fluoropolymers, such as PTFE, and as surfactants and surface treatment agents in textiles, paper, paints, and fire-fighting foams.[13,19]

We identified perfluorooctanesulfonamides (FOSAs) and perfluorooctanesulfonamidoethanols (FOSEs) as PFOS precursors, while PFOA precursors primarily include fluorotelomer alcohols (FTOHs), FOSAs, and FOSEs.[28,29] Some precursors, such as FTOHs, FOSAs, and FOSEs, are industrial products and may be released into the environment during manufacturing and use.[30] Both FOSAs and FOSEs are produced from perfluorooctanesulfonyl fluoride (POSF), whereas FTOHs are manufactured from fluorotelomers (FTs).[31,32] These precursors are also released as impurities or byproducts during the manufacture of other FT-based or POSF-based products, including PFOS and PFOA.[18,33] Emission data for these precursors were collected or calculated based on their production and use, corresponding emission factors, and their occurrence as impurities in different industrial processes and products.[29] However, the transformation of precursors to PFOS or PFOA was not explicitly quantified in the inventory. This treatment was adopted because precursor degradation pathways, conversion efficiencies, and transformation time scales are highly compound-specific and strongly dependent on environmental conditions.[34−36] These factors preclude a robust national-scale estimation of the precursor-to-PFOS/PFOA conversion. Accordingly, precursor results are reported as direct precursor emissions rather than as indirect PFOS or PFOA formation. Because the most recent data on the production and use of PFOS and PFOA are available for 2023 (Supporting Information, Section SI1), this study covers the period from 1985 to 2023 and is confined to mainland China.[26] Annual provincial production, consumption, and the proportions of use across major application industries from 1985 to 2023 were obtained from published literature and statistical sources (Figure S1).

2.3. Inventory Development

This study explores the evolution of PFOS and PFOA releases in China from production and use between 1985 and 2023 using a DMFA approach.[21,37,38] The analysis primarily focused on PFOS and PFOA emissions to air, water, and soil across different life cycle stages. The emission factors (EFs) used in this study (Tables S7−S10) were obtained from the literature,[13−15,18,25] the European Technical Guidance Document (https://publications.jrc.ec.europa.eu/repository/handle/JRC23785), and the OECD Emission Scenario Documents (https://www.oecd.org/en.html). These sources document industrial processes and emission release characteristics that are comparable to those employed in China, where PFOS and PFOA production has historically relied on electrochemical fluorination technology and fluoropolymer manufacturing pathways that follow internationally established methods. The EFs have been widely used and validated in previous PFAS emission inventories covering similar industrial sectors and time periods in China.[11,13,15]

The stock flow relationship was explicitly represented for product-use and end-of-life stages. Specifically, based on sector-specific lifetime distributions, PFOS and PFOA entering product applications in year t*t* were either retained in in-use stocks or transferred to end-of-life flows in subsequent years.[21,39,40] The annual discarded amount was therefore derived from historical product inputs and lifetime functions, rather than treated as an independent activity variable. This mass was then distributed among recycling, incineration, and landfill pathways using time-dependent waste-management shares.[21] It should be noted that several processes remain outside this boundary or are treated only in simplified form, including secondary emissions from recycled or reused materials,[41] diffuse nonpoint releases (e.g., from washing, abrasion, and urban runoff), and product-specific heterogeneity in the PFOS/PFOA content of internationally traded goods.[42,43]

(1) Emissions from the PFOS/PFOA production[21,37]

$$E_{1,j,t} = \sum_{i} (A_{1,i,t} \times EF_{1,j}) \tag{1}$$

where *i*, *j*, and *t* denote province, emission media (air, water, and soil), and year, respectively; $E_{1,j,t}$ denotes the amount of emission from PFOS/PFOA production to medium *j* in year *t*; A_1 denotes the annual production volumes (tons) of PFOS/PFOA in province *i* in year *t* (Section SI1, Tables S1 and S2); and $EF_{1,j}$ denotes the medium-specific emission factor of the PFOS/PFOA production process.

(2) Emissions from downstream product manufacturing[38,44]

$$E_{2,k,j,t} = \sum_{i} (A_{2,i,k,t} \times EF_{2,k,j}) \tag{2}$$

where *k* denotes the specific main product industry using PFOS/PFOA in China; $E_{2,k,j,t}$ denotes emissions of sector *k* to medium *j* in year *t*; $A_{2,i,k,t}$ denotes the amount of PFOS/PFOA used in sector *k* from province *i* in year *t* (Section SI1 and Figure S1); and $EF_{2,k,j}$ denotes the sector- and medium-specific emission factor of the downstream product manufacturing processes.

(3) Emissions during the use of products[28,45]

Some PFOS and PFOA materials were exported to international markets, while most of the remaining materials were further processed into various products for domestic use (Tables S1 and S2). Both pathways contribute to PFOS and PFOA emissions during the product use phase. We assumed that imported and exported products of the same type contain equivalent amounts of PFOS and PFOA, given the similarity in primary PFOS and PFOA application industries in China and other countries.[46] Using import and export data for PFOS- and PFOA-containing products obtained from the National Bureau of Statistics (https://www.stats.gov.cn/), the UN Comtrade Database (https://comtradeplus.un.org/), and relevant literature (Table S11), we analyzed the mass flows of PFOS and PFOA associated with international trade. This information was then incorporated into the product use phase to estimate subsequent emissions.[21,38]

$$E_{3,k,j,t} = \sum_{i} \left[(A_{2,i,k,t} - E_{2,i,k,t}) \times \left(1 + \frac{I_{i,k,t} - E_{i,k,t}}{P_{i,k,t}}\right) \times [1 - \varphi_k] \times EF_{3,k,j} \right] \tag{3}$$

where $E_{3,k,j,t}$ denotes emissions from the use of PFOS/PFOA-containing product *k* to medium *j* in year *t*; $I_{i,k,t}$ denotes the product (tons) annual import quantity or import value of product *k* from province *i* in year *t*; $E_{i,k,t}$ denotes the product (tons) annual export quantity or export value of product *k* from province *i* in year *t*; $P_{i,k,t}$ denotes the product (tons) annual domestic production of product *k* from province *i* in year *t*; φ_k denotes the cumulative distribution function of PFOS/PFOA life span in products (Table S12); and $EF_{3,k,j}$ denotes the emission factor during the use of respective products.

(4) Emissions from the disposal of end-of-life products[15,21]

Preliminary estimations of emissions from the disposal of end-of-life products were conducted by integrating the relative proportions of waste disposal methods in China (i.e., incineration, recycling, and landfill).[21] The total amount of PFOS/PFOA entering end-of-life products in a year *t* can be expressed as:

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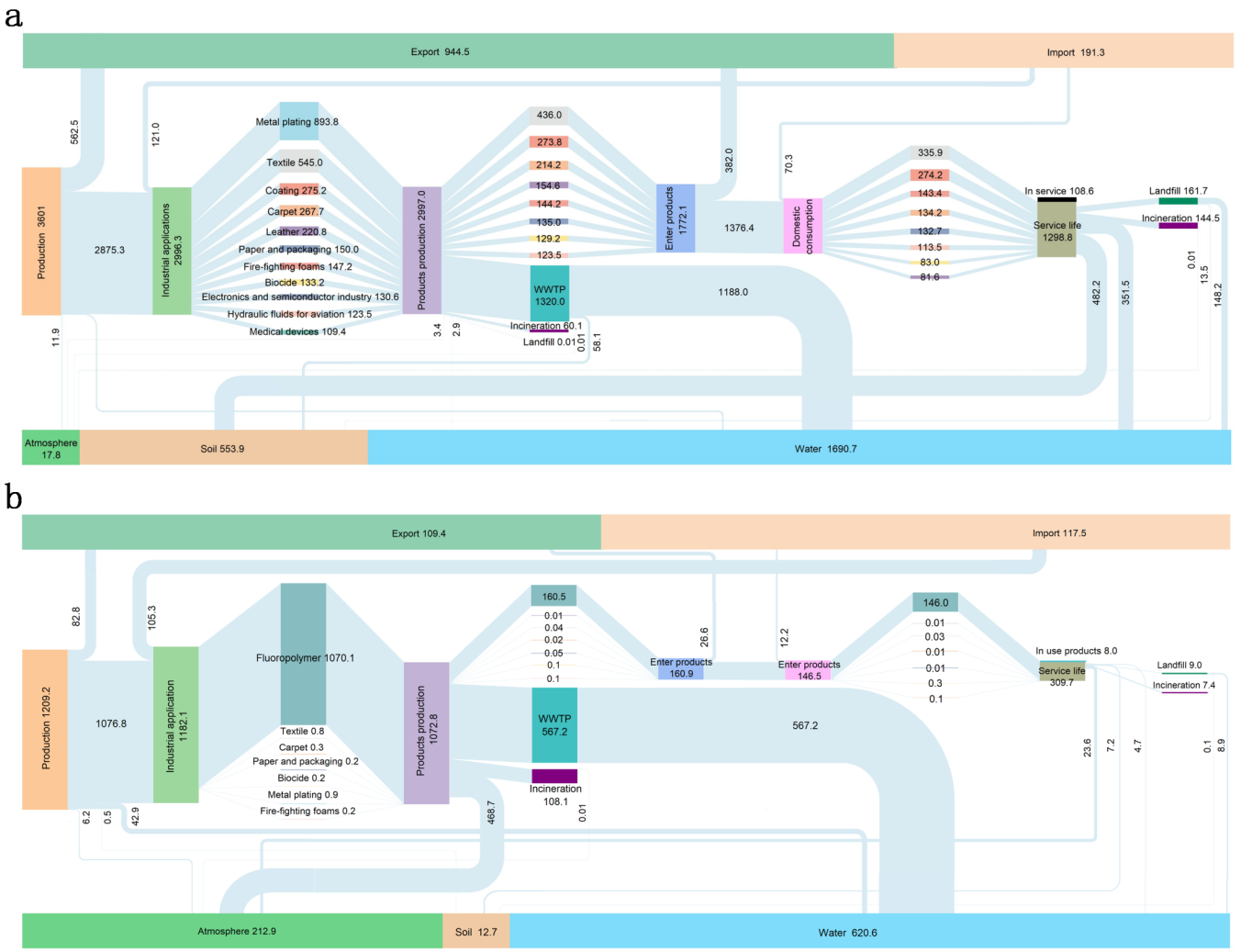

Figure 2. Flows of PFOS (a) and PFOA (b) in China from 1985 to 2023, with the flow width indicating the scales of mass.

$$DIS_{4,d,k,t} = \sum_{i} (A_{2,i,k,t} - E_{2,i,k,t} - E_{3,i,k,t}) \times f_{k,t} \times \alpha_{d,t} \quad (4)$$

where $DIS_{4,d,k,t}$ denotes the total amount of PFOS/PFOA discarded through waste-management pathway d for product k in year t; $E_{2,i,k,t}$ and $E_{3,i,k,t}$ denote the total releases during downstream product manufacturing and product use across all environmental media, respectively; $f_{k,t}$ denotes the life span distribution function of PFOS/PFOA in the product k (Section SI2), and $\alpha_{d,t}$ denotes the shares (%) of recycling, incineration, and landfilling treatments in year t (Table S13).

2.4. Emission Gridding

We spatially interpolated provincial PFOS and PFOA emission data into grid cells at a spatial resolution of 0.1° × 0.1° latitude and longitude from 1985 to 2023.[47,48] Emission distributions from upstream PFOS and PFOA production were determined based on the known locations of PFAS-related industrial facilities (Figure S2). For emissions associated with product manufacturing, the locations and capacities of industrial plants involved in the production of fire-fighting foams, metal plating, carpets, leather, textiles, paper and packaging, coatings, biocides, and fluoropolymers were used as spatial proxies (https://www.qcc.com/). The locations of landfills, incineration facilities, and wastewater treatment plants were employed to represent emissions during the disposal phase (https://www.cnopendata.com/). Owing to limited information, emissions during the use phase were allocated using population-based proxies (https://www.resdc.cn/data.aspx?DATAID=260).

2.5. Uncertainty Analysis

We employed Monte Carlo simulation to assess the uncertainty of the emission inventory based on probabilistic distributions of key parameters.[22,23,26] These parameters included activity levels, multimedia PFOS and PFOA EFs, and the shape and scale parameters of the Weibull lifetime distribution functions that govern product-specific service lifetimes (Table S12). Activity levels follow normal distributions with coefficients of variation of 10%.[49,50] The PFOS and PFOA EFs were assumed to follow normal distributions,[11] and the Weibull lifetime parameters were also treated as random variables with distributions derived from the literature.[51] The simulations were run 10,000 times using Python. By treating the lifetime distribution parameters probabilistically, lifetime-related uncertainty was propagated into the emission estimates, ensuring that variability in in-use stock accumulation and end-of-life product flows is appropriately reflected in the final inventory and its 95% confidence intervals. Median values were selected as the best estimates, and (P97.5−P50)/P50 and (P50−P2.5)/P50 were used to represent the upper and lower uncertainty bounds of the simulation results, respectively.[20,23,52] Here, P97.5, P50, and P2.5 denote the 97.5th, 50th, and 2.5th percentiles of the probability distributions for PFOS and PFOA inputs and multimedia emissions (Figure S3). Detailed information is provided in Section SI3.

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3. RESULTS AND DISCUSSION

3.1. Production and Use of PFOS/PFOA in China

Tables S1 and S2 present the temporal trends in annual production and consumption of PFOS and PFOA in China from 1985 to 2023. China's production of PFOS and PFOA was negligible during the 1980s and 1990s but increased sharply during the 2000s, peaked around 2010, and then declined in response to regulatory controls. China was reported to be the sole country still producing POSF, a PFOS precursor, and its output surged following 3M's phaseout of PFOS in 2002.[53] By contrast, after PFOS was listed under the Stockholm Convention in 2009, China began to curtail its production and use. From 1985 to 2023, the cumulative production of PFOS and PFOA reached 3,601.0 tons and 1,209.2 tons, respectively. Import and export activities for these substances were relatively limited over the last four decades, with total imports and exports of 121.0 tons and 562.5 tons for PFOS and 82.8 tons and 105.2 tons for PFOA, respectively.

Between 1985 and 2023, approximately 2,996.3 tons of PFOS and 1,182.1 tons of PFOA were used in industrial applications (Tables S1 and S2). Most PFOS and PFOA produced in China were consumed domestically in specific industries (Tables S3 and S4). PFOS was used primarily in metal-plating mist suppressants (29.8%), textile treatments (18.2%), and surface coating processes (9.2%) (Table S3).[11,25] Over the study period, the proportions of PFOS used in metal plating, textiles, and coatings exhibited an upward trend (Figure S4). In other industries, such as biocides, fire-fighting foams, and paper and packaging, the absolute volume of PFOS use decreased, whereas the relative proportion of use remained comparatively stable in recent years, as illustrated by the annual sectoral-use data (Figure S4 and Table S3). By contrast, PFOA was used almost exclusively as a processing aid in fluoropolymer production.[20] Emulsifiers derived from PFOA are used in the manufacture and processing of PTFE, polyvinylidene fluoride, and related resin materials (Table S4). Over the past four decades, PFOA use has undergone a marked transition from widespread application to more limited use, reflecting increasing concerns regarding its safety.[54,55]

A substantial proportion of PFOS and PFOA produced since the 1980s has been incorporated into consumer and industrial products. By 2023, it is estimated that approximately 60% of cumulative PFOS applications have been transferred into products such as textiles, coatings, carpets, leather, foams, and paper. However, a large share (92.3%) of the PFOS stock has already been discarded or emitted during use, leaving only a small fraction (7.7%) remaining in active use (Figure 2a). By contrast, the in-use stock of PFOA is considerably smaller because it is primarily used as a processing aid in fluoropolymer manufacturing rather than as an integral component of consumer products (Figure 2b),[13,56] resulting in only a minor fraction remaining in finished goods. From 1985 to 2023, 382.0 tons of PFOS and 26.6 tons of PFOA were exported, while 70.3 tons of PFOS and 12.2 tons of PFOA were imported. Following product use, approximately 161.8 tons of PFOS and 9.0 tons of PFOA were discarded with end-of-life products over time. The quantity of waste generated is largely determined by PFOS and PFOA consumption levels and the service lifetimes of end-use products over the same period.

3.2. Emissions of PFOS and PFOA into the Environment

The total PFOS and PFOA emissions in China from 1985 to 2023 were estimated at 2,289.6 tons (95% CI: 308.7−8,360.3) and 846.1 tons (95% CI: 115.0−3,075.9), respectively (Figure 2). From a life cycle perspective, most PFOS and PFOA releases in China originated from product manufacturing and use of products stages. Our estimates indicate that product manufacturing was the dominant emission stage, accounting for approximately 54.7% of total PFOS emissions and 88.8% of total PFOA emissions. Within industrial processing, metal plating accounted for 67.4% of PFOS emissions from industrial applications, followed by medical devices, textiles, electronics and semiconductor industries, leather, and carpet treatments (Table S3). By contrast, fluoropolymer processing accounted for over 99% of PFOA emissions from industrial applications between 1985 and 2023, based on the cumulative share of fluoropolymer sectors in total industrial emissions (Figure 2b). This proportion is comparable to the 98.3% reported by Li et al.[15] for 2004−2012. In-use products represented the second largest emission source, contributing 36.4% of PFOS emissions and 4.2% of PFOA emissions. Specifically, coatings, fire-fighting foams, biocides, and textiles contributed 46.7%, 24.6%, 9.67%, and 8.92%, respectively, to total PFOS emissions from in-use products. The use of aqueous fluoropolymer dispersions accounted for 84.3% of PFOA emissions from in-use products, while the remaining 10.5% and 5.2% were attributed to FT-based and POSF-based substances, respectively. The PFOS production and end-of-life stages contributed 1.8% and 7.1% to total PFOS emissions and 5.9% and 1.1% to total PFOA emissions, respectively. Within the end-of-life stage, landfilling and incineration were the two main subsources of PFOS and PFOA emissions.

The distribution of emissions among environmental media also differed between the two substances. According to our analysis, most cumulative PFOS emissions entered water (75.0%), while emissions to soil and the atmosphere were comparatively smaller, accounting for 24.2% and 0.8% of total PFOS emissions, respectively. Across the life cycle, releases to water occurred during all stages except incineration, with 69.3% of PFOS emissions to water arising from the industrial application stage. Our estimates of the multimedia distribution of PFOS emissions are comparable with those reported by Xie et al.[11] (87.7% for water, 9.6% for soil, and 2.7% for air). The present study further provides quantitative and dynamic source-resolved distributions of PFOS emissions. For PFOA, the majority of cumulative emissions were released directly into water (73.3%), followed by emissions to air (25.2%) and soil (1.5%) (Figure 2b). Notably, the proportion of emissions entering water was almost identical between PFOS and PFOA, indicating that water serves as the dominant reservoir for both compounds. By contrast, atmospheric PFOS emissions constituted only 0.8% of its total releases, which is substantially lower than the 25.2% for PFOA. This difference is likely attributable to the lower volatility of PFOS and its higher octanol−air partition coefficient (log K_{OA} = 4.84−8.07) compared with PFOA (log K_{OA} = 4.24−7.23).[57] The result indicates a stronger tendency of PFOS to partition into environmental organic phases. Laboratory studies have shown that the organic carbon-normalized partition coefficient (log K_{OC}) for PFOS (3.0−3.7) is significantly higher than that for PFOA (2.4−2.8).[58,59]

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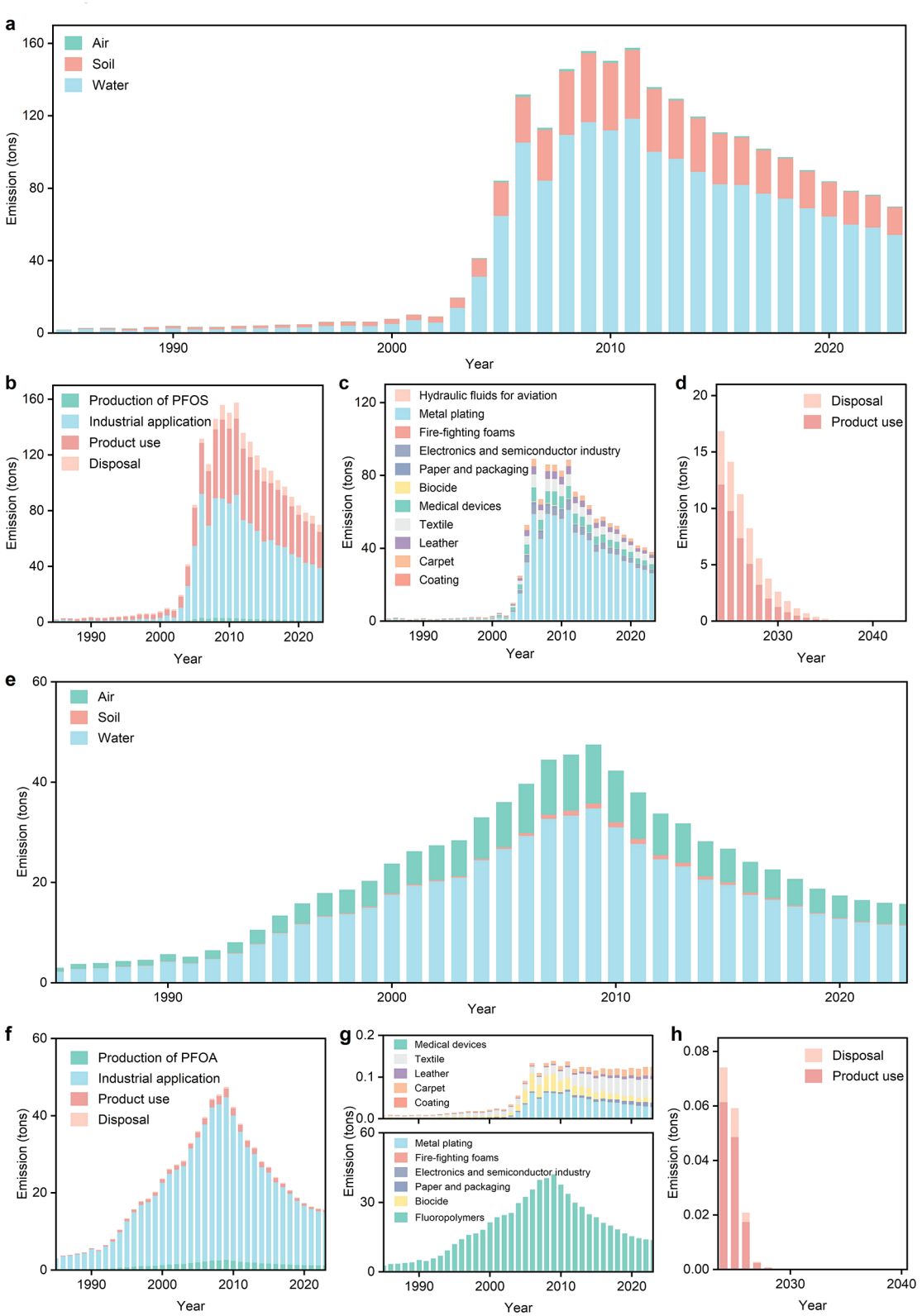

Figure 3. Temporal evolution of annual PFOS and PFOA total emissions and their decomposition (1985−2050): (a−d) PFOS and (e−h) PFOA. (a and e) Emissions to air, water, and soil; (b and f) emissions decomposed by life cycle stage (production, industrial application, use, and disposal); (c and g) emissions decomposed by major use sectors; and (d and h) projected use-phase emissions for 2024−2050 under the scenario described in Section 2.

3.3. Temporal Trends of Annual PFOS and PFOA Emissions

Historical emission trends of PFOS and PFOA in China are shown in Figure 3 across different life cycle stages and sectors. The temporal patterns of PFOS and PFOA emissions were closely correlated with PFAS production and use (Tables S1−S3). Total PFOS emissions in China increased from 1.8 tons

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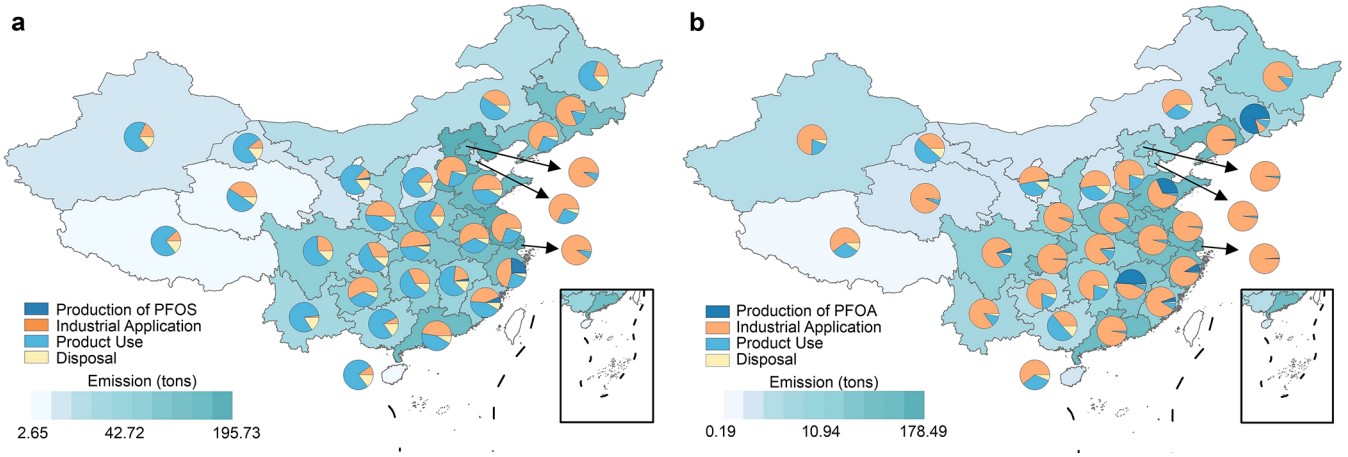

Figure 4. Provincial distribution of PFOS and PFOA emissions. (a) PFOS and (b) PFOA emissions by province in China for 1985−2023.

(95% CI: 0.24−6.6) in 1985 to a peak of 155.9 tons (95% CI: 21.0−568.9) in 2009, followed by a decline to 69.9 tons (95% CI: 9.4−255.1) in 2023 (Figure 3a). Between 1985 and 2000, total PFOS emissions increased by a factor of 3.3, with emissions from in-use products contributing 37.4% of the increase, accounting for approximately 53.6% of the total growth. During 2000−2009, total PFOS emissions surged by more than 18-fold, primarily driven by industrial application sectors, which accounted for more than 56% of the overall increase (Figure 3b). Over the same period, atmospheric PFOS emissions increased from 0.1 to 1.3 tons, PFOS entering waste streams rose from 2.7 to 38.0 tons, and aquatic PFOS releases increased from 5.0 to 116.5 tons. The growth in PFOS emissions to soil and aquatic media was largely attributable to in-use products and industrial application sectors, respectively, which together accounted for approximately 95.0% of the total increase in these categories. By contrast, the increase in atmospheric emissions was predominantly driven by PFOS production, contributing about 80% of the total rise. After 2010, PFOS emissions declined at an average rate of 8 tons per year, primarily as a result of PFOS being listed under the Stockholm Convention in 2009, which led China to restrict its production and use.

Figure 3e shows that PFOA emissions in China increased rapidly from 3.0 tons (95% CI: 0.4−10.9) in 1985 to 47.5 tons (95% CI: 6.5−172.7) in 2009, followed by a subsequent decline. Between 1985 and 2019, atmospheric PFOA emissions increased from 0.8 to 11.7 tons, soil emissions rose from 0.02 to 1.0 tons, and aquatic discharges increased from 2.2 to 34.8 tons. This growth in multimedia PFOA emissions was primarily driven by the production and use of fluoropolymers. For example, China's production of PTFE, a key fluoropolymer, increased by more than an order of magnitude, from 1,012.5 tons in 1985 to 52,098.2 tons in 2010 (Table S4). Despite continued expansion of PTFE production and the use of aqueous fluoropolymer dispersions, the reduced application rate of PFOA in fluoropolymer manufacturing has contributed substantially to emission reductions. Between 2010 and 2023, annual atmospheric PFOA emissions decreased by 59.8%, water emissions declined by 63.1%, and soil emissions were reduced by 85.4%. As noted in previous studies, this decline can be attributed to strengthened control measures for persistent organic pollutants.

The production and use of PFOS and PFOA were banned in China at the end of 2023 under the Priority Control List of New Pollutants; however, emissions from in-use stocks persist. As a result, the dominant emission sources are expected to shift from industrial applications to in-use stocks in the future (Figure 3d and h). Long-lived consumer products, such as textiles, leather, and carpets, that contain PFOS or PFOA will continue to release these substances as they age or enter waste streams over the coming decade. By contrast, short-lived products, including paper, pesticides, and foams, are expected to be largely phased out by approximately 2030. Overall, most in-use stocks are projected to decline substantially, with a large fraction depleted by 2030, while residual emissions from durable goods are expected to persist into the 2040s and beyond as materials age and degrade. In addition to direct releases from in-use stocks, indirect formation from precursors, such as FTOHs, FOSAs, and FOSEs (Table S6), as well as the slow degradation of fluorinated materials in products and waste streams, may sustain PFOS and PFOA burdens over longer time scales. However, precursor-to-PFOS and PFOA transformation was not included in the projections because degradation rate constants remain highly uncertain.

3.4. Spatial Distribution

The emissions of PFOS and PFOA across different provinces in China are illustrated in Figure 4, revealing distinct provincial patterns. PFOS emissions were generally higher in eastern coastal regions than in western regions, with major concentrations in the Jiangsu, Hebei, Shanghai, Beijing, Shandong, Guangdong, and Zhejiang provinces (Figure 4a). Zhejiang is the primary producer of PFOS, accounting for approximately 70% of the national production capacity. Shanghai, Hebei, Beijing, Jiangsu, and Jilin provinces also have substantial production of metal-plating products, with cumulative production shares of 16.2%, 14.3%, 11.6%, 10.7%, and 9.9%, respectively, from 1985 to 2023 (Figure S1). In-use products containing PFOS in these provinces are also significant, accounting for 1.7%, 5.3%, 1.5%, 5.9%, and 2.0% of the cumulative PFOS usage over the same period, respectively. Consequently, PFOS emissions are higher in these regions. By contrast, PFOA emissions in China primarily originate from production and manufacturing processes, with Shanghai, Jiangsu, Shandong, Guangdong, and Liaoning identified as the largest contributors, accounting for 18.8%, 11.4%, 10.2%, 9.4%, and 9.2% of the total emissions,

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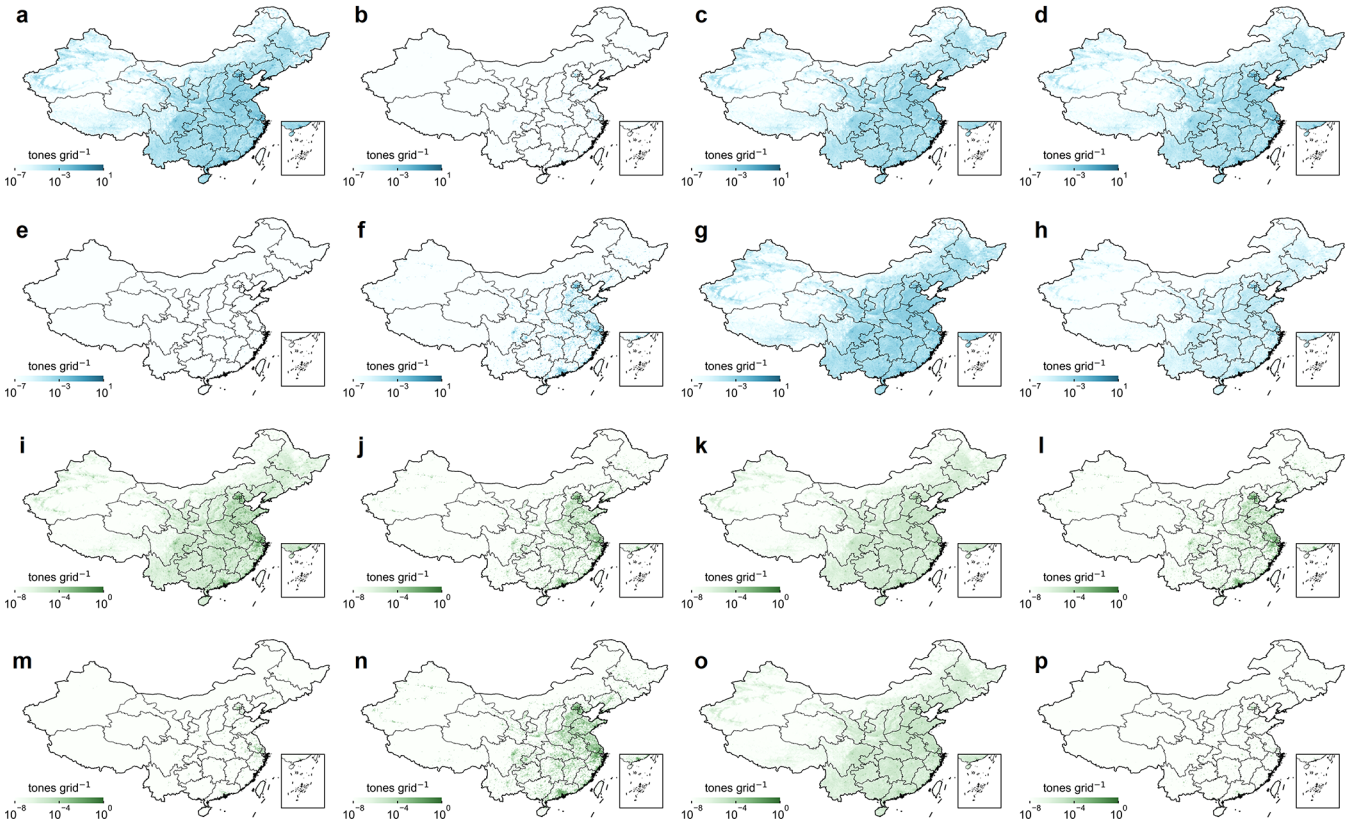

Figure 5. Annually averaged gridded PFOS and PFOA emissions across China from 1985 to 2023 at 0.1° × 0.1° resolution. Panels (a−h) show PFOS, and panels (i−p) show PFOA; within each group, maps correspond to total, air, soil, water, production, industrial, use, and disposal.

respectively (Figure 4b). This spatial pattern reflects the distribution of PFOA-related industrial activities across the country.

Figure 5 presents the spatial distribution of gridded PFOS and PFOA emissions at a resolution of 0.1° × 0.1°. Emissions were first calculated at the provincial level and then allocated to individual grid cells using various types of spatial information (see Section 2.4). Considerably high PFOS and PFOA emissions were observed in relatively developed regions, including the Beijing−Tianjin−Hebei region, the Pearl River Delta, the eastern coastal areas of Shandong, Jiangsu, and Zhejiang, as well as the central regions of Henan and eastern Sichuan−Chongqing. These regions are characterized by large populations and developed economies, resulting in substantial demand for PFOS and PFOA applications and corresponding emissions. The spatial patterns of gridded PFOS and PFOA emissions differed slightly among sectors, as shown in Figures 5e−5h and 5m−5p. Emissions associated with the use phase (Figures 5g and 5o) and disposal phase (Figures 5h and 5p) were more spatially dispersed, whereas emissions from application sectors were more spatially concentrated (Figures 5f and 5n). To evaluate the spatial plausibility of the inventory, we compiled literature-reported PFOS and PFOA concentration data in air, soil, and surface water across China (Supporting Information Data 1). As the temporal coverage of these observations is too sparse to support a year-by-year trend correlation, we conducted a spatial pattern comparison between the observed concentration distributions and our gridded emission inventory. The observed concentration hotspots were broadly consistent with the high-emission regions identified by the inventory, mainly in eastern China. The results support the reliability of the estimated spatial distribution (Figures 5 and S5).

We further examined temporal changes in the spatial distribution of multimedia PFOS and PFOA emissions. Figure S6 illustrates emission patterns at 5-year intervals. Between 1985 and 1990, PFOS and PFOA production and use were at an early stage, with average annual emissions of 1.9 tons and 6.5 tons, respectively, largely confined to a limited number of provinces. PFOS emissions were mainly concentrated in Jiangsu, Beijing, and Guangdong, whereas PFOA emissions were primarily observed in Jilin, Guangdong, and Liaoning. From 1990 to 2000, the average annual emissions of PFOS and PFOA increased to 8.2 tons and 27.2 tons, respectively, reflecting an expansion of high-emission areas. During this period, Zhejiang emerged as the dominant PFOS emitter, while PFOA use increased notably in Guangdong, Liaoning, and Zhejiang. From 2000 to 2010, PFOS and PFOA emissions continued to rise, largely driven by the relocation of substantial production capacities from developed countries to China. At the same time, emission hotspots expanded geographically from coastal regions to inland areas. However, the listing of PFOS and PFOA under the Stockholm Convention, together with strengthened control measures in China, effectively reversed this trend. From 2010 to 2023, substantial reductions in PFOS and PFOA emissions were observed nationwide, particularly in traditional hotspots such as Zhejiang and Shandong. Figures S7 and S8 show five-year changes in provincial PFOS and PFOA emissions. These results reveal a clear temporal shift: from 1985 to 2010, most provinces exhibited predominantly positive changes, mainly due to industrial expansion associated with new facility construction

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cy7Dm3ZL_9Cf3qfKAc485ysgAAALcwgbQGCSqGSIb3DQEHBqCBpjCBowIBADCBnQYJKoZIhvcNAQcBMB4GCWCGSAFlAwQBLjARBAyDgnSSXuztk5SGx98CARCAcP_zG5ENdKw_hQo0y5h3M1C9FUOOQ1NbvxbtCMWx9MvM44b2PStOJYIHLoEWjIxRAE0wwIhqOUyq-yK1yMF88YEoDkNWP20yyv

and capacity scaling at existing plants. By contrast, from 2010 to 2023, emission trends reversed, with an increasing number of provinces showing negative changes.

3.5. Comparisons with Other Studies

As shown in Tables S14 and S15, our estimated PFOS emissions were 150.45 tons in 2010 and 135.89 tons in 2012, which are substantially higher than the corresponding estimates of 69.55 tons and 68.90 tons reported by Xie et al.[11] and Liu et al.[14] These differences are primarily attributable to methodological advances in the present study, including the consideration of additional emission sources that were omitted in earlier assessments, such as paper manufacturing, medical devices, and biocides, as well as the inclusion of in-use and end-of-life emissions through a DMFA framework. Our results are partially consistent with the temporal trends in PFOS emissions reported by Paul et al.[60] and Miao et al.[61] (Figure S9), although notable discrepancies remain in the absolute emission estimates. PFOS emissions during 1985−2002 in this study were approximately five times higher than those reported by Paul et al.,[60] whereas for 2004−2019, our estimates were 23.0% lower than those of Miao et al.[61] These differences can largely be explained by variations in data sources and the emission categories considered. For PFOA, our estimated cumulative emissions of 374.7 tons for the period from 2004 to 2012 exceeded the 250.8 tons reported by Li et al.,[15] mainly due to updated emission factor estimates for fluoropolymer processing and production, as well as for POSF- and FT-based substance production and use (Tables S8−S10). Du et al.[13] estimated that PFOA emissions in China may have reached 39.2 tons in 2019, which is approximately twice the level estimated in the present study for the same year. In that study, emissions were derived based on changes in source activities relative to 2012 and direct emission estimates for 2012 reported by Li et al.[15]

The gridded emissions in this study were also compared with POPE (Figure S10),[62] a global data product that integrates multiple PFAS inventories and allocates emissions using population density and gross domestic product as spatial proxies.[18,31,33,63] In terms of emission magnitudes, scatter plots reveal distinct patterns for PFOS and PFOA at a 0.5° resolution. The correlation coefficients between our inventory and POPE were 0.44 ($p < 0.001$) for PFOS and 0.13 ($p < 0.001$) for PFOA (Figures S10c and S10f). We further calculated absolute differences (AD), defined as $AD = E_{our} - E_{POPE}$, where E_{our} and E_{POPE} (>0) represent grid-level emissions from this study and POPE, respectively. The spatial distribution of positive and negative mean AD values averaged over mainland China highlights regions where PFOS and PFOA emissions were under- or overestimated relative to POPE. Specifically, POPE tends to underestimate PFOS emissions, particularly in urban areas (Figure S10g), while it generally overestimates PFOA emissions but underestimates emissions in urban regions compared with our inventory (Figure S10h). POPE estimates are largely based on historical extrapolations before 2015, predating the inclusion of PFOA under the Stockholm Convention in 2019 and China's 2023 priority pollutant framework.[20,64] By contrast, our inventory incorporates more recent activity data that reflect production declines and phase-out efforts. Consequently, POPE's pre-Stockholm Convention baseline for PFOA likely overestimates current emissions by not accounting for regulatory actions implemented after 2019, whereas our estimates better represent contemporary emission conditions. For PFOS, POPE's pre-2015 data partially overlap with the post-2009 implementation of the Stockholm Convention, which likely explains the stronger correlation relative to PFOA. From a spatial perspective, the population- and gross domestic product-based allocation approach used by POPE produces a relatively homogenized distribution of emissions across urban areas,[65] which inadequately captures industrial concentrations that drive PFAS emission hotspots. By contrast, our facility-based methodology, as described in Section 2.4, employs detailed industrial location data, including production facilities, processing sites, and waste management infrastructure, as spatial proxies. This approach more accurately identifies emission hotspots and resolves pronounced spatial gradients beyond major industrial clusters, which is critical for exposure assessment and policy development.

4. IMPLICATIONS

This high spatiotemporal resolution emission inventory of PFOS and PFOA provides a quantitative foundation for chemical management and regulatory evaluation in China. The emission trajectories exhibit a clear increase−peak−decline pattern that aligns with the implementation of international controls under the Stockholm Convention and subsequent national measures in China, demonstrating that coordinated regulatory interventions can substantially reduce primary emissions of these toxic chemicals.[9,20] Moreover, industrial restructuring, technology upgrades in manufacturing processes, shifts in international market demand, and relocation of production capacity may have also influenced these trends.[66] These findings underscore the importance of continued compliance monitoring and the timely integration of international obligations into domestic chemical control frameworks.[10] PFOS and PFOA emissions were concentrated in eastern China and other industrialized provinces, identifying these regions as priorities for enforcement, targeted environmental monitoring, and site-specific risk management.[67] The observed westward shift in emission hotspots further indicates that industrial restructuring and relocation can redistribute PFAS burdens, highlighting the need for proactive regulation in emerging industrial regions to prevent pollution transfer.

With PFOS and PFOA now included in China's List of New Pollutants for Priority Control,[10] the increasing contribution of in-use products and end-of-life waste reflects a shift in dominant emission drivers. Future emission reductions are therefore likely to depend less on production controls and more on life cycle management measures, including product registration, extended producer responsibility, waste treatment standards, and controls on secondary material flows such as recycling and landfill leachate.[68] Regulatory attention should extend beyond manufacturing to downstream use and disposal stages, where emissions may persist for decades. From a regulatory science perspective, this inventory provides valuable input for multimedia fate modeling, exposure assessment, and cumulative risk evaluation, supporting the development of regionally differentiated control strategies.

Several limitations of this study should be acknowledged. First, the emission inventory focuses on only two long-chain PFAS; future research should incorporate additional emerging PFAS.[67,69] Second, uncertainties in EFs contribute to uncertainty in the estimated PFOS and PFOA emissions, and additional field measurements, particularly those capturing sector-specific application processes and waste management

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practices, are needed to better characterize releases. Since time-varying EFs are unavailable for most processes relevant to PFOS and PFOA emissions, consistent with earlier PFOS/PFOA inventories,[11,13−15,18,25] we used the emission factors retrieved from the literature for the years corresponding to each process as fixed-year values. This simplification could introduce biases in absolute emissions and in process contributions, for example, overestimating recent emissions when controls, recovery, or waste management have improved, and underestimating earlier emissions. Third, although the DMFA ensured consistency in material-flow accounting and emission estimation across life-cycle stages, the gridded inventory was generated by downscaling provincial emissions using stage-specific spatial proxies rather than tracking actual interprovincial product and waste trajectories.[44,45] Hotspots associated with facility-based production and industrial application are expected to be more spatially robust, whereas use-phase and end-of-life hotspots may lead to larger spatial uncertainty.[40] Such uncertainty should be considered when the inventory is applied in multimedia fate models. Fourth, because of limited knowledge regarding degradation rates, major precursor emissions were estimated, but their conversion to PFOS or PFOA was not quantified.[70] Finally, the assessment of regulatory impacts relies on temporal alignment between emission trends and policy milestones rather than on explicit modeling of policy implementation, enforcement, or compliance, and concurrent industrial, technological, and market factors may also shape the observed trends. As a result, the inferred policy effects should be interpreted as indicative rather than strictly causal. Within these scope boundaries, this inventory is positioned as a screening-level tool for national- and regional-scale emission trend analysis, multimedia fate modeling, and hotspot identification. Its 0.1° × 0.1° gridded results provide a spatially explicit baseline but are not designed for site-specific regulatory compliance. Despite these limitations, the inventory provides up-to-date and comprehensive estimates of PFOS and PFOA emissions in China and supports the evaluation of ecological and human health risks, as well as the design of PFAS-related policies.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.6c02449.

Additional description of the life-cycle material flow analysis framework, activity data, and emission factors used to construct the PFOS and PFOA emission inventory for China; details of data sources, parameter settings, stock and end-of-life modeling, and Monte Carlo uncertainty analysis; supplementary figures and tables on temporal trends of PFOS and PFOA emissions by life-cycle stage, environmental compartment, and sector; spatial distributions at 0.1° × 0.1° resolution for selected years; and comparison with previous PFOS/PFOA emission inventories (PDF)

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Notes

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