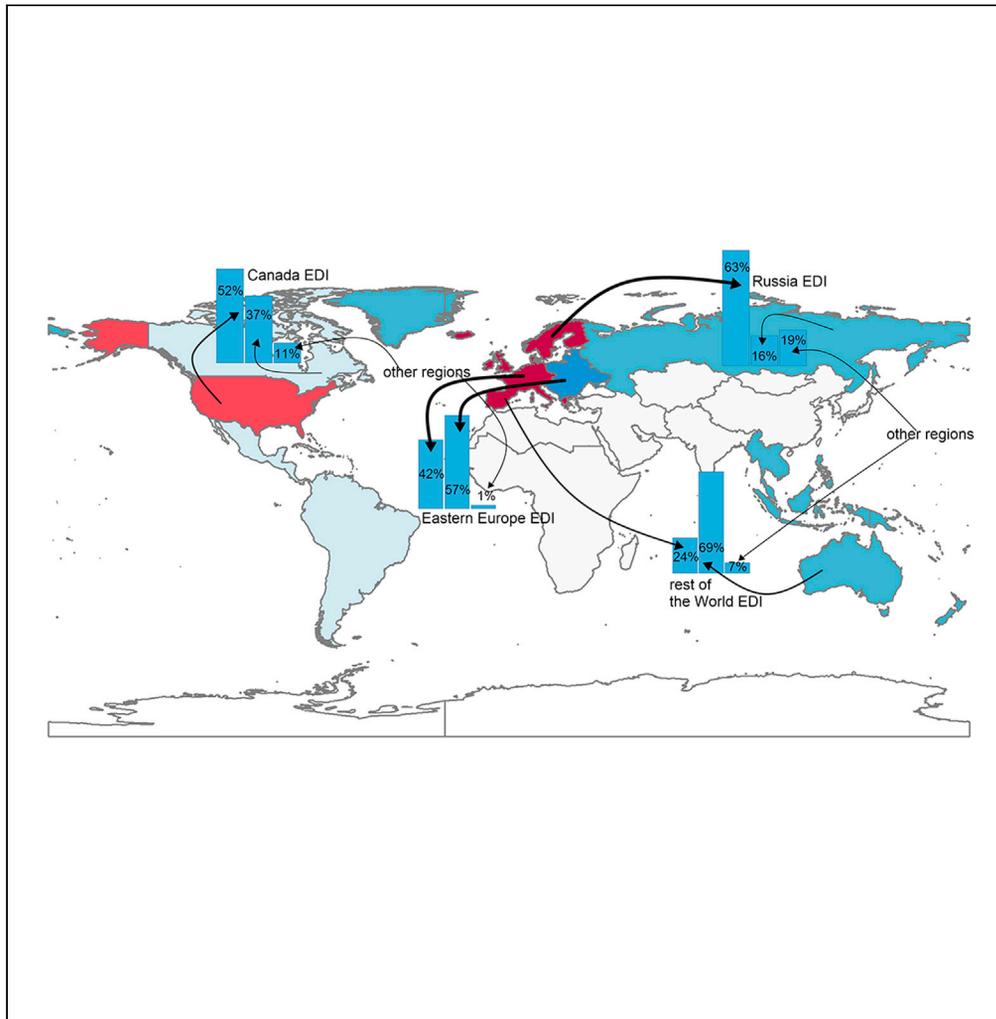


Article

The footprint of dioxins in globally traded pork meat



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Highlights

High PCDD/Fs EDI is caused by high dioxin environmental pollution and pork consumption

Global pork trade caused larger PCDD/Fs EDI flows than atmospheric transport

Western Europe and the United States yield most of the PCDD/Fs EDI transfer through pork export

Pig feed trade can affect pork consumers' exposure to dioxins

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Article

The footprint of dioxins
in globally traded pork meat

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SUMMARY

The bioaccumulation of polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs), known as dioxins, in fatty meat is one of primary pathways of entry into the human body, but levels of human exposure to dioxins in fatty meat subject to global trade are unknown. We show high dioxin estimated dietary intake (EDI) via pork consumption in Europe, the United States, and China, owing to stronger dioxin environmental contamination and high pork consumption in these countries. The dioxin risk transfer embodied in pork trade is mostly significant in high-latitude countries and regions of Canada, Russia, and Greenland because these regions with low dioxin environmental levels import large amounts of pork meat from more severely dioxin-contaminated Europe and the United States. We demonstrate that global pig feed trading decreases the exposure of pork consumers to dioxins via the import of feed from countries with low dioxin environmental contamination by pig breeding countries.

INTRODUCTION

Polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs), known as dioxins, are one of the most toxic persistent organic pollutants (POPs) (Srogi, 2008). Dioxins are unintentionally present in the environment by human activities, with their sources mainly from waste incineration and some uncontrolled combustion processes (Kulkarni et al., 2008; Duan et al., 2011; White et al., 2021; Ni et al., 2009). The persistence of dioxins in the environment enables toxic chemicals to be absorbed by living organisms and to enter the human food web, posing potential health risks to humans (Hsu et al., 2010; She et al., 2016; Weber et al., 2008). Dioxins enter the environment through atmospheric transport, deposition, sedimentation, and multimedia exchange (Hageman et al., 2015; Jurado et al., 2004). Given their high lipophilicity, these toxic chemicals tend to accumulate and amplify in the food web as the endpoint of POPs in the environment (Fernández-González et al., 2015). Up to 90% of human exposure to PCDD/Fs and dioxin-like-polychlorinated biphenyls (dl-PCBs) is attributed to the intake of animal-derived food products. PCDD/F levels in meat and fish have been found to be considerably higher than those in plants (Zhang et al., 2008a; Bocio et al., 2007; Törnkvist et al., 2011).

Dioxin (and other toxic chemicals) contamination of the environment and food web is traditionally attributed mostly to atmospheric transport and deposition on terrestrial and water surfaces from "sources" to "sinks" (Pan et al., 2013; Castro-Jiménez et al., 2010; Cohen et al., 2002; Hassanin et al., 2005). About 65%–68% of dioxins released from their sources ends up in global land surfaces owing to source proximity (this study), contaminating the terrestrial food web via accumulation in the terrestrial environment and food web. Recent studies linked domestic and international food trade with human health risks by the intake of toxic chemicals in marine fish embodied in the food trade (Huang et al., 2020; Jiang et al., 2019; Bedi et al., 2020). In the last two decades, the acceleration of economic globalization and the increasing demands on environmental carrying capacities have considerably enhanced rapid global food trade. Concerns are raised if the food harvested from those countries and regions exporting food is contaminated by toxic chemicals, and the risk would be transferred to the countries and regions importing the food. Although food inspection is extensively carried out for traded food, such inspection is expensive and often randomly conducted, which experiences increasing difficulties in meeting dramatically expanded food safety demands. As a result, food safety assessments without taking the food origin into consideration could overlook the risk embodied in traded food. In this context, food trade might provide a new and more efficient pathway for the transport of POPs because the food trade by shipping is not subject to dispersion and

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degradation, which often occur in atmospheric and oceanic transport (Ng Carla and von Goetz, 2017). This pathway has, however, been paid less attention.

Human exposure to dioxin depends not only on toxic chemical levels in food but also on food consumption. The quantity of food intake plays a vital role in exposure risk to the food. Among animal-derived foodstuffs, pork production is markedly higher than beef. Figure S1 compares global total pork and beef (cattle + buffalo) production from 2000 to 2018, indicating that pork production was 1.63 times the beef production. Pork meat accounted for 40.1% of the global meat consumption in 2015, higher than poultry (34.1%) and beef (21.0%) (Szűcs and Vida, 2017). In China alone, the average pork consumption per capita was about 39.5 kg, accounting for 66% of all meat consumption, whereas beef only accounted for 8% of the total meat consumption. Pork is also a very important component in the global meat trade. According to pork trade matrix data from the United Nations Commodity Trade Commission (UN Comtrade, <https://comtrade.un.org>), the total pork quantity in global trade reached 13.9 mt in 2012, accounting for the most share of global meat trade. Considering the higher lipid content and strong bioaccumulation of PCDD/Fs in pork meat than other meat items, PCDD/Fs tend to be transferred from pork producers to consumers via global pork trade, potentially enhancing or mitigating human exposure. Knowledge about global pork trade and human exposure risk is, however, poor. The present study aims to fill this knowledge gap by quantitatively assessing human exposure to PCDD/Fs embodied in the international pork trade. The approach adopted in the present study can be readily extended to other fat-containing food items and toxic chemicals in the global food trade.

We first built a global gridded atmospheric dioxin emission inventory (STAR Methods). We then performed extensive model simulations using a global-scale atmospheric transport model and multicompartment model (CanMETOP) for POPs to predict dioxin concentration levels in various environmental media. We combined a food web model (Huang et al., 2016) with CanMETOP to predict dioxin contamination in traded pork (STAR Methods). Finally, we implemented global pork trade matrix data to assess human exposure to globally traded pork. To highlight the significance of global trade as a new pathway of toxic chemicals, we also compare human exposure to dioxin caused by pork trade with that induced by atmospheric transport. We conducted multiple model scenario simulations by taking and not taking global pork trade into account. Considering that in many cases pigs are held indoors, pig feed might play a key role in pork contamination by dioxins. In light of this, further investigations were conducted to examine the health effect of pig feed contamination by dioxins, which are also subject to global trade, on traded pork and pork consumers.

RESULTS

Human exposure to dioxins embodied in global pork trade

The detailed information of models and data referred in this section and other sections are provided in STAR Methods, Figures S1–S17, and Tables S1, S2, S3, S4, S5, and S6.

Figure 1 shows the simulated EDI from the intake of pork produced locally and imported from other countries or regions in 2012. The high EDI primarily resulted from relatively high dioxin environmental contamination and pork meat consumption. High PCDD/Fs emissions and concentrations in air and soil are identified in Western Europe, eastern China, India, some Southeast Asian countries, Japan, the eastern United States, Sub-Saharan Africa, and coastal countries (Figures S2, S4, and S11). In particular, we have identified simulated PCDD/F levels in pork exceeding the maximum limit (1 pg TEQ/g fat) and the action level (0.75 pg TEQ/g fat) recommended by the European Community (European Food Safety Authority, 2012), in several countries, including Israel, Kuwait, Lebanon, Haiti, and El Salvador. Among three Middle East countries, the majority of the populations in Kuwait and Lebanon is Muslim but pork production for small non-Muslim populations is recorded by the FAO and UN Comtrade (STAR Methods). Israel as a non-Muslim country produces and consumes pork meat with low pork export. The estimated EDI values are also large in Western Europe, Greenland, China, and the United States. Among those countries with high EDIs, Albania has the highest EDI value at 0.74 pg TEQ per kg bw per day (95% confidence interval [CI]: 0.31–1.79), followed by Burkina Faso (0.68, 95% CI: 0.28–1.62), Belgium (0.38, 95% CI: 0.16–0.91), Spain (0.37, 95% CI: 0.15–0.88), and the Netherlands (0.35, 95% CI: 0.15–0.84). Muslim countries, such as those in the Middle East and Sub-Saharan Africa, have the lowest EDI embodied in global trade because of almost zero pork meat consumption in these countries (Figure S5). Low EDIs are also identified in Mongolia because pork is traditionally not a meat item in this country. We notice that dioxin environmental levels

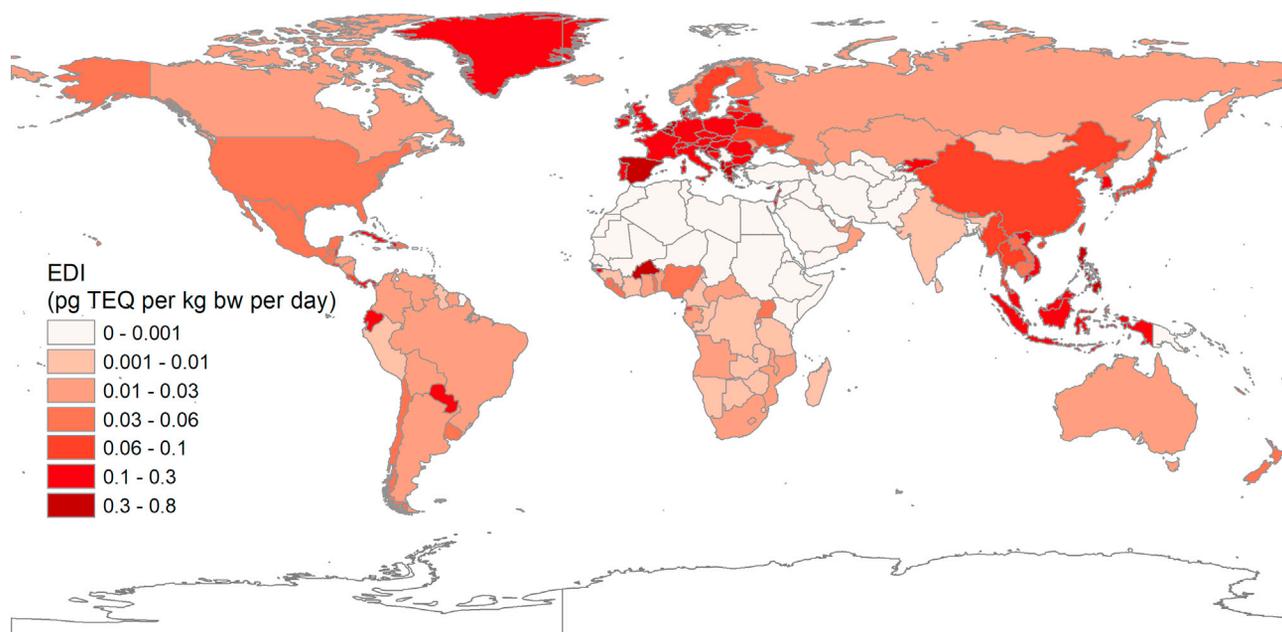


Figure 1. Modeled PCDD/Fs EDI (pg TEQ per kg bw per day) from the trade simulation in 2012

in some of these low- or zero-pork consumption countries are not low. For instance, both PCDD/Fs emissions and environmental levels in Kuwait are high, but its EDI is very low owing to its low pork consumption. Moderate EDIs are observed in Russia, Australia, Canada, and Latin America owing to low PCDD/Fs emissions and environmental contamination (Figures 1, S2, and S11).

Transfer of PCDD/Fs EDI via global pork trade

The risk transfer of PCDD/Fs embodied in the global pork trade is assessed quantitatively by comparing the dioxin EDIs simulated from the “trade” and “no trade” model scenarios. For the “no trade” case, we assumed that pork meat taken in by pork consumers was entirely locally produced and pigs were fed by the local pork feeds. The EDIs from the “no-trade” simulations are illustrated in Figure 2A. The “no-trade” model results exhibit a similar spatial pattern to the “trade + local” case, as shown in Figure 1. A direct glance at Figures 1 and 2A can identify a significant change in Greenland, where a large amount of pork meat was imported from countries of high dioxin contamination, as shown by very small EDI values from the “no trade” simulation (Figure 2A), but large EDI values are seen in the “trade + local” modeling results (Figure 1). This result implies that the local population exposure to PCDD/Fs has been altered by the consumption of imported pork meat.

The contribution of global pork trade to human exposure (EDI) via pork intake is further demonstrated by the percentage change in EDI between the “trade” and “no trade” simulations, as shown in Figure 2B. Pork trade significantly altered the PCDD/Fs EDIs in most countries, especially those importing pork from countries whose environment and pork are, to a large extent, contaminated by dioxins. We find that countries with higher dioxin levels in their locally produced pork benefit from importing pork from less dioxin-polluted countries. Our modeled EDI change rate between the “trade” and “no trade” scenarios showed that global pork trade reduced dioxin EDIs in most Latin American countries, Northeast Asia, and some European countries. For example, the PCDD/Fs EDI from pork intake embodied in pork trade decreases 301% in Burundi, 90% in the Netherlands, and 50% in Albania (Figure 2B). In contrast, the countries importing pork from those countries and regions with higher environmental PCDD/F levels enhanced the PCDD/Fs EDI. As shown in Figure 2B, PCDD/Fs EDI via pork consumption in Europe, Central and Northern Asia, and some Southeast Asian countries shows positive EDI fractions, manifesting increasing human exposure. Again, Greenland bears the largest increase in PCDD/Fs EDI via pork consumption imported from other countries, enhancing 97.5% of EDI, followed by Brunei (93.3%), Mongolia (92.9%), and Australia (78.3%). A large amount of pork meat consumed by Mongolians was imported from China and Poland, where

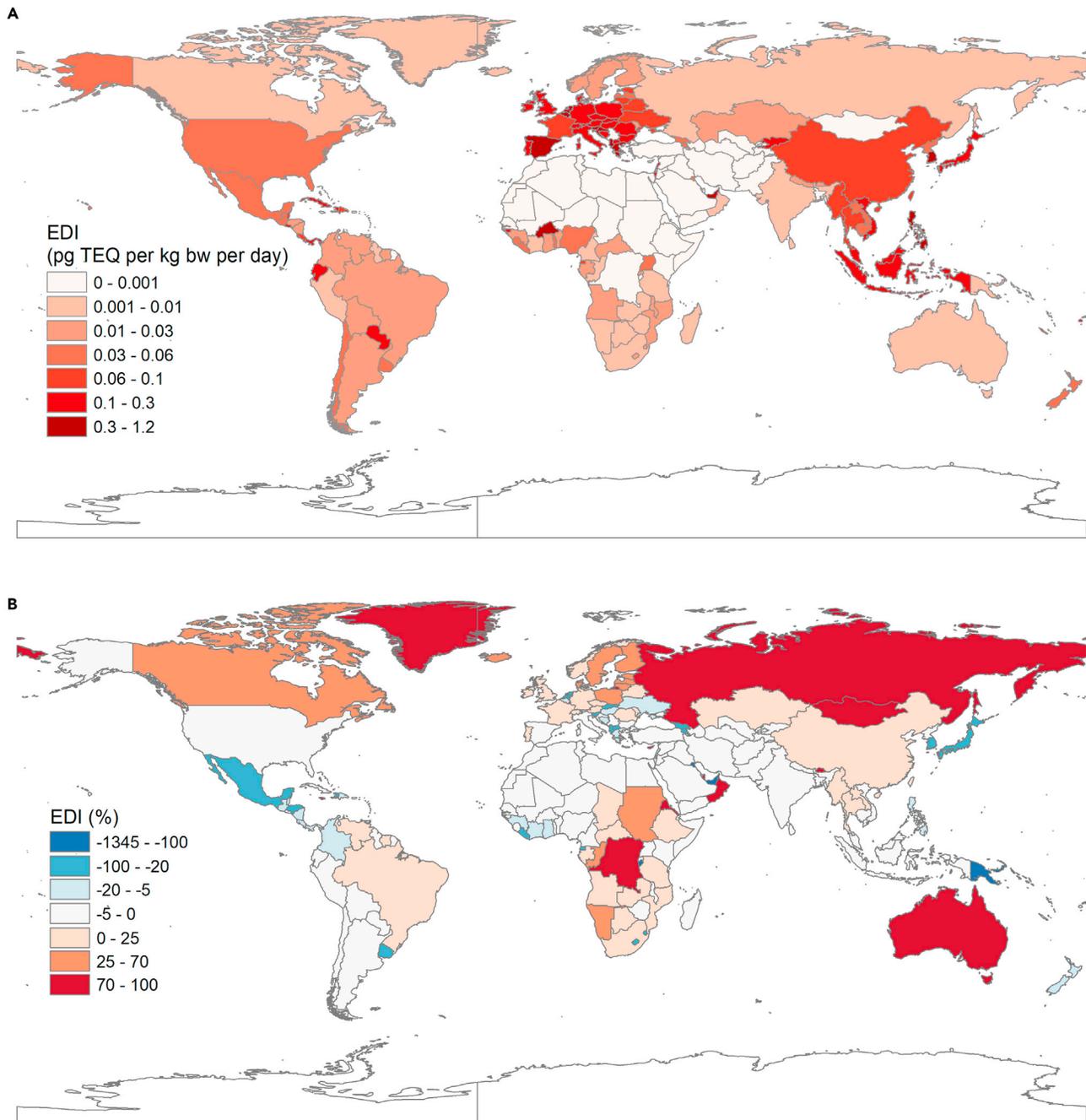


Figure 2. Modeled PCDD/Fs EDI subject to “no trade” simulation and the fraction (%) of PCDD/Fs EDI between “trade” and “no trade” simulations

(A) PCDD/Fs EDI embodied in the no trade simulation.

(B) Percent change in PCDD/Fs EDI between trade and no trade simulation in 2012, estimated as $EDI_{GT} = EDI_{DF} \times 100/EDI_T$ (%), where $EDI_{DF} = EDI_T - EDI_{NT}$ is the difference between trade and no trade simulated PCDD/Fs EDIs.

the dioxin level was significantly higher than that in Mongolia, leading to a marked increase in the PCDD/Fs EDI in Mongolia in the “trade” simulation. It is notable that high-latitude countries and regions tend to get more exposure to dioxins via pork trade, such as Greenland, Canada, and Russia (Figure 2B). This is likely that the high cost of pig farming prompts these northern regions to import pork meat from their nearby countries under higher dioxin contamination.

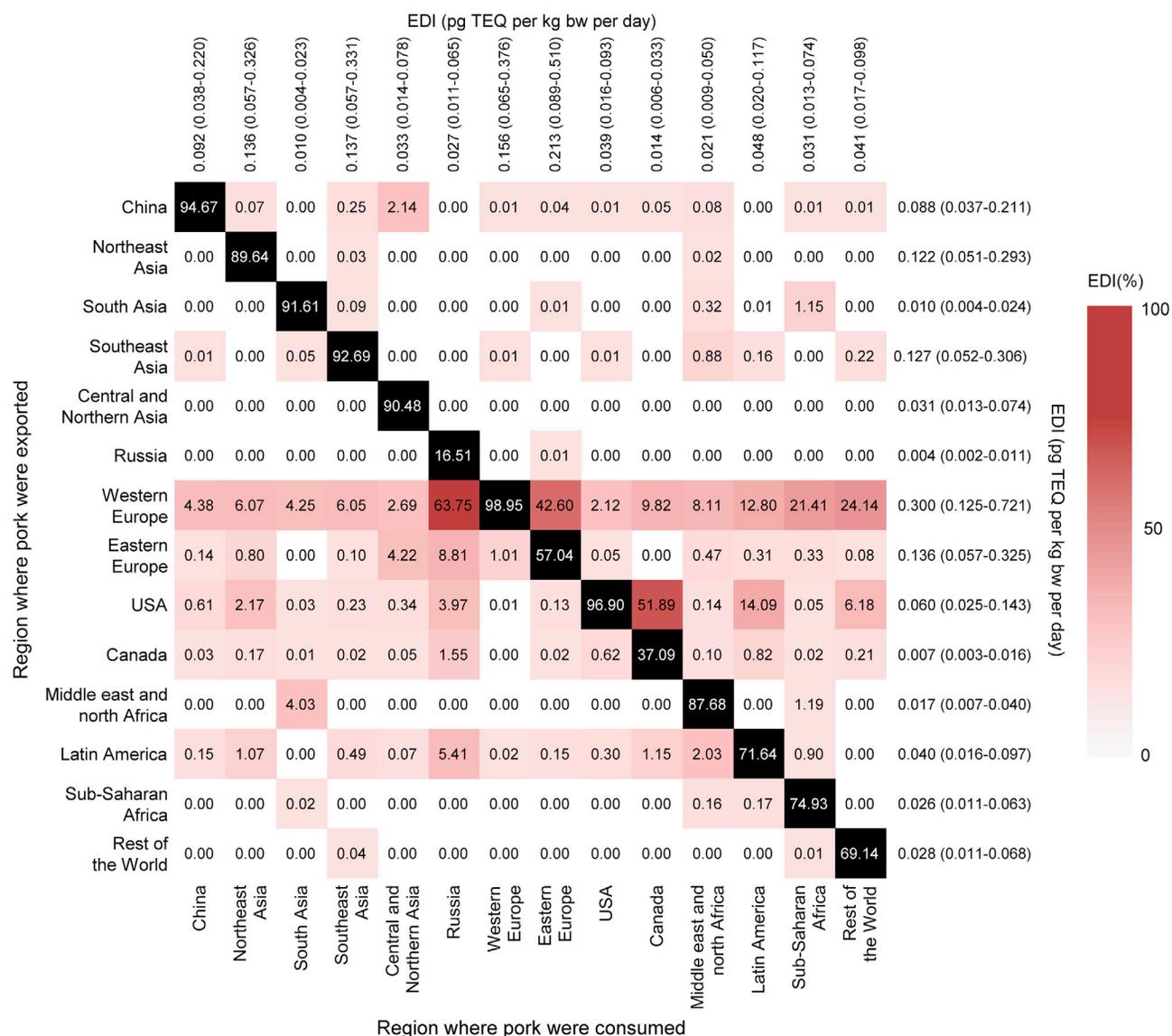


Figure 3. Proportion of PCDD/Fs EDI (%) via locally produced and traded pork intake in each of 14 selected regions

Each cell in the matrix shows the fraction of EDI (%) from pork consumption in each region indicated on the bottom x axis to that produced in the region indicated by the left y axis. The dark cells on the diagonal show the EDI (%) in each region induced by the consumption of pork produced locally. The values in parentheses represent the ranges that span the 95% CI. see [Figure S12](#)

The other important factor contributing to PCDD/Fs EDIs in a country of concern is the proportion of imported pork to the country's total pork consumption. In some countries, most of the pork supply is imported from abroad ([Figure S6](#)). If pork trade is not taken into consideration, the estimated PCDD/Fs EDI in these countries will be significantly reduced. Taking Canada as an example, since 20% of Canada's pork supply came from the United States, where dioxin levels in pork are five times higher than those in Canada ([Figure S13](#)), if imported pork from the United States was produced by Canada itself (no trade), the PCDD/Fs EDI in Canada would be reduced from 1.37×10^{-2} to 6.48×10^{-3} pg TEQ per kg bw per day. In another case, the proportion of pork imports in total pork consumption in South Asia is very low, and its PCDD/Fs EDI is not strongly associated with the global pork trade. [Figure 3](#) illustrates the proportion (%) of PCDD/Fs EDI from locally produced and traded pork in 14 regions or the contribution of the intake of locally produced and traded pork to the total PCDD/Fs EDI in each of the 14 regions. Among these regions, locally produced pork in China, South Asia, Southeast Asia, Central and Northern Asia, West Europe, and the United States contributed over 90% of their respective PCDD/Fs EDIs to their total

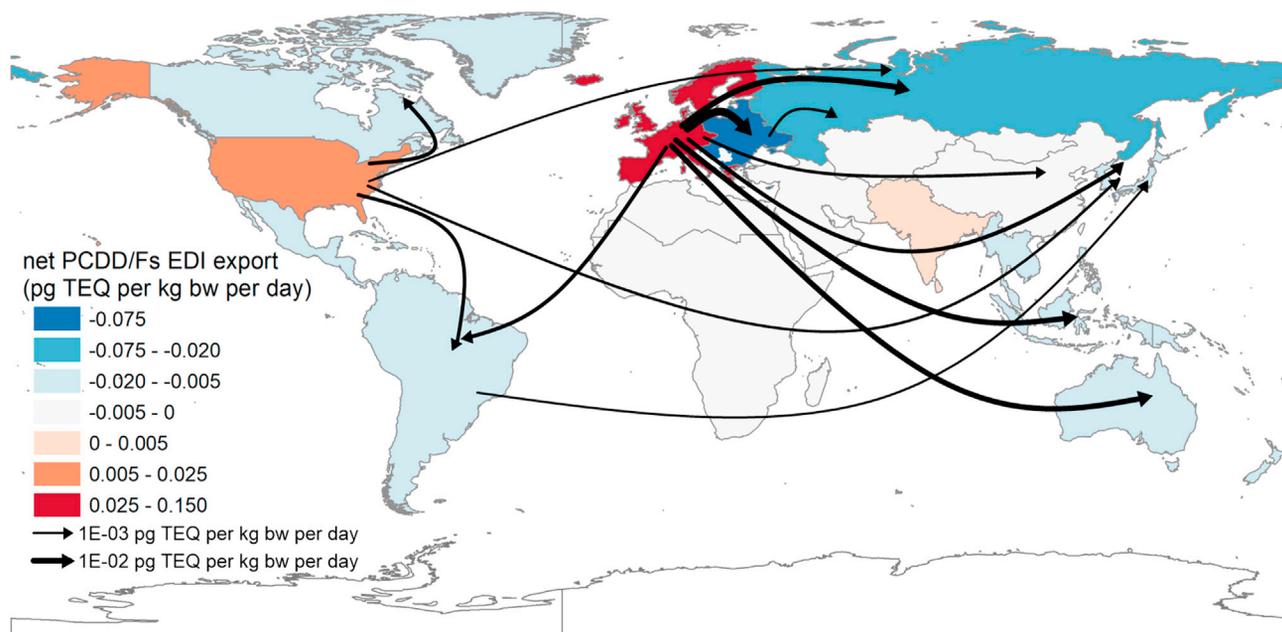


Figure 4. Net PCDD/F EDI export through global pork trade and transfer of PCDD/F EDIs among 14 regions

A positive value indicates net PCDD/Fs EDI exports in the pork trade. Arrow lines represent the pathway of EDI transfer embodied in the global pork trade.

EDIs, suggesting that dioxin exposure via pork intake in these countries mainly comes from local pig breeding. In this case, while China imported a very large amount of pork from overseas to meet its pork demand, human exposure to dioxin from pork consumption is primarily associated with local pork intake. On the other hand, human exposure to dioxin through pork intake in Russia was predominantly induced by pork import from overseas, accounting for 83.5% of dioxin EDI from imported pork consumption (Figure 3). The most significant transfer of PCDD/Fs exposure occurring in Russia follows the pathway of pork import/export between Western Europe and Russia. Considering the high environmental level of PCDD/Fs in Western Europe (Figures 1, S2, and S11), this region supplies 63.8% of the total EDI to Russia. Likewise, Canada received 51.9% of the PCDD/Fs EDI via the consumption of imported pork from the United States and the other 9.8% from Western Europe. As a result, these two countries exhibit a moderate level of PCDD/Fs EDI (Figure 1), although their national mean dioxin emissions are low (Figure S2).

Overall, our results indicate that the regions with large pork exports contribute a large portion of the PCDD/Fs EDI to those importing pork. In addition to the main EDI export pathways mentioned above (EDI transfer from Western Europe to Russia and from United States to Canada), Western Europe also contributes 21.4% to the total PCDD/Fs EDI through pork intake in Sub-Saharan Africa via pork export, 24.14% to the rest of the world, and 12.8% to Latin America. The United States contributes 14% to the total EDI in Latin America and 6% to the rest of the world (Figure 3). Figure 4 further illustrates the net EDI outflow ($EDI_{\text{export}} - EDI_{\text{import}}$) via global pork trade in 2012, characterized as the pathways of PCDD/Fs EDI transfer from one region to another via trade. As seen, Western Europe and the United States are two main (source) regions exporting dioxin EDIs through pork trade. The EDI transfer from Western Europe and US pork exports accounts for 91.6% of the total EDI transfer embodied in the global pork trade (Figure 3). As a traditional pig breeding region, Western Europe yields the largest EDI transfer at 1.45×10^{-1} pg TEQ per kg bw per day through pork export to 13 other regions. The largest EDI transfer pathway is from Western Europe to Eastern Europe, with an EDI export of 8.10×10^{-2} pg TEQ per kg bw per day, which is far greater than the EDI outflows among all other pathways (Figure 4). Another primary PCDD/Fs EDI transfer pathway is from Western Europe to Russia, in which Western Europe transfers 1.70×10^{-2} EDI (pg TEQ per kg bw per day) to Russia. This traditional and largest pig feeding region also transfers 8.25×10^{-3} and 9.83×10^{-3} (pg TEQ per kg bw per day) EDIs to Northeast Asia and the rest of the world, respectively. As the second largest dioxin EDI exporter, the United States transfers 7.09×10^{-3} (pg TEQ per kg bw per day) EDI to Canada, 6.68×10^{-3} (pg TEQ per kg bw per day) to Latin America, and 2.95×10^{-3} pg TEQ per kg bw per day to Northeast Asia.

Effect of pig feed under global trade

Feeds are the main source of dioxin contamination in farm livestock and determine dioxin levels in pork (Malisch and Kotz, 2014; Kim et al., 2007, 2011). The “trade” and “no-trade” model scenarios presented above, in reality, assume that pig feeds are provided locally. In many cases, however, pig feeds are also subject to global trade. We carried out an additional model scenario simulation to associate the global pig feed trade with pork pollution by dioxins, aiming to quantify the contribution of the pig feed trade to human exposure to dioxins via pork intake. The national level pig feeds were collected from the UN Comtrade and estimated using Global Livestock Environmental Assessment Model (GLEAM, FAO, <http://www.fao.org/gleam/resources/en/>, see Table S3 for details). Dioxin levels in feed ingredients were estimated using the same food web model (STAR Methods). Figure S14 compares the simulated dioxin concentration in pig feed by taking traded feed and local feed into consideration, showing marked differences of PCDD/Fs concentration in feed between “feed” and “trade” scenarios. We then calculated the fraction (%) (or percentage change) of dioxin concentration in pork meat from “feed” model results to that from the “trade” simulation, defined as $Con_{FT} = Con_{DF}/Con_T \times 100\%$ (%), where $Con_{DF} = Con_F - Con_T$ with Con_F and Con_T as dioxin concentration from “feed” and “trade” simulations, aiming to discern the net effect of feed trade on dioxin exposure (Figures S15 and S16). Negative Con_{FT} values are observed in many countries across the globe, suggesting that dioxin levels in pork decrease in these countries after taking pig feed trade into account, including China, South and Southeast Asia, Northeast Asia, and some Latin American countries. These regions import pig feeds from countries with lower PCDD/Fs levels in the environment (Figure S15 and S16), so pork imports in these countries benefit from the global pig feed trade. Positive Con_{FT} values are seen in Canada, Russia, Australia, northern Europe, South America, and several other countries in Sub-Sahara and Western Europe, indicating enhanced dioxin levels in pork embodied in the global pig feed trade. The largest increase in PCDD/Fs concentration in pork occurs in Brunei (1,263.44%), followed by Mongolia (639%), Sweden (143%), and Canada (114%).

Taking feed trade into account could either enhance or decrease pork consumers’ exposure to dioxins (EDI), depending on the change in the dioxin contamination level in both importers’ and exporters’ environments. Figure 5 shows the fractions (Figure 5A) of PCDD/Fs EDI (%) and the differences between “feed” and “trade” model scenario simulations (Figure 5B). In most selected regions and countries, the EDI fraction and differences are negative, indicating declining EDI values after the feed trade was taken into consideration. Of note, the positive PCDD/Fs EDI fraction and differences between the “feed” and “trade” scenarios occur in Canada and Russia, manifesting increased human exposure to dioxin-contaminated pork meat. The changes in the PCDD/Fs EDI in a certain region or country can be attributed to the direct and indirect pork and pig feed trade. Taking Canada as a case study, as mentioned above, a higher PCDD/Fs EDI in pork consumption in Canada was induced, to a large extent, by direct pork import from the United States, which has a higher dioxin environment level than Canada. However, pig feed import of the United States from low dioxin-contaminated regions and countries reduces the EDI via pork consumption in the United States, indirectly benefiting the EDI reduction in Canada’s pork meat imported from the United States. The increasing dioxin level in pork after accounting for both pork and pig feed trade in Canada also enhances PCDD/Fs EDI via pork exposure to other countries and regions. As seen in Figure 5B, the EDI embodied in pork exports from Canada to Russia increases 4.74×10^{-4} (pg TEQ per kg bw per day), followed by 4.49×10^{-4} (pg TEQ per kg bw per day) to Latin America. On the other hand, the PCDD/Fs EDI embodied in Western Europe’s pork export to other countries and regions, after accounting for pig feed trade, decreases markedly with the largest decline in Eastern Europe at -2.99×10^{-3} (pg TEQ per kg bw per day), followed by Russia at -1.3×10^{-3} (pg TEQ per kg bw per day) and Sub-Saharan Africa at 3.8×10^{-4} (pg TEQ per kg bw per day). Overall, the global pig feed trade reduces, to some extent, dioxin exposure in Western European pork consumers, benefiting from pig feed imports from lower dioxin-contaminated countries.

DISCUSSION

We demonstrate that risk transfer via the global food trade network provides another likely more efficient and direct pathway for global and regional toxic transport beyond atmospheric and oceanic transport. Here, we combined several models to quantify human exposure to PCDD/Fs embodied in the global pork meat trade. We find that pork trade leads to increased or reduced levels of dioxin exposure via pork intake in different countries or regions. Typically, by replacing locally produced pork meat under a high dioxin-contaminated environment with imported pork from regions with lower dioxin levels, Kuwait and Burundi reduced the PCDD/Fs EDI up to $-1,131\%$ and -301% , respectively. Canada, Russia, and

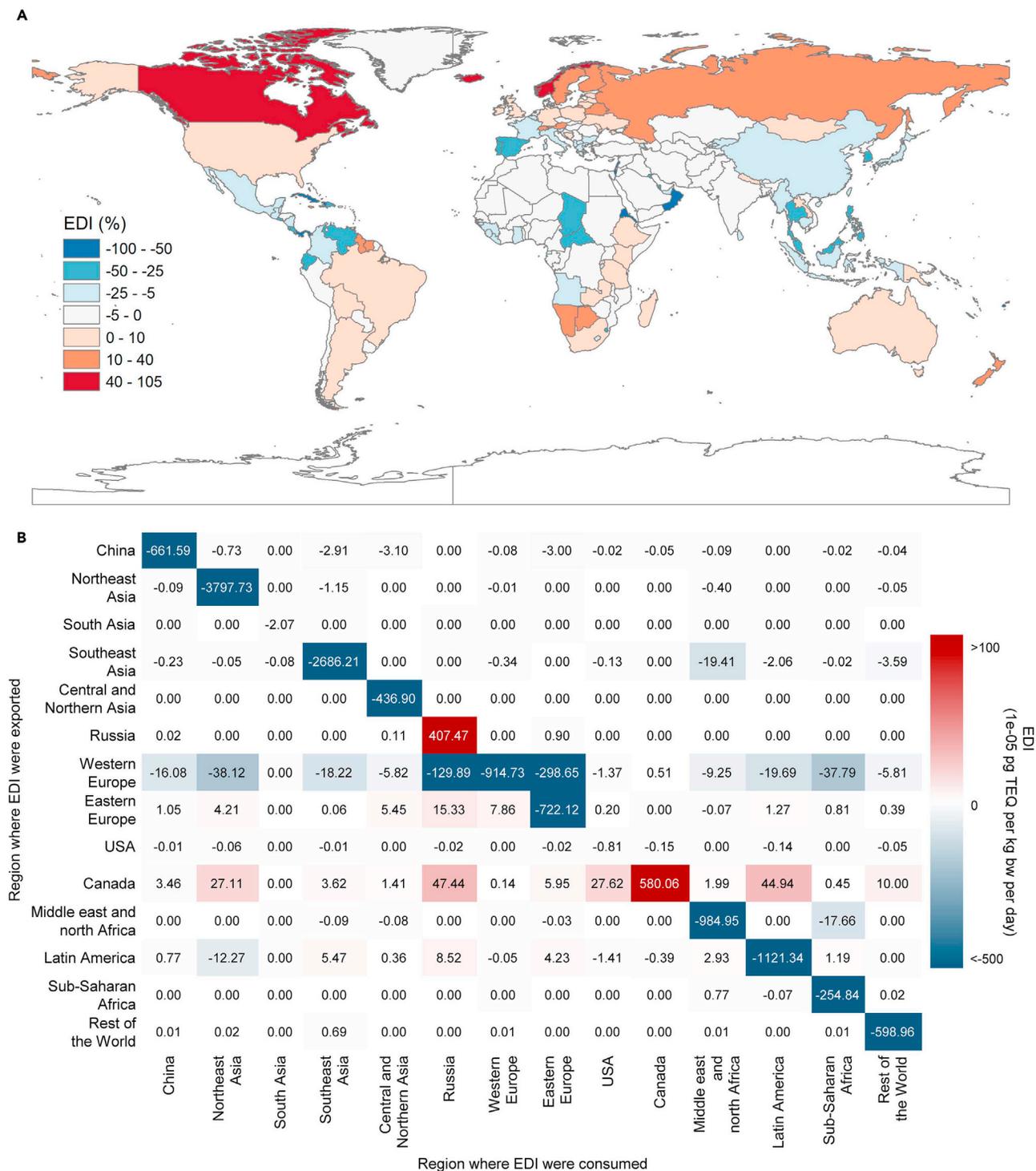


Figure 5. Fractions of PCDD/Fs EDI from “feed” simulations to that from “trade” simulation and the differences of EDI between feed and trade simulation

(A) Fraction of PCDD/Fs EDI (%) in pork between trade and feed simulations, estimated as $EDI_{GT} = EDI_{FT} \times 100/EDI_T$ (%), where $EDI_{FT} = EDI_F - EDI_T$ and EDI_F and EDI_T are feed and trade simulated EDIs.

(B) EDI_{FT} in 14 regions. To display more clearly the EDI differences, the values of the differences are multiplied by 10^5 .

Australia, where the dioxin level in the environment is low, receive increasing EDIs due to pork and pig feed imports from those regions with higher PCDD/Fs environmental contamination. Our emission inventory and modeling results reveal that most Western European countries suffer from relatively stronger dioxin pollution, in line with the field monitoring data in air and soil (Figures S7 and S8). Many European countries also have traditional pig breeding and pork production industries. Efforts have been made in Europe to assess and mitigate dioxin contamination in food and feed in recent years (European Food Safety Authority, 2012; EFSA CONTAM Panel et al., 2018; Hoogenboom et al., 2015). The Expert Panel on Contaminants in the Food Chain under the umbrella of the European Food Safety Authority (EFSA) has conducted a comprehensive review of the risks to human and animal health from dioxins in food and feed. They reviewed previous data (tolerable daily intake at 1–4 pg TEQ/kg bw provided by WHO in 1998) and proposed a new tolerable weekly intake (TWI) of dioxins at 2 pg TEQ/kg bw (EFSA CONTAM Panel et al., 2018). A continuous decline in dioxin levels in the environment and food is expected in Europe subject to the new TWI, which would enhance confidence and stimulate both pork (and other food items) import and export among countries involved in the global pork trade. This would pose a new challenge to the local efforts in dioxin emission reduction in Europe because the new TWI adopted in Europe might not be followed by other, particularly less-developed countries. The increasing demands for food items with low cost, which is one of the factors motivating international food trade, might enhance dioxin exposure if these food items would be imported from those countries or regions where present or future projected toxic contaminants would not be properly mitigated. As a result, the international food trade might harm efforts to control toxic emissions.

Extensive studies on the atmospheric and oceanic transport of bioaccumulative toxic chemicals have been conducted to assess environmental and food web contamination in receptor regions (Jiang et al., 2019; Xu et al., 2013). Compared with dioxin EDI transfer from the international pork trade, the risk embodied in atmospheric transport is considerably lower than that embodied in the pork trade (see STAR Methods and Figure S17). The total net EDI outflow among 14 regions through pork trade is 1.83×10^{-1} pg TEQ per kg bw per day, one order of magnitude higher than those from atmospheric transport (1.96×10^{-2} pg TEQ per kg bw per day). Atmospheric transport from Western Europe only contributes 5.54×10^{-6} and 1.66×10^{-7} EDI (pg TEQ per kg bw per day) to Northeast Asia and the rest of the world, three to four orders of magnitude lower than direct pork export-induced EDI values of 8.25×10^{-3} and 9.83×10^{-3} (pg TEQ per kg bw per day), respectively. The international food trade provides a unique pathway for toxic chemicals between the Southern and Northern Hemispheres because the air masses between these two hemispheres are seldom communicated. We would anticipate that rapidly increasing regional and global food trade would play an increasingly important role in the global transport of bioaccumulative toxic chemicals from their origins to destinations.

Limitations of the study

Rapid growing global food trade under globalization might enhance risk to those countries or regions importing food from food-exporting countries or regions where the environment was contaminated by toxic chemicals. To assess risk embodied in global food trade, it is necessary to link food origins to toxic pollutant origins. However, owing to the lack of domestic trade data or trade data with more specific information of locations, which often interacts with international food trade, the risk embodied in domestic food trade still experiences difficulties. Further efforts need to be made to predict the multiscale risk transfer of toxic pollutants embodied in traded food from food production and export (origin) to food consumption and import (endpoint).

STAR★METHODS

Detailed methods are provided in the online version of this paper and include the following:

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- Emission gridding
- Atmospheric transport model
- Food-web model
- Health exposure embodied in global pork trade
- Pig feed trade impact
- The impact of atmospheric transport
- Model evaluation and uncertainty

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.isci.2021.103255>.

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AUTHOR CONTRIBUTIONS

J.M., T.H., and K.C. designed the study; K.C. wrote original draft; K.C. performed model simulations; K.C., X.Z., X.L., Y.H., L.W., X.J., A.G., and Y.Z. collected the data; J.M., K.C., T.H., S.T., H.G., and J.L. participated in the acquisition, analysis, and interpretation of data.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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STAR★METHODS

KEY RESOURCES TABLE

REAGENT or RESOURCE	SOURCE	IDENTIFIER
Other		
Global pork trade data	UN Comtrade	https://comtrade.un.org
Pork meat consumption data	The Food and Agriculture Organization (FAO)	http://www.fao.org/faostat/en/#data
national PCDD/Fs atmospheric emission inventories	Stockholm Convention for Persistent Organic Pollutants	http://www.pops.int/

RESOURCE AVAILABILITY

Lead contact

Further information and requests should be directed to and will be fulfilled by the lead contact, Dr. Jianmin Ma (jmma@pku.edu.cn).

Materials availability

This study did not generate new unique reagents.

Data and code availability

Some publicly available datasets used in this article are listed in the [Key resources table](#). The other data associated with model inputs are available as reported in this article and [STAR Methods](#).

This paper does not report original code.

Any additional information required to reanalyze the data reported in this paper is available from the lead contact upon request.

METHOD DETAILS

Several models and datasets were applied in this study. We developed a multiple linear regression model to build a gridded global atmospheric emission inventory of dioxins at a 1° longitude by 1° latitude resolution. A modified version of the CanMETOP (Canadian Model for Environmental Transport of Organochlorine Pesticides) model ([Ma et al., 2003](#); [Zhang et al., 2008b](#); [Ma et al., 2004](#)) with the same spatial resolution was employed to simulate PCDD/Fs concentrations in the atmosphere, soil, and water. We combined CanMETOP with a food web model to calculate the concentrations of PCDD/Fs in pork meat, pig feed, and other food items. We estimated human exposure to PCDD/Fs associated with global pork trade by combining international food trade data and PCDD/Fs concentrations in pork across the globe. To quantify the impact of pork trade on dioxin EDI transfer, we performed two model scenario simulations. Scenario 1 is referred to as the 'trade' simulation, which takes global pork trade into consideration. The second model scenario, referred to as the 'no trade' simulation, assumes that all pork consumed is not subject to trade but produced locally. In the 'trade' scenario, we have assumed that pork is traded only once and that no processed pork meat is considered. Given that pig feed plays a crucial role in dioxin levels in pork meat, we also set an additional 'feed' scenario simulation, which incorporates globally traded pig feed in the pork food web model to quantify the dioxin EDI.

Gridded global atmospheric emission inventory of dioxins

Multiple linear regression model. [Fiedler \(2007\)](#) and [Cao et al. \(2013\)](#) summarized 86 PCDD/Fs emission inventories reported in the Stockholm Convention for POPs and identified the correlations among PCDD/Fs emission intensities and economic status in different countries. [Wang et al. \(2016\)](#) developed a multiple linear regression model to estimate the total global PCDD/Fs release. However, these inventories were created on a national and provincial level but not gridded. To create a global atmospheric emission inventory of dioxins, we collected 139 national PCDD/Fs atmospheric emission inventories in 94 countries from the Stockholm Convention for Persistent Organic Pollutants (POPs, <http://www.pops.int/>). We separated

all national inventories into a training set and a validation set. Among 139 national inventories, we took 94 dioxins atmospheric inventories in 2012 submitted by each of 94 countries to the Stockholm Convention for POPs as a training set to develop an MLR model. The other 45 dioxins inventories in other years submitted from these countries were served as a validation set to verify the MLR model predicted emissions. Following Wang et al. (2016), we selected four factors: area, gross national/regional income (GNI), GNI per capita (ppGNI), and CO₂ emission per unit GDP (CO₂pGDP) as independent variables to establish the multiple linear regression model and predict national PCDD/F atmospheric inventories. To eliminate the influence of different units and orders of magnitude on the results, we take logarithm for each factor to establish the emission regression model, given by:

$$\log E_{air} = 0.185 \log \text{Area} + 0.477 \log \text{GNI} - 0.668 \log \text{ppGNI} + 0.148 \log \text{CO}_2\text{pGDP} + 1.398$$

$$R^2 = 0.534,$$

(Equation 1)

where E_{air} is the dioxin air emission in the unit of g TEQ/year, TEQ is total toxic equivalency calculated by using 2005 World Health Organization (WHO) toxicity equivalency factors (TEF) (Van den Berg et al., 2006). We use this multiple linear regression model to predict national-level atmospheric PCDD/Fs emissions. The result shows that total dioxin emission to the atmosphere in 2012 in 196 countries is 37471.16 g TEQ/yr, among which India emitted 2992.19 g TEQ/yr, followed by China at 2954.23 g TEQ/yr, and Indonesia at 985.73 g TEQ/yr, respectively (Figure S2). The weak technological infrastructure, outdated industrial equipment and technology, and extensive open burning released a large number of dioxins into the air in many African countries (Klánová et al., 2009; Ssebugere et al., 2019; Tue et al., 2016; White et al., 2021). Given advanced industrial equipment, techniques, and emission control, dioxins emission and environmental levels in some developed countries were low (Van Metre et al., 2015; Kanan and Samara, 2018). The MLR model predicted emissions are compared to 45 emission inventories which are independent of the MLR model. The results revealed that 83% of predicted PCDD/Fs emissions are within 0.1 and 10 times of 45 PCDD/Fs emissions collected from the Stockholm Convention for POPs. 17% of predicted PCDD/Fs emissions are within 10 and 100 times of PCDD/Fs emissions reported by Madagascar (Figure S3). In general, our MLR model performs reasonably well in the prediction of PCDD/F emissions across the globe.

Atmospheric emission inventory of 17 dioxins homologues

To simulate the environmental fate of 17 dioxins homologues with different physical and chemical properties (Table S2), we also develop the emission inventory for individual dioxin homologue. There are ten main categories of emission sources in the UNEP Toolkit (http://toolkit.pops.int/Publish/Popups/07_Table1.html). The proportion of different sources varies from country to country. To calculate the proportion of each source in different countries, we separate countries who submitted their emission data to the UNEP to the four groups according to their respective personal income based on the World Bank definition (<https://databank.worldbank.org/home.aspx>). These four groups of countries are low-income, lower middle income, upper middle income, and high-income countries, respectively. According to the emission inventories submitted to the Stockholm Convention for POP (<http://www.pops.int/>), we first calculated the mean emission ratios of each emission source to the total emission in each group. We then multiplied the total emissions by the ratio (%) of each emission source to obtain the emission inventory of each emission source. The ratios of emissions from sources in the four emission groups are shown in Table S1. The result indicates that open biomass burning is the most important source and the countries with higher income always have lower ratios of this source. The possible reason is the strict restrictions on open burning in developed or high-income countries. Typically, waste incineration and heat and power generation are the two significant sources in high-income countries, in line with previous studies (Quaß et al., 2004; Dwyer and Themelis, 2015; Coudon et al., 2019). Finally, we collected 17 dioxins homologues emission factors for each emission source from literature to allocate dioxins emission to 17 homologues. The emission inventory of 17 dioxin homologues can be obtained by summing the emissions from each source sector.

Emission gridding

To allocate the national-level and provincial-level dioxin emissions to the areas where dioxin is emitted, population intensities in each country are used as a surrogate parameter to convert national and provincial emissions to gridded air emissions. In terms of significant correlations between provincial total PCDD/Fs emission and population intensities (Huang et al., 2015), the gridded population intensities were used as surrogate data to establish gridded global atmospheric emission inventory of PCDD/Fs on 1° longitude

by 1° latitude resolution. The global gridded population intensity data and map (1 latitude by 1 longitude) are obtained from the CIESIN (Center for International Earth Science Information Network) of Columbia University (available at <http://sedac.ciesin.columbia.edu/data/sets>). This modeling investigation established and employed a gridded atmospheric emission inventory of PCDD/Fs from 2002 to 2012. As shown in the gridded emission inventory, higher dioxin emissions occur in China's Henan, Shandong, and Hebei provinces. These areas have many steel and metal processing industries, power generation and heating, and waste incineration, resulting in large amounts of dioxins emissions (Huang et al., 2015). Northern India, Western Europe, and central Nigeria were also significant sources of dioxins (Figure S4).

Atmospheric transport model

Model configuration and model input data. A modified version of the CanMETOP (Canadian Model for Environmental Transport of Organochlorine Pesticides) model was employed to simulate PCDD/Fs concentrations in the atmosphere, soil, and water. CanMETOP is a three-dimensional atmospheric transport model coupled with a dynamic fugacity-based soil–air exchange model and a water–air exchange model based on the two-thin-film theory. Three-dimensional atmospheric advection, eddy diffusion, and dry/wet deposition processes are also included in the model (Huang et al., 2015; Huang et al., 2016; Ma et al., 2003; Ma et al., 2004; Jiang et al., 2017; Zhang et al., 2008b). The model needs to input meteorological data, geographic data, and physicochemical properties of the persistent organic chemicals. Meteorological data (winds, atmospheric pressure, temperature, precipitation, etc.) used the 6-hourly objectively analyzed data on a 2.5° × 2.5° latitude/longitude resolution provided by National Centers for Environmental Prediction (NCEP) Final Operational Global Analysis, the United States (US). The meteorological data were then interpolated into the CanMETOP model grids on a spatial resolution of 1° × 1° latitude/longitude and a time step length of 20 min. The coarse resolution of NCEP reanalysis winds and temperature data in the surface boundary layer (~100m) interpolated onto CanMETOP model grids are adjusted by the Monin-Obukhov similarity theory in the constant flux layer. Geographic data used in the model include terrain height, land use, soil organic content (SOC), and the surface roughness length. These data were also interpolated into the spatial resolution of 1° × 1° latitude/longitude. In addition to the meteorological data, the physicochemical properties of 17 dioxins homologues for dioxin modeling were collected from the literature (Mackay et al., 2006) and presented in Table S2. The model was integrated from 2002 to 2012 to simulate the long-term trend of 17 dioxins homologues in multiple environmental compartments (Detailed physicochemical properties and references are provided in Table S2).

PCDD/Fs concentrations in the environmental media. Figure S11 shows modeled concentrations (for 2012) of PCDD/Fs. The PCDD/Fs concentrations in ambient air ranged from 0.005 to 1958 fg TEQ m⁻³, with the mean of 12.8 fg TEQ m⁻³, and the PCDD/F concentrations in the atmospheric particle phase are higher than that in the gas phase. It can be seen that the places with high concentrations of dioxin are identified in some areas of eastern China and northern India. The distribution of dioxin concentrations in air and soil is basically consistent with its gridded emissions.

Model performance evaluation and validation. The CanMETOP has been extensively evaluated and verified against available monitored air concentration data for different organic chemicals collected from the globe (Zhang et al., 2009; Jiang et al., 2017; Huang et al., 2020; Xu et al., 2013). To establish the level of confidence in the present model investigation and to verify and evaluate PCDD/Fs emission inventory, we further conducted model validation and evaluation by comparing modeling results with measured PCDD/Fs concentrations in air and soil. There have been no extensive measurement data available worldwide due to the high cost of sampling and laboratory analysis. So the routine dioxin measurements have not been taken up to now. The UNEP Stockholm Convention for POPs has carried out the Global Monitoring Plan on Persistent Organic Pollutants and provided a harmonized organizational framework for sampling POP air concentration across the globe, including PCDD/Fs, aiming to identify the changes in their concentrations over time and their regional and global environmental transport (<https://www.pops-gmp.org/>). The dioxin monitoring data in the ambient air are mostly provided by AMAP (Arctic Monitoring and Assessment Programme), China National POPs Monitoring Project, EMEP (The co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe), and GAPS (Global Atmospheric Passive Sampling). We also collected sampled PCDD/Fs concentrations from literature and some national POPs sampling programs, such as the four-year National Dioxins Program established by the Australian Government (<http://www.environment.gov.au/protection/chemicals-management/dioxins>). The sampling locations are shown in Figure S7A. Detailed references

of measured data in ambient air are provided in Table S4. Figure S7B is a correlation diagram between modeled and field sampling data. Most simulated air concentrations are within 0.1 and 10 times of field sampling concentrations with a correlation coefficient of $r = 0.4$ ($p < 0.001$). Measured dioxin concentrations in soil are collected from the literature (Figure S8A). No extensive sampling programs were ever conducted around the globe. Figure S8B is a correlation diagram between CanMETOP simulated and sampled dioxin soil concentrations. In general, modeled soil concentrations match reasonably well with the field observation data at a correlation coefficient of $r = 0.69$ ($p < 0.001$). However, the modeled concentrations underestimated the measured concentrations slightly in soil. It is important to note that most of the measured data were collected from industrial areas. In contrast, our simulation results on a single grid cell at $1^\circ \times 1^\circ$ lat/lon resolution covering industrial, rural, and urban areas tend to underestimate the PCDD/Fs level in the soil. Still, model-predicted concentrations are within the same order of magnitude. (Detailed references of measured data in soil are provided in Table S5). Overall, the comparison between modeled and field sampling concentrations of PCDD/Fs in both air and soil shows reasonable agreements, suggesting that gridded PCDD/Fs emission inventory established in the present study and model performance are, to a large extent, reliable.

Food-web model

Model description. The contamination of 17 dioxin homologs in different food items was estimated using a food-web model (Huang et al., 2016). PCDD/Fs accumulation in plants or fodder, which are the significant components of pig feed, was obtained via the CanMETOP model predicted PCDD/Fs air-soil exchange, wet and dry particle deposition, and root uptake from soil and translocation within the plants. PCDD/Fs accumulation in pork meat was calculated by a bioconcentration model:

$$C_{\text{pork}} = \text{BCF}_{\text{pork}} \times C_d \quad (\text{Equation 2})$$

where C_{pork} is the PCDD/Fs concentration in pork (pg TEQ/g fresh weight). BCF_{pork} is a fresh weight bioconcentration factor of pork. C_d is the PCDD/Fs concentration in pig diet (pg TEQ/g fresh weight) (Huang et al., 2016; Harrad and Smith, 1997). Further details can be found in Huang's study (Huang et al., 2016).

Food web model evaluation and validation. To evaluate the food-web model performance, measured PCDD/Fs concentrations in pork were collected to compare with food-web modeling results worldwide. Because the sampled pork data were very scarce, we mainly collected measured PCDD/Fs concentrations in pork meat from different sources at different locations in Europe, China, the USA (Figure S9A). Figure S9B is a correlation diagram between modeled and measured dioxin concentration in pork meat ($r = 0.44$, $p < 0.001$). As shown, our simulated concentrations in pork meat match reasonably well with the measured concentration. Most modeled and measured dioxins concentrations have the same order of magnitude, suggesting that the food-web model can predict the PCDD/Fs level in pork nicely. It should be noted that since most samples were collected at a single or a few of food markets, the predicted PCDD/Fs concentration in pork meat at a model grid with a grid spacing of $1^\circ \times 1^\circ$ latitude/longitude stands for the mean concentration over this grid cell, which could cause bias between the predicted and measured PCDD/Fs concentrations. Detailed references of sampled concentrations in pork meat are provided in Table S6.

Health exposure embodied in global pork trade

The estimated daily intake (EDI) of PCDD/Fs through pork consumption, in units of picograms WHO₂₀₀₅-TEQ per kilogram of body weight per day (pg TEQ per kg bw per day), is defined by

$$\text{EDI} = C_{\text{pork}} \times I/W \quad (\text{Equation 3})$$

where $C(\text{pork})$ is the PCDD/Fs concentration in pork meat (pg TEQ per g fresh weight), I is the per capita daily consumption of pork (g per day), and W is the mean adult body weight, taken as 65 kg in this study (Huang et al., 2016; Xu et al., 2013). The domestic consumption of pork and global trade data in each country in 2012 are obtained from the FAO (<http://www.fao.org/faostat/en/#data/CL>) and are shown in Figure S5. In the calculation of per capita daily pork intake, we have assumed that countries with Muslim populations exceeding 90% of the national total do not consume pork. For Muslim countries with small non-Muslim populations and where the FAO reports pork meat production and trade (<http://www.fao.org/faostat/en/#data>), we also take pork consumption in the non-Muslim population into account. The population data by country in Muslim countries are collected from the Pew Research Center (<https://www.pewforum.org/2011/01/27/table-muslim-population-by-country/>). Global pork trade data are collected from UN Comtrade (<https://comtrade.un.org>) and are shown in Figures S5 and S6.

Pig feed trade impact

The farm livestock are often raised by local or industrial feed in many countries. Such industrial feed might play a crucial role in the pork contamination by dioxins if the feed is polluted by these toxic chemicals (Bernard et al., 2002; Heres et al., 2010; Hoogenboom et al., 2015; Kim et al., 2007). Given that the industrial feed is also subject to global trade, the pig feed in a country might be imported from another country, which enhances uncertainties in the assessment of pork contamination in the country importing pig feed from overseas. We designated a "feed" simulation to quantify the impact of feed trade on pork pollution by dioxins. First, we obtained the composition of pork diets from the FAO Global Livestock Environmental Assessment Model (GLEAM; <http://www.fao.org/gleam/resources>). GLEAM divides the pork production system into three sub-systems according to specific ratios in different regions across the world, namely Backyard, Intermediate, and Industrial pig production systems, and provides the regionally averaged feed ratio of pork diet for each system. Because maize, wheat, and soybean are the three main feed sources. We assume that all feed is composited and produced by these three cereals. We then recalculated the composition ratio of pig feed. Results are presented in Table S3. Next, based on the composition ratio of pig feed as well as the pork consumption from FAO (<http://www.fao.org/faostat/en/#data/BC>) and trade data of three cereals from UN Comtrade (<https://comtrade.un.org>), we estimated dioxin levels in pig feed with and without taking feed trade into consideration using Equations 4, 5, 6, and 7, defined as:

$$C_{\text{feed},i}^{\text{TRADE}} = C_{\text{feed},i} \times R_{\text{feed},i} + \sum_j C_{\text{feed},i,j} \times R_{\text{feed},i,j} \quad (\text{Equation 4})$$

$$C_{\text{feed},i}^{\text{NO-TRADE}} = C_{\text{feed},i} \times R_{\text{feed},i} + \sum_j C_{\text{feed},i} \times R_{\text{feed},i,j} \quad (\text{Equation 5})$$

$$R_{\text{feed},i} = M_i \times M_{\text{feed},i} + W_i \times W_{\text{feed},i} + S_i \times S_{\text{feed},i} \quad (\text{Equation 6})$$

$$R_{\text{feed},i,j} = M_{i,j} \times M_{\text{feed},j} + W_{i,j} \times W_{\text{feed},j} + S_{i,j} \times S_{\text{feed},j} \quad (\text{Equation 7})$$

where $C_{\text{feed},i}^{\text{TRADE}}$ and $C_{\text{feed},i}^{\text{NO-TRADE}}$ are the dioxin levels (pg TEQ/g fresh weight) in pig feed in country i with and without considering feed trade, respectively. $C_{\text{feed},i}$ is the dioxin level (pg TEQ/g fresh weight) in feed in country i obtained by the food-web model. $C_{\text{feed},i,j}$ is the dioxin level (pg TEQ/g fresh weight) in the feed from exporter j to importer i . $R_{\text{feed},i}$ is the ratio of feed produced locally to total feed consumption in country i . $R_{\text{feed},i,j}$ is the ratio of feed imported from country j to the total feed consumption in country i . M_i , W_i , and S_i are the ratios of maize, wheat, and soybean produced locally to total maize, wheat, and soybean consumption in country i , respectively. $M_{i,j}$, $W_{i,j}$, and $S_{i,j}$ are the ratios of maize, wheat, and soybean imported from country j to maize, wheat, and soybean consumption in the country i , respectively. $M_{\text{feed},i}$, $W_{\text{feed},i}$ and $S_{\text{feed},i}$ are the percentage of maize, wheat, and soybean in feed in country i , obtained from the GLEAM. $M_{\text{feed},j}$, $W_{\text{feed},j}$ and $S_{\text{feed},j}$ are the percentage of maize, wheat, and soybean in feed in country j .

The impact of atmospheric transport

The significant contribution of dioxin transfer to human exposure risk through the global pork trade paves a direct pathway from pork production to consumption. We anticipate that this new pathway of toxic chemicals is more efficient than that of atmospheric and oceanic transport in the context of risk transfer. To verify it, we examine the dioxin EDI through pork intake via atmospheric transport and deposition. We employed the CanMETOP model and food-web model and carried out several model scenario runs by turning on/off the dioxin emission in each of the 14 regions to quantify the contribution of the dioxin emission from a source region to a receptor region. The predicted air and soil concentrations and dry/wet deposition from the source-receptor scenario simulations are then input into the food-web model to obtain dioxin levels in pork meat and the EDI attributable to the atmospheric transport. Figure S17 shows the specific transfer of PCDD/Fs EDI via atmospheric transport in each of the 14 regions. 90% of estimated EDI in receptor regions are resulted from local dioxin emissions. The fraction of PCDD/Fs EDI related to the atmospheric transport ranges from 0.55% in South Asia to 9.69% in Russia. Likewise, the 95% of total dioxin EDI from the pork intake in central and northern Asia is attributed to local dioxin pollution; only very small portions of EDI via atmospheric transport can be traced back to the Middle East and North Africa (1.88%), Russia (1.09%), and South Asia (0.68%). Russia received 3.6% of EDI from eastern Europe and 1.28% from central and north Asia. As shown in Figure S17, the atmospheric transport yields very small dioxin EDI values through pork intake ranging from 10^{-3} to 10^{-4} pg TEQ per kg bw per day, one order of magnitude smaller than that from global pork trade (Figures 2 and 3). The most significant atmospheric transport-related EDI occurs in the nearby regions around a source region. The EDI associated with continental

transport only takes place between Africa-south America with very small EDI values at about 10^{-4} pg TEQ per kg bw per day.

Model evaluation and uncertainty

Our model results are subject to uncertainties from numerous variables and data. Potential uncertainties can be summarized below:

1. The emissions inventory which drives the atmospheric modeling. There are many uncertainties in the national/international PCDD/F inventories. For example, role of diffusive combustion sources, accidental/incidental fires, legacies from past chemical manufacture and use;
2. Potential sources of PCDD/Fs to pig feed, which is non-atmospheric, e.g., chemical contamination in the feedlot; contaminated soils ingested by the pigs;
3. Key uncertainties inherent in the atmospheric modeling;
4. Differences in animal feeding/husbandry practices, differences in the processing of the meat before entry to the food chain; the age of animals before slaughter, etc., which may differ in different countries;
5. Scope for huge within-country differences. e.g., Chinese scenarios are all modeled as one, but there are substantial within-country differences in PCDD/F sources, pathways, and exposures.

Given considerable data constraints, it is impossible to address all these uncertainties. For an atmospheric modeling practice, the emission inventory is often ranked as the largest source of uncertainties. As aforementioned, the emission inventories of 17 dioxin homologues were developed using national emission inventories of 139 countries reported to the UNEP following the 10 main categories of emission sources in the UNEP toolkit. Potential uncertainties are likely from reported data quality, particularly in those developing countries, emission gridding, and the lack of source heterogeneity in provincial or regional levels (e.g., the fifth uncertainty listed above). Since our model outputs gridded ($1^\circ \times 1^\circ$ lat/lon) dioxin soil and water concentrations, modeling results could be used to address contaminated soils and water ingested by the pigs. However, due to the coarse resolution and the lack of high-resolution pig farm locations, the present study did not consider this uncertainty.

Monte Carlo analysis (Rugen and Callahan, 1996; Huang et al., 2015; Callahan, 1996) was conducted to evaluate the uncertainties of global PCDD/Fs emission inventory calculated by the MLR model. In the uncertainty analysis, the coefficient of variation (CV, %) for national statistical data, such as energy and economy statistics, is within 20% (Zhao et al., 2011). Therefore, we assume that the CV for the factors in the MLR model, including gross national/regional income (GNI), GNI per capita (ppGNI), is 15%. For CO₂ emission, we set a CV of 5% proposed by Le Quéré et al. (2014). The Monte Carlo model was run repeatedly 10,000 times at the 95% confidence level. The frequency of distribution from the Monte Carlo uncertainty modeling is shown in Figure S10. Compared with the total PCDD/Fs emission estimated in 2012 in this study (37471.16 g TEQ), the emission ranged from 22510.9 to 68929.4 g TEQ.

PCDD/Fs concentrations in the environment and food simulated by CanMETOP and food-web model are also subject to uncertainties. Due to model complexity and multiple input variables, it is impossible to run the Monte Carlo model in the uncertainty assessment. Instead, we used a first-order error propagation approach to evaluate the uncertainties of simulated PCDD/Fs concentration (Cf_{out}) in the various environment and food by CanMETOP (MacLeod et al., 2002; Huang et al., 2020). The Cf_{out} is estimated by:

$$Cf_{out} = \exp\sqrt{\sum (\ln Cf_i)^2 \times S_i^2} \quad (\text{Equation 8})$$

where Cf_{out} and Cf_i are the confidence factors that span 95% confidence interval around the median of a log-normally distributed variable, and S_i is the relative sensitivity of the model output toward a change in input parameter i .

Cf of each parameter in Equation 8 is calculated as follows:

$$Cf = e^{2\sigma} \quad (\text{Equation 9})$$

where σ is the standard deviation and can be obtained by:

$$CV = \sqrt{e^{\sigma^2} - 1} \quad (\text{Equation 10})$$

where CV is the coefficient of variation, also defined as the ratio of the standard deviation to the mean. The sensitivity S in Equation 8 is calculated by:

$$S = \left(\frac{\Delta I}{I} \right) / \left(\frac{\Delta O}{O} \right) \quad (\text{Equation 11})$$

where ΔI and ΔO are the relative changes in input (I) and output (O) parameters of interest, respectively. The average sensitivity for increasing and decreasing an input parameter was calculated by varying each input parameter by $\pm 10\%$. We only choose several significant parameters to estimate their uncertainties (Cf) among multiple variables in CanMETOP and food-web model. The Cf value of model input parameters is mostly obtained from the literature (Huang et al., 2020; MacLeod et al., 2002) and presented in Table S2. The CV of PCDD/Fs emission uses the output of the Monte Carlo analysis discussed above, and the Cf value is calculated by Equations 8, 9, 10, and 11. The total Cf of global averaged EDI was 2.43, and the CF in 14 regions ranges from 2.39 to 2.48, respectively.