Release Inventories of Polychlorinated Dibenzo-*p*-Dioxins and Polychlorinated Dibenzofurans

Heidelore Fiedler

Abstract For unintentionally generated persistent organic pollutants such as polychlorinated dibenzo-*para*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF), the development and maintenance of national release inventories is an obligation for parties to the Stockholm Convention on Persistent Organic Pollutants. About 20 years after the first dioxin inventories have been published, a systematic approach has been developed and now is applied worldwide to establish complete, comparable inventories that are consistent in format and content. The basis for such inventories is the "Toolkit," a collection of emission factors and description of activities and processes that form and release PCDD/PCDF. The Toolkit uses a five-vector approach, i.e., not only releases to air but also to other compartments such as water, land, product, and residue are included. The assessment of the quantitative data for releases from ten source groups to five release vectors provides interesting insight in the country's geographic, economic, and development status. After the first round of reporting PCDD/PCDF inventories, 86 inventories have been assessed, and it can be seen that the total releases of PCDD/PCDF from the ten source categories have a positive correlation with the size of the population and a negative correlation with economic status.

Keywords Economic status, Polychlorinated dibenzofurans, Polychlorinated dibenzo-*p*-dioxins, Regional assessment, Release inventory, Stockholm Convention

H. Fiedler (✉)
Örebro University, School of Science and Technology, MTM Research Centre, SE-70182 Örebro, Sweden

Division of Technology, Industry and Economics/Chemicals Branch, United Nations Environment Programme, chemin des Anémones 11-13, CH-1219 Châtelaine(GE), Switzerland
e-mail: heidelore.fiedler@oru.se

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Abbreviations

µg	Microgramme
CEE	Central and Eastern European (group of countries)
COP	Conference of the Parties (to the Stockholm Convention)
EF	Emission factor
g	Gramme
GNI	Gross national income
GRULAC	Group of Latin American and Caribbean Countries
HCB	Hexachlorobenzene
ISO	International Organization for Standardization
kg	Kilogramme
km^2	Square kilometer
NIP	National implementation plan
p	*para*
PCB	Polychlorinated biphenyls
PCBz	Pentachlorobenzene
PCDD	Polychlorinated dibenzo-*para*-dioxins
PCDF	Polychlorinated dibenzofurans
pg	Picogramme
POPs	Persistent organic pollutants
PPP	Purchasing power parity
SG	Source group
TEF	Toxicity equivalency factor
TEQ	Toxic equivalent
UNEP	United Nations Environment Programme
WEOG	Western European and Others Group
WHO	World Health Organization
yr	Year

1 Introduction

Polychlorinated dibenzo-*para*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) have never been produced for any purpose other than laboratory experiments. They are unintentionally formed in industrial-chemical processes [1–3], such as chemical manufacture or pulp and paper production, and thermal processes [4–6], such as waste incineration, recycling of metals, the production of minerals or forest fires, and release to the environment. The predominant mechanism or pathway to generate PCDD/PCDF can vary from process to process resulting in a wide range of source-specific emission factors that also take into account different factors, such as reduction and abatement technologies, to reduce the releases [7]. It is generally accepted that the main sources of PCDD/PCDF are human activities [8].

The Stockholm Convention on Persistent Organic Pollutants (POPs) entered into force on 17 May 2004. In August 2015, 179 countries were party to the Convention through ratification, acceptance, approval, or accession [9]. An updated list of parties to the Convention and full information on the Stockholm Convention is available on the Web site of the Treaty Section of the United Nations at the following URL address: http://untreaty.un.org. The Convention intends to stop production and use of intentional POPs (e.g., pesticides such as DDT or industrial chemicals such as polychlorinated biphenyls (PCB)) and "reduce the total releases derived from anthropogenic sources" of unintentional POPs (i.e., polychlorinated dibenzo-*para*-dioxins/polychlorinated dibenzofurans (PCDD/PCDF), hexachlorobenzene (HCB), polychlorinated biphenyls (PCB), and pentachlorobenzene (PCBz)). In order to track the environmental and human exposure to these compounds, the Convention requires the parties to undertake continued measurements of their releases and concentrations. Article 5 of the Stockholm Convention [10] requests countries to develop an action plan "designed to identify, characterize and address the release of the chemicals" regulated by the Convention. The action plan shall include "[...] an evaluation of current and projected releases, including the development and maintenance of source inventories and release estimates."

In order to assist countries to establish release inventories of polychlorinated dibenzo-*p*-dioxins and dibenzofurans at national or regional level and to fulfill the requirements on release reduction under Article 5 of the Convention, UNEP through an expert group has developed a Toolkit for the development of release inventories of unintentional POPs [11]. The information contained therein comes from published scientific literature, government reports, Internet sources and through personal communication to the UNEP expert group. The Toolkit (published in 2013) is the most comprehensive available compilation of emission factors for all relevant PCDD/PCDF sources and is useful particularly in countries where measurement data are limited, enabling the elaboration of source inventories and release estimates by using the default emission factors. It is also useful in countries where national measurement data are available, as a reference document for data comparison and validation purposes. Therefore, the Conference of the Parties at its fifth meeting in 2011 (COP-5) encouraged the use of the Toolkit and adopted the

reporting format from the Toolkit. Through its structure for reporting, i.e., ten source groups and five release vectors, it is possible to gain some further insight into the global situation as to the sources of PCDD/PCDF releases [12].

This paper gives a brief overview on PCDD/PCDF release inventories as of December 2014 and then presents assessments of national inventories according to the total releases and specific releases of PCDD/PCDF, such as according to source groups or release vectors. Further, statistical assessments are presented in particular in relation with geographic and socioeconomic factors.

2 History

Sources of PCDD/PCDF have been addressed systematically since about 1980 when Esposito et al. [13] published the first comprehensive report on sources of "dioxins," especially tetrachlorodibenzodioxin (TCDD). Since about 25 years, dioxin inventories have been presented. For other persistent organic pollutants, attempts have been undertaken to quantify their releases and regional distribution such as by Breivik et al. [14]; however, the abundance of information as exists for PCDD/PCDF is not available for any other POP.

At Dioxin'90 [15], Fiedler and coworkers [16] published the paper entitled "Dioxin Emissions to the Air: Mass Balance for Germany Today and in the Year 2000." The results were mainly based on measured emission data and had a total annual emission of 928.5 g I-TEQ to air from sources in Germany (former Federal Republic of Germany). Although releases in solid residues such as slags, fly ashes, and sludges have been quantified, a systematic approach for estimating these releases had not been undertaken.

In 1999, UNEP published a report presenting the results of 15 emission inventories; the reference year was around 1995 [17]. Figure 1 shows the distribution of sources within and between countries.

With the entry into force of the Stockholm Convention and the recommendation to use one methodology for the development and presentation of national PCDD/PCDF inventories and report results back to the Conference of the Parties, comparison of release inventories became easier.

The Toolkit uses the five-vector approach and countries are able to estimate PCDD/PCDF releases from each source category to the following environmental media: air, water (surface and ground water, including marine and estuarine water), and land (surface soils) as well as to these process outputs: products (such as chemical formulations, including pesticides or consumer goods such as paper, textiles, etc.) and residues (including certain liquid wastes, sludge, and solid residues, which are handled and disposed of as waste or may be recycled).

In the early 2000s and presented at Dioxin 2004 in Berlin, 23 national release inventories were available that have been made with the UNEP Toolkit methodology [18]. Among the most important sources, open fires in agriculture/forests as well as open burning of wastes have been identified as the major sources of PCDD/

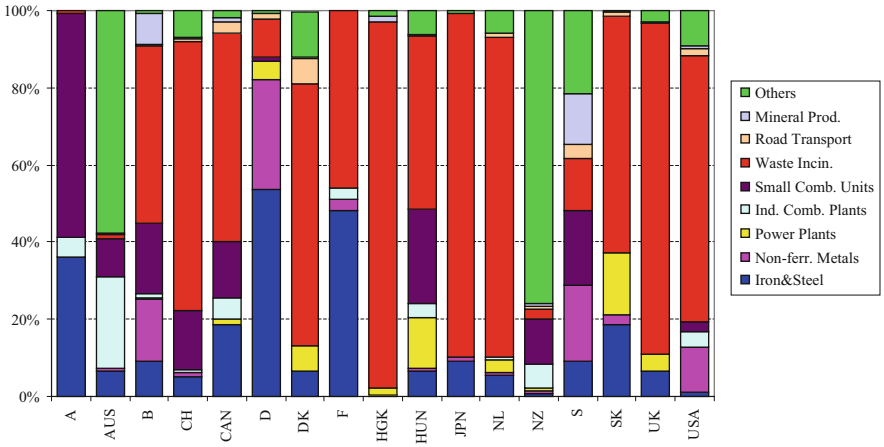


Fig. 1 Contribution of PCDD/PCDF emission source groups per country (% on the basis of TEQ yr^{-1}) [17]

PCDF. At Dioxin2007, Fiedler [19] presented the actual status of global POPs inventories, which were divided into two types of PCDD/PCDF inventories:

1. Estimated releases of PCDD/PCDF to air and as totals by countries that did not apply the UNEP Toolkit methodology (12 countries): Air emissions were 3,804 g TEQ yr^{-1} and total releases were 4,148 g TEQ yr^{-1}. Without a common methodology, most country estimates reported emissions to air only, did not address the same sources, and typically did not assess releases to water, land, residues, or products.
2. Estimated releases by countries that applied the UNEP Toolkit methodology (43 countries; different from countries included in (1) above): Air emissions were 10,911 g TEQ yr^{-1} and total releases 23,877 g TEQ yr^{-1}. Since these inventories used the same methodology, typically all sources listed in the Toolkit were assessed and the five release vectors included. Therefore, the results between countries are better comparable.

In this paper [19], for the first time, so-called population equivalents in μg TEQ per year were included. Such normalization was found to be helpful to put results into perspective and which can also serve as an orientation for a country if the own estimate fits into the scale of estimates from other countries. Across all countries, it was found that the following average releases per capita and per year did apply: 12 μg TEQ yr^{-1} $person^{-1}$ to air and 21 μg TEQ yr^{-1} $person^{-1}$ for total releases.

At Dioxin 2012, Fiedler et al. [20] presented a geographic and socioeconomic assessment of PCDD/PCDF inventories, based on 68 national release inventories; the majority of them were developed using the 2005 version of the UNEP Standardized Toolkit. The total releases accounted for 58,700 g TEQ per year.

Based on the same dataset, the quantitative releases have been correlated to geographic, demographic, and source-specific information, exploring the release

patterns of PCDD/PCDF influenced by economic status and methodology that is fair, accurate, and objective enough to assess International PCDD/PCDF Reduction Burden [21].

In this research, the results of national release inventories of PCDD/PCDF that have been developed by using the UNEP Toolkit are presented and evaluated. Using the same methodology, the Toolkit, it is ensured that the source inventories and release estimates are complete in the sense of the Toolkit methodology, transparent, and consistent in format and content. The results will allow parties and others to compare results, identify priorities, mark progress, and follow changes over time at the national, regional, and global levels.

3 Materials and Methods for the Assessment of Dioxin/Furan Inventories

3.1 Inventory Methodology

The "Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs under Article 5 of the Stockholm Convention" [11] or "PCDD/PCDF Toolkit" for short provides emission factors for the five release vectors, i.e., air (EF_{Air}), water (EF_{Water}), land (EF_{Land}), product ($EF_{Product}$), and residue ($EF_{Residue}$). Together with national activity data, this approach allows the development of release inventories for total releases (all source groups and all five release vectors) but also presentation of releases to, e.g., air only or sectoral consideration according to source groups.

Release inventories are obtained by measuring the "activity rate" of dioxin-releasing activities and multiplying them by a specific "emissions factor." For a given country, the total releases are given by

$$\mathrm{TEQ_{PCDD\text{-}PCDF}} = \sum \mathrm{j\ ActivityRate}_j \times \mathrm{EmissionFactor}_j$$

where $\mathrm{ActivityRate}_j$ is the activity rate of the source j and the $\mathrm{EmissionFactor_j}$ is the emission factor for this source j for each of the five vectors, including air, water, land, product, and residue.

For this assessment, the national inventories, compiled according to the UNEP Toolkit methodology for estimating PCDD/PCDF releases into the environment with five vectors and ten source groups, have been entered into an MS Excel databank and assessed further.

Values of PCDD/PCDF releases are presented as toxic equivalent (TEQ) using the concept of toxic equivalency which measures the relative dioxin-like toxic activity of different congeners of polychlorinated dibenzo-*p*-dioxins and dibenzofurans and expresses the result in a single number, the toxic equivalent (TEQ). The Stockholm Convention on Persistent Organic Pollutants initially uses the toxicity

equivalency factors (TEFs) established by a World Health Organization (WHO) expert group in 1997 and published in 1998 [22] and not yet the scheme established in 2005 [23]. The TEFs rank the toxicity of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in comparison with 2,3,7,8-tetrachlorodibenzo-*p*-dioxin.

Following the UNEP Toolkit methodology, the TEQ is not adhered to a specific scheme of toxicity equivalency factors (TEFs). For their "order-of-magnitude" estimates of emission factors, the differences between the WHO_{1998}-TEFs and other TEF schemes previously or later established are negligible. Therefore, the TEF scheme accompanying the emission factors is not detailed further in the Toolkit, and as a consequence, the national releases are expressed in g TEQ per year without further specification of the TEF scheme.

3.2 Data Sources

Early PCDD/PCDF inventories have been compiled from the published literature and national reports. With the entry into force of the Stockholm Convention on Persistent Organic Pollutants, official national reporting has gained importance and information has been drawn from, e.g., submissions of parties to the Stockholm Convention according to national reporting under Article 15 or from national implementation plans prepared according to Article 7. Statistical data on population, economics, and major pollutant emissions have been extracted from the World Bank database .

In order to obtain comparable results for assessment, PCDD/PCDF inventories have been compiled from reports submitted by parties to the Stockholm Convention in their national implementation plans (NIPs) [24], national reporting formats according to Article 15 [25], other national reports or the scientific literature. Within its program of work, the Chemicals Branch of the United Nations Environment Programme (UNEP) regularly searches and updates a database, which is maintained in MS Excel. Inventory information is compiled according to ten source groups covering the main sources of PCDD/PCDF and five release vectors, i.e., air, water, land, product, and residue.

The statistical data such as population, land area, and gross national income per capita were extracted from the World DataBank, compiled and published by the World Bank [26]. For denominators such as population and GNI, these informations were referred to according to the reference year of the PCDD/PCDF inventory. Economies are classified according to GNI per capita (gross national income per capita) using the World Bank Atlas method. The four economic groups are (http://data.worldbank.org/about/country-classifications; economies are divided according to 2012 GNI per capita, calculated using the World Bank Atlas method) as follows: low income (L), lower middle income (LM), upper middle income (UM), and high income (H). It should be noted that both denominators were found to be highly volatile; e.g., whereas countries belonging to the low-income group in 1999 had a GNI PPP^{-1} lower than 755 international dollar, the threshold in 2011 was at 1,205 international dollar

(http://econ.worldbank.org/WBSITE/EXTERNAL/DATASTATISTICS/0,,contentMDK:20487070~menuPK:64133156~pagePK:64133150~piPK:64133175~theSitePK:239419,00.html).

4 Results

4.1 Overall Results

In 2014 and used in this review, 85 national inventories and one inventory for Hong Kong, Special Administrative Region (CHN-HKG SAR), have been available for assessment (Table 1). For all countries, the most recent inventory has been used. It should be noted that the reference years for which the releases have been estimated range over 12 years with the inventory for the Philippines being the oldest (reference year 1999) and for Zimbabwe being the most recent one (reference year 2011). Of the 86 inventories, seven countries reported releases to air only (AUT, BGR, CHE, DEU, FIN, FRA, RUS).

Figure 2 shows the graphical sketch for each country in Table 1 for the total releases and releases per vector; the releases summed-up per release vector are shown in Table 2. Countries colored in green have releases much lower than the average of all countries, and countries colored in red have releases much higher than the average (for both 5% or 95%, resp.). With respect to total releases, there are 17 countries with total releases significantly lower than the average of all countries and 11 countries that have significantly higher emissions (shown in red color). Countries releasing ±10% of the calculated average are shown in yellow color.

4.2 Quantitative Inventory Results Using Country Basis

4.2.1 Releases According to Five-Vector Approach

The total releases accounted for 70,819 g toxicity equivalents per year (g TEQ yr^{-1}). Of these, 47% were emitted to air (approximately 33,500 g TEQ yr^{-1}), 32% were found in residues (approximately 22,600 g TEQ yr^{-1}), 11% released to land (approximately 7,700 g TEQ yr^{-1}), and smaller amounts, i.e., 8%, were attributed to products (approximately 5,700 g TEQ yr^{-1}) and only 2% to water (approximately 1,300 g TEQ yr^{-1}) (Table 2). It should be noted that the ashes generated in open burning processes such as forest or agricultural fires or from open burning of waste were assigned as "release to land." Numerically, the highest releases were from China (10,238 g TEQ yr^{-1}) followed by India (8,658 g TEQ yr^{-1}) (among others, colored red in Fig. 2); the lowest releases were reported for Niue (0.56 g TEQ yr^{-1}) followed by Brunei Darussalam and Samoa (1.4 g TEQ yr^{-1}).

Table 1 National releases of PCDD/PCDF per country and release vector ($n = 84$)

Country	ISO-3	Air	Water	Land	Product	Residue	Total	Reference year
Albania	ALB	58.7	57.5	0.007	26.8	0.07	143	2004
Argentina	ARG	874	3.1	241	29.4	964	2,111	2003
Armenia	ARM	5.49	5.27	0.83	17.5	22.9	52	2001
Australia	AUS	498	3.41	1,278			1,780	2002
Austria	AUT	39.8					40	2004
Azerbaijan	AZE	67.7	9.22	1.00	1.92	47.9	128	2003
Bangladesh	BGD	188.4	62.1	0.0	198.0	37.3	486	2005
Belarus	BLR	26.1	0.46	1.71	0.05	73.1	101	2006
Benin	BEN	182	12.8	7.50	65.3	111	379	2002
Brazil	BRA	1,168	22.6	79.3	419	546	2,235	2008
Brunei	BRN	0.75	0.02	0.03	0.04	0.56	1.4	2001
Bulgaria	BGR	255					255	2003
Burkina Faso	BFA	300	12.6	59.7	10.7	402	785	2002
Burundi	BDI	190	0.10	0.00	0.05	4.91	195	2004
Cambodia	KHM	273	0	14.6	0	319	607	2004
Cameroon	CMR	397	0.02	186	0.03	13.7	597	2009
Canada	CDN	157	0.20	6.90			164	2004
Cape Verde	CPV	18.5	0.0004	0.001	0.00003	0.16	19	2005
Chile	CHL	51.7	2.55	16.9	7.17	7.31	86	2003
China	CHN	5,043	41.2	953	174	4,026	10,238	2004
China Hongkong SAR	CHN-HKG	2.70	0.86	0.48	0.06	16.7	20.8	2003
Colombia	COL	479	20.0	18.4	32.8	240	790	2002
Côte d'Ivoire	CIV	416		6.00		9.8	432	2002
Croatia	HRV	116		1.70	0.80	49.5	168	2001
Cuba	CUB	163	1.50	27.8	25.2	7.69	225	2002
Djibouti	DJI	50.8	55.6			12.9	119	2003
Ecuador	ECU	65.0	3.43	9.30	3.32	16.5	98	2002
Estonia	EST	13.7	0.15	0.12		15.2	29	2000
Ethiopia	ETH	154	3.84	5.95	29.6	21.5	215	2003
Fiji	FJI	11.2	0.12	0	7.31	0.60	19	2002
Finland	FIN	26.2					26	2005
France	FRA	247					247	2003
Gabon	GAB	135	5.27			32.8	173	2005
Gambia	GMB	107	3.96			65.4	177	2000
Germany	DEU	116					116	2002
Ghana	GHA	386	0.12	279	0	3.04	668	2004
India	IND	2,827	22.7	30.3	314	5,464	8,658	2009
Indonesia	IDN	1,847	81.2	436	3,545	1,443	7,352	2003
Iran	IRN	1,071	0.10	31.8	399	66.6	1,568	2005
Jordan	JOR	64.3	0.42	0.07	0.35	16.4	82	2003

(continued)

Table 1 (continued)

Country	ISO-3	Air	Water	Land	Product	Residue	Total	Reference year
Kenya	KEN	3,103	2.97	2.29	17	1,613	4,738	2005
Lao PDR	LAO	46.9	0.02	18.8		38.0	104	2005
Lebanon	LBN	79.0	1.20	0.02	3.05	82.6	166	2004
Liberia	LBR	186		7.50		121	315	2004
Lithuania	LTU	37.4	0.10	0.30	0.44	18.6	57	2005
Macedonia	MKD	163	3.14			8.61	175	2001
Madagascar	MDG	119	1.00	29.6	4.67	180	334	2002
Mali	MLI	35.0		0.53	2.17	1.79	39	2005
Mauritius	MUS	19.6	5.41	0.32	0.50	4.58	30	2003
Moldova	MDA	13.5	755	4.21		2.85	776	2001
Morocco	MAR	167	3.30	0.22	19.1	45.95	236	2003
Nepal	NPL	202	0.05	43	6.90	80.0	332	2003
New Zealand	NZL	17.4	1.58	35.5		34.9	89	2008
Nicaragua	NIC	191	0.47	303		3.15	498	2004
Nigeria	NGA	2,784	0.03	2,521		34.4	5,340	2004
Niue	NIU	0.39		0.004		0.17	0.56	2004
Norway	NOR	32.9	0.30	2.00		0.00	35	2004
Palau	PLW	0.10	0.00	0.00	0.00	2.14	2.25	2007
Panama	PAN	48.2	0.38	13.9	0.01	37.2	100	2005
Paraguay	PRY	70.7	0.20	8.50	0.22	76.3	156	2002
Peru	PER	193	0.16	61.5	4.14	165	424	2003
Philippines	PHL	328	43.8	46.9	77.6	38.1	534	1999
Portugal	POR	38.5	0.36	7.55	8.78	40.1	95	2006
Romania	ROU	136	0.15			454	590	2004
Russia	RUS	1,785					1,785	2007
Samoa	WSM	1.05				0.33	1.4	2004
Serbia	SRB	123	34.8	30.1	0.70	209	398	2006
Seychelles	SYC	4.06			0.07	1.27	5.4	2003
Slovenia	SVN	6.19	0.93		3.31	20.0	30	2005
South Africa	RSA	709	2.70	64.2	30.9	1,956	2,763	2006
Sri Lanka	LKA	171	0.08		6.45	79.5	258	2002
Sudan	SDN	376		52.4	24.0	540	992	2004
Swaziland	SWZ	47.3	0.01	69.5	0.13	0.23	117	2006
Switzerland	CHE	16.7					17	2005
Syria	SYR	352		208	61.9	0.88	623	2006
Tajikistan	TJK	32.0				141	173	2003
Tanzania	TZA	528	0	184	0	252	964	2007
Thailand	THA	286	1.33	6.64	8.36	767	1,070	2005
Togo	TGO	432	0.12	14.0		72.6	519	2002
Tunisia	TUN	139	0.50	0.66	0.97	67.2	209	2004
Turkey	TUR	1,249	0.30	96.0	123	695	2,163	2006
Uruguay	URY	18.7	0.15	1.24		26.8	47	2003
Venezuela	VEN	618	1.72	37.6	8.11	435	1,100	2007

(continued)

Table 1 (continued)

Country	ISO-3	Air	Water	Land	Product	Residue	Total	Reference year
Vietnam	VNM	16.0	1.46	1.05	2.19	48.2	69	2002
Zambia	ZMB	290		48.4		145	483	2004
Zimbabwe	ZWE	154	0.27	131		1.03	285	2011
Total		33,459	1,296	7,746	5,722	22,595	70,819	

Concentrations in g TEQ per year. Empty cells indicate that releases were either not estimated or do not occur (e.g., no emission factor in the Toolkit)

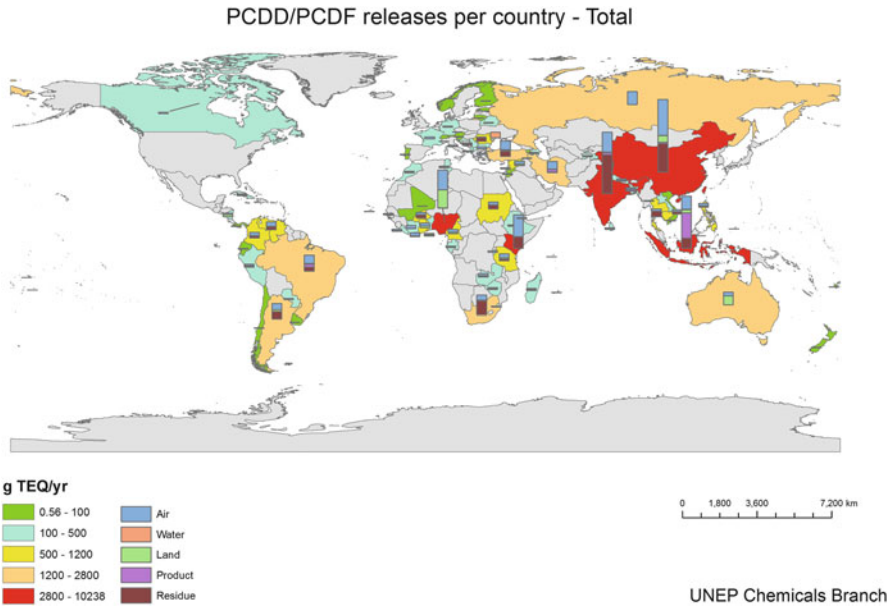


Fig. 2 Countries that reported low (*green* or *blue* color, resp.) or high releases of PCDD/PCDF (*orange* and *red* color, resp.). For countries colored in *grey*, no information has been provided so far; they are not included in Table 1. Countries with releases ±10% around the average calculated release are shown in *yellow* color. The *bar graphs* demonstrate the share for each release vector

Table 2 Summary of PCDD/PCDF releases per vector (based on 86 reports)

	Air	Water	Land	Product	Residue	Total
Release per vector (g TEQ yr^{-1})	33,459	1,296	7,746	5,722	22,595	70,819
Release per vector (% of total)	47	2	11	8	32	100

The 85 assessed countries (without CHN-HKG or Hong Kong SAR) constitute 44% from the total of 193 member states in the United Nations [27] but represent 68% of the global population of 6.966 billion in 2011 [28] (the reference year of the most recent inventory).

4.2.2 Regional Distribution of Releases and Socioeconomic Factors

Often, and as a recommended approach in the Stockholm Convention, release inventories may be developed on a regional basis. The total releases as well as the releases to the five release vectors are shown in Table 3. Accordingly, the highest overall releases are reported to originate in Asia (49%) followed by Africa (30%) and GRULAC (11%); CEE (6.4%) and WEOG (3.7%) together account for ~10% of the total releases. However, it should be noted that from the Asian and the African regions, 25 and 26 inventories have been reported, respectively, whereas from the other regions, the number of available inventories was lower (CEE = 13, GRULAC = 12, WEOG = 10). Further, the majority of the people "covered" by these inventories were from Asia (3.34 billion people or 71% of the population covered by these inventories), whereas from Africa, a population of "only" 560 million (0.56 billion or 12% of the population covered by these inventories) is included (Table 4).

Table 3 Summary of PCDD/PCDF releases per vector and per UN region (based on 86 reports)

UN region	No	Total (g TEQ yr^{-1})	Air (g TEQ yr^{-1})	Water (g TEQ yr^{-1})	Land (g TEQ yr^{-1})	Product (g TEQ yr^{-1})	Residue (g TEQ yr^{-1})
Africa	26	21,122	11,425	111	3,670	205	5,712
Asia	25	34,659	14,165	266	1,888	4,929	13,411
CEE	13	4,559	2,739	858	39	50	873
GRULAC	12	7,869	3,941	56	819	529	2,524
WEOG	10	2,610	1,190	5.8	1,330	8.8	75
Grand total	86	70,819	33,459	1,296	7,746	5,722	22,595
		100%	47%	2%	11%	8%	32%

Table 4 Number of countries reporting PCDD/PCDF releases per region and population covered (based on 86 reports)

Row labels	Number of inventories	Population covered by region in million inhabitants
Africa	26	560
Asia	25	3,344
CEE	13	212
GRULAC	12	383
WEOG	10	237
Grand total	86	4,736

Table 5 Summary of country statistic data used for assessment (based on 86 reports)

	Population (million)	Area (km^2)	GNI PPP $capita^{-1}$ (international dollar)
Minimum	0.0022	260	200
Maximum	1,296	17,098,240	43,740
Average	55	999,458	9,107
Total of all 86 countries	4,736	85,953,345	

Table 6 Summary of PCDD/PCDF releases per denominator and year (based on 86 reports)

	Total releases per denominator and year			Releases to air per denominator and year		
	mg TEQ $capita^{-1}$	mg TEQ km^{-2}	mg TEQ 1000 USD GNI PPP^{-1}	mg TEQ $capita^{-1}$	mg TEQ km^{-2}	mg TEQ 1,000 USD GNI PPP^{-1}
Minimum	0.87	18	0.032	0.20	17	0.006
Maximum	256	22,917	3,560	178	9,603	2,315
Average	39	3,004	340	20	1,480	169

The assessed countries exhibit a large range of characteristics with respect to population, area of the country, or gross national income per capita (Table 5). For example, the population of countries ranges over six orders of magnitude and has the smallest population in Niue (2,200 people in the year 2004) and the largest population in the People's Republic of China (1,30 billion in 2007). With an area of 260 km^2, Niue is also the smallest country; the largest country is the Russian Federation with an area of more than 17 million square kilometer. The poorest country within this dataset and according to the World Bank's statistics expressed as PPP is Liberia with a gross national income (GNI) of 200 international dollar per capita in 2004 and the economically strongest country is Brunei Darussalam with 43,740 international dollar in 2001, followed by Norway with 42,550 international dollar in 2004; the average GNI PPP per capita is 9,107 international dollar (for the countries included in this assessment and the corresponding reference year).

The minimum, maximum, and average releases of PCDD/PCDF for all release vectors ($PCDD/PCDF_{total}$) and to air are summarized in Table 6. The average total release per year (39 mg TEQ yr^{-1}) is about twice as high as the release to air only (20 mg TEQ yr^{-1}) underpinning the importance of the vector to air (paired with the fact that some countries estimated releases to air only; see Fig. 2).

According to income category following the World Bank classification (http://data.worldbank.org/about/country-classifications; economies are divided according to 2012 GNI per capita, calculated using the World Bank Atlas method), the number of countries reporting PCDD/PCDF releases within each income category

Table 7 Overview of annual releases to five release vectors according to income category (86 inventories)

Income category	# Countries	Air	Water	Land	Product	Residue	Total
		(g TEQ yr^{-1})					
High income	22	5,238	14	1,511	177	1,341	8,281
Upper middle income	31	11,873	217	1,801	1,223	9,933	25,043
Lower middle income	28	15,545	1,060	4,261	4,287	10,993	36,147
Low income	5	803	5	174	34	328	1,344
Total	86	33,459	1,296	7,746	5,722	22,595	70,819

varies: The annual PCDD/PCDF releases per release vector within each income category (L, LM, UM, H) are shown in Table 7. The highest number of countries belong to the economic group of upper middle income ($n = 31$), followed by the lower middle income ($n = 28$); the two extreme income groups have the lowest representation with five and 22 countries in the low-income and the high-income categories, respectively.

Annual PCDD/PCDF release of the 86 countries/regions ranged from 0.56 (Niue, UM) to 10,237 g TEQ yr^{-1} (China, UM) (Tables 1 and 8). The annual releases of the five largest emitters were from five upper middle-income countries (China = 10,238 g TEQ yr^{-1}; India = 8,658 g TEQ yr^{-1}; Indonesia = 7,352 g TEQ yr^{-1}; Nigeria = 5,340 g TEQ yr^{-1}; and Kenya = 4,738 g TEQ yr^{-1}), whereas the smallest releases were from two upper middle-income countries (Niue = 0.6 g TEQ yr^{-1} and Palau = 2.25 g TEQ yr^{-1}), one lower middle-income country (Samoa = 1.4 g TEQ yr^{-1}), and two high-income countries (Brunei Darussalam = 1.4 g TEQ yr^{-1} and Seychelles = 5.4 g TEQ yr^{-1}). The top five emitters accounted for 51% of the total releases from all available inventories (86 inventories), demonstrating that population size and economic status have a positive correlation toward PCDD/PCDF releases (see also Pulles et al. [29] and Ren and Zheng [30] for smaller datasets).

Graphical sketches of releases according to economic status are shown in Figs. 3, 4, and 5 for total releases and releases to air ($PCDD/PCDF_{total}$ and $PCDD/PCDF_{air}$). Whereas the normalization to area as mg TEQ per km^2 (Fig. 4) results in a scattered picture, using population-based releases, it can be seen that high-income countries (H) have the lowest PCDD/PCDF releases per capita. Not much difference was found between the other income categories (Fig. 3). Normalization to GNI PPP^{-1} shows that lowest releases are found in higher-income countries (H and UM), whereas the LM and L countries exhibit higher releases (Fig. 5).

Table 8 Summary of total releases and releases to air according to income category (and reference year) (86 inventories)

Income group	# Countries	Sum air (g TEQ yr^{-1})	Sum total (g TEQ yr^{-1})	Pop. *mio.	Land area (km^2)	Average total (mg TEQ $capita^{-1}$)	Average air (mg TEQ $capita^{-1}$)
High income	**22**	**5,238**	**8,281**	**521**	**38,685,052**	**19**	**11**
Asia: BRN, CHN-HKG, SYC, TUR	4	1,256	2,190	76	790,882	26	17
CEE: EST, HRV, LTU, RUS, SVN	5	1,958	2,069	153	17,285,580	21	13
GRULAC: CHL, CUB, VEN	3	833	1,411	55	1,778,030	22	13
WEOG: AUS, AUT, CAN, CHE, DEU, FIN, FRA, NOR, NZL, PRT	10	1,190	2,610	237	18,830,560	15	6.3
Upper middle income	**31**	**11,873**	**25,045**	**1,934**	**29,645,964**	**43**	**23**
Africa: MAR, GAB, MUS, SWZ, TUN, ZAF	6	1,217	3,528	90	2,116,324	57	32
Asia: AZE, CHN, FJI, IRN, JOR, LBN, NIU, PLW, SYR, THA, WSM	11	6,977	13,897	1,470	12,251,099	50	26
CEE: ALB, BGR, BLR, MKD, ROU, SRB	6	762	1,662	52	697,450	43	26
GRULAC: ARG, BRA, COL, ECU, PAN, PER, PRY, URY	8	2,917	5,960	322	14,581,550	23	11

(continued)

Table 8 (continued)

Income group	# Countries	Sum air (g TEQ yr^{-1})	Sum total (g TEQ yr^{-1})	Pop. *mio.	Land area (km^2)	Average total (mg TEQ $capita^{-1}$)	Average air (mg TEQ $capita^{-1}$)
Lower middle income	**28**	**15,357**	**35,661**	**2,026**	**15,256,910**	**50**	**23**
Africa: BEN, BFA, CIV, CMR, CPV, DJI, GHA, GMB, KEN, MLI, NGA, SDN, TGO, TZA, ZMB	15	9,404	16,250	359	8,468,650	60	36
Asia: BGD, IDN, IND, KHM, LAO, LKA, NPL, PHL, TJK, VNM	10	5,932	18,572	1,798	6,741,880	17	6.7
CEE: ARM, MDA	2	19	828	6.7	63,580	115	2.7
GRULAC: NIC	1	191	498	5.4	130,370	93	36
Low income	**5**	**803**	**1,344**	**111**	**2,217,390**	**35**	**22**
Africa: BDI, ETH, LBR, MDG, ZWE	5	803	1,344	111	2,217,390	35	22
Total	86	33,459	70,819	4,736	85,953,345	39	20

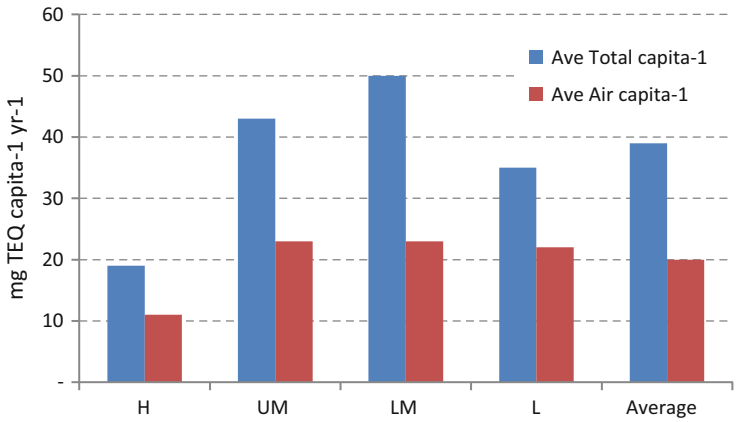


Fig. 3 Average annual releases of PCDD/PCDF per capita and year (mg TEQ $capita^{-1}$ yr^{-1})

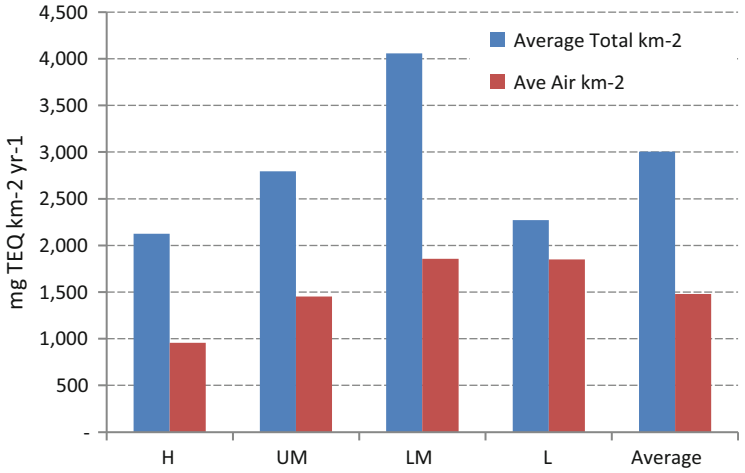


Fig. 4 Average annual releases of PCDD/PCDF per square kilometer and year (mg TEQ km^{-2} yr^{-1})

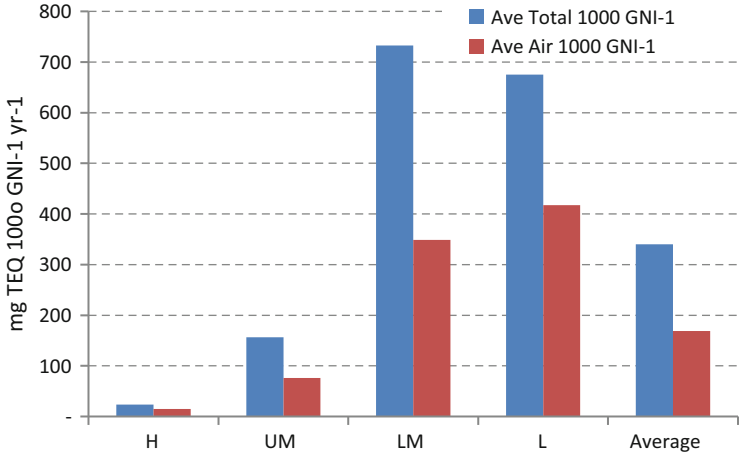


Fig. 5 Average annual releases of PCDD/PCDF per gross national income per capita purchase parity and year (mg TEQ 1,000 USD GNI PPP^{-1} yr^{-1})

4.3 Quantitative Inventory Results Using Source Groups

4.3.1 Releases According to Ten Source Groups

When applying the Toolkit methodology, releases of PCDD/PCDF cannot only be assessed according to the five release vectors but also according to the ten source groups (SG). Of these, nine are quantitative, whereas the tenth source group represents hot spots, which cannot be assigned to a reference year. For the two most important release parameters – total releases and releases to air – the statistics are as shown in Tables 9 and 10 and in Fig. 6, respectively. For both releases, the

Table 9 Descriptive statistics for releases PCDD/PCDF$_{total}$ by source groups

	Total_SG1	Total_SG2	Total_SG3	Total_SG4	Total_SG5	Total_SG6	Total_SG7	Total_SG8	Total_SG9	Total_SG10
Mean (%)	13	11	12	1	2	45	5	1	8	0
Median (%)	6	2	5	0	0	43	0	0	1	0
Std error (%)	2	2	2	0	1	4	1	0	2	0
Std dev. (%)	19	18	18	3	9	33	11	2	15	1
25th percentile (%)	1	0	1	0	0	11	0	0	0	0
75th percentile (%)	17	13	15	1	1	77	4	0	10	0
Minimum (%)	0	0	0	0	0	0	0	0	0	0
Maximum (%)	89	85	80	14	79	100	60	14	97	7
Count	86	86	86	86	86	86	86	86	86	86

Table 10 Descriptive statistics for releases PCDD/PCDF$_{Air}$ by source groups

	Air_SG1	Air_SG2	Air_SG3	Air_SG4	Air_SG5	Air_SG6	Air_SG7	Air_SG8	Air_SG9	Air_SG10
Mean (%)	17	9	15	2	3	51	1	1	0	0
Median (%)	9	3	7	1	1	56	0	0	0	0
Std error (%)	2	2	2	0	1	4	1	0	0	0
Std dev. (%)	23	15	20	4	10	33	5	3	0	0
25th percentile (%)	1	0	2	0	0	24	0	0	0	0
75th percentile (%)	22	10	21	3	2	80	0	1	0	0
Minimum (%)	0	0	0	0	0	0	0	0	0	0
Maximum (%)	100	70	91	20	89	100	45	14	1	0
Count	85	85	85	85	85	85	85	85	85	85

Note: Armenia reported total releases only; therefore, this one country cannot be included here; total of 85 reports

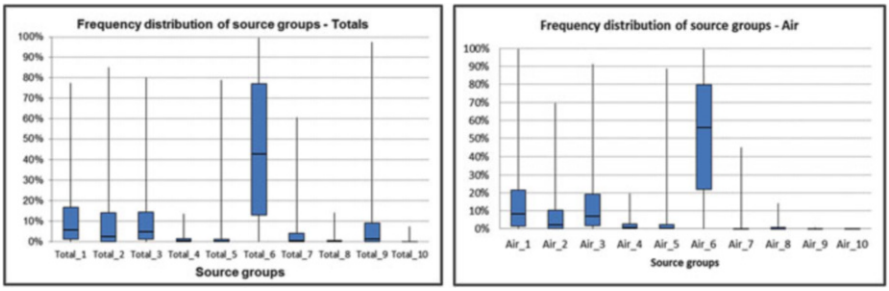


Fig. 6 Frequency distribution of source groups according to country. Source groups are as follows: SG1 = waste incineration, SG2 = metal production, SG3 = power and heat, SG4 = mineral production, SG5 = transport, SG6 = open burning, SG7 = industry, SG8 = miscellaneous, SG9 = disposal, SG10 = hot spots

source group 6, corresponding to open burning processes, is by far the largest source group with a 75th percentile of 77% and 80%, respectively. For total releases, the second and third largest source groups are waste incineration (SG1 = 17%) and metal production and heat and power (SG2 and SG3; both with 14%). For air releases, the second and third largest contributors are waste incineration (SG1 = 22%) and heat and power (SG3 = 19%).

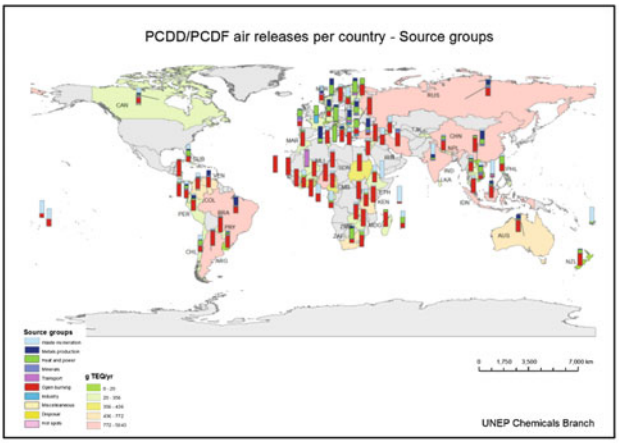
The releases to air, expressed as g TEQ per year according to income categories by source group, are displayed in Table 11. As can be seen, the highest contribution to all air releases was from SG6, open burning processes, such as open burning in agriculture and forests and waste, occurring in lower middle-income countries (>10,300 g TEQ yr^{-1}). Interestingly, SG6 had the highest releases for all income categories. For the high- and upper higher-income groups, the second most important source group is SG2, the production of ferrous and nonferrous metals (relatively biggest emitter SG6). Notably, the industrial process for the production of consumer goods (SG7) is the source group with the lowest releases to air (174 g TEQ yr^{-1}).

Graphical sketches of the distribution of source groups to total and air releases of PCDD/PCDF (g TEQ yr^{-1}) are shown in Fig. 7.

Table 11 Sums of air emissions by source group and income category (g TEQ yr^{-1})

Income group	ΣSG1	ΣSG2	ΣSG3	ΣSG4	ΣSG5	ΣSG6	ΣSG7	ΣSG8	ΣSG9	Subtotals
High	345	1,541	719	553	101	1,833	125	23	-	5,238
Upper middle	1,263	3,417	2,129	708	214	3,983	44	116	0.2	11,873
Lower middle	2,833	1,000	949	243	98	10,366	5	45	1.0	15,545
Low	60	0.2	33	0.4	1.0	708	0	0.1	0.4	803
Total	4,501	5,958	3,829	1,504	413	16,889	174	183	1.6	33,459

Numbers may not add up at last digit due to rounding procedure



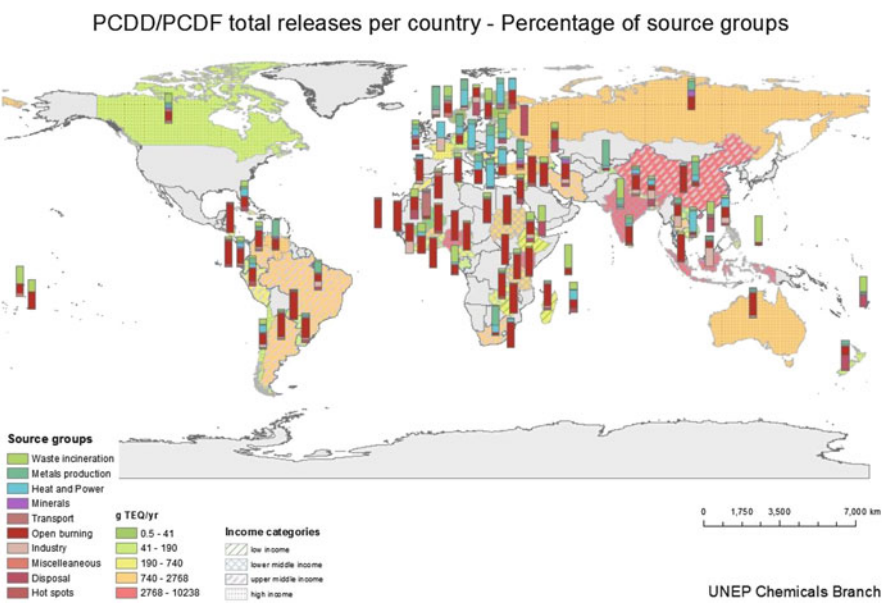


Fig. 7 (continued)

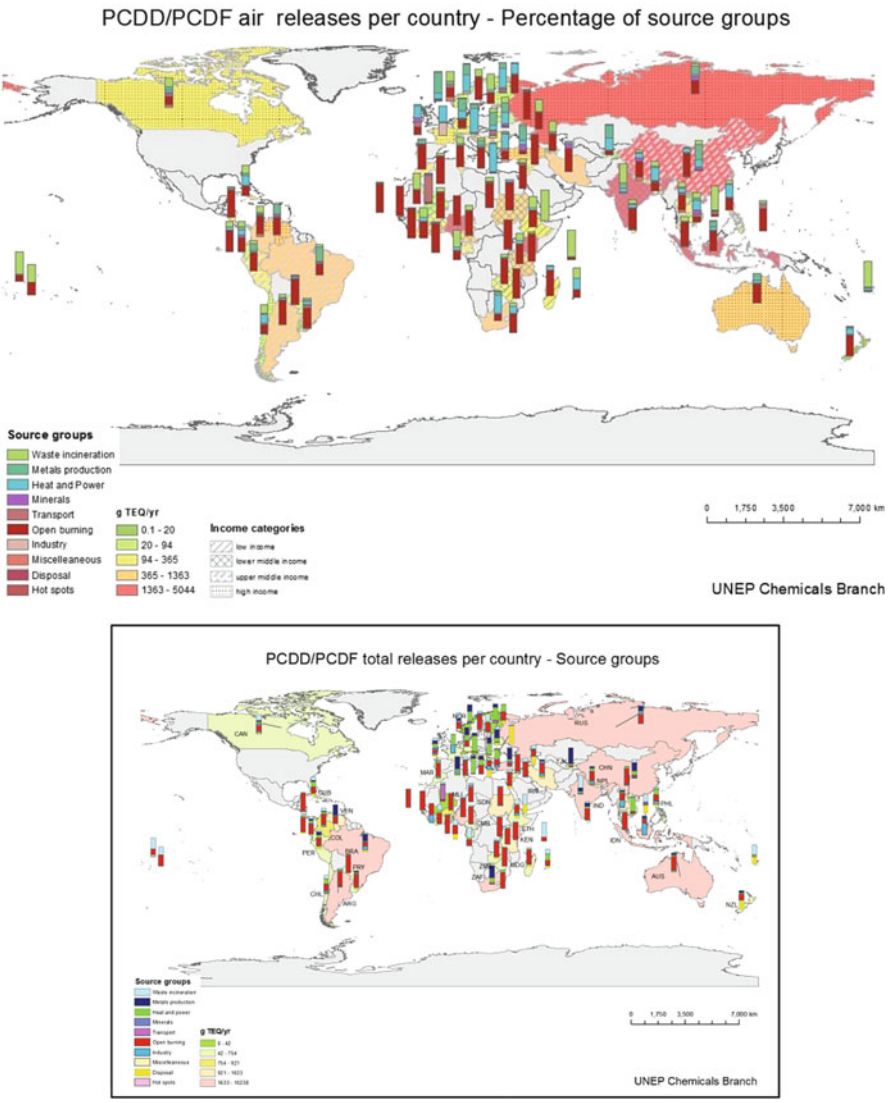


Fig. 7 Presentation of total annual releases and releases to air per source group and income category based on total and air releases, respectively (g TEQ yr^{-1}). For countries colored in *grey*, no information has been provided so far; they are not included in Table 1

5 Conclusions

The development and periodic updating of national release inventories for unintentional POPs is an obligation for countries that have ratified the Stockholm Convention. The Conference of the Parties had endorsed a reporting format according to Article 15 of the Convention where countries report their national inventories for ten source groups and five release vectors. According to schedule in the legally binding instrument, these inventories should be revised and updated every five years. In order to report national inventories in a transparent, complete, and comparable manner, the Toolkit has been developed by UNEP and is being revised and updated periodically. The first round of reporting took longer than anticipated and still does not yet include all parties (86 reported whereas the Convention has 179 parties; status: June 2015). Nevertheless, an abundance of information has been generated from these inventories including political and technical results.

At policy level and for practical reasons, it is recommended that inventory activities be focused on PCDD/PCDF only, as these substances are indicative of the presence of other unintentional POPs (HCB, PCB and PeCBz according to Annex C of the Stockholm Convention). It is also recommended to use the TEQ approach. PCDD/PCDF are considered to constitute a sufficient basis for identifying and prioritizing sources of all these substances as well as for devising applicable control measures for all Annex C POPs and for evaluating their efficacy. Only in the context of research or other projects it is advisable to analyze emissions of all unintentional POPs listed in Annex C in order to produce useful information for the purpose of deriving emission factors [11].

At technical level, across all inventories, the most important release vector is air, receiving 47% of all PCDD/PCDF releases; the second most important vector is residue with 32%. Of the other releases vectors, water does not play a role (receiving only 2% of all PCDD/PCDF releases); releases to land or product account for 11% and 8%, respectively. Among the source groups, SG6 (open burning) constitutes the largest emitter for PCDD/PCDF$_{total}$ (mean = 45%) and for PCDD/PCDF$_{air}$ (mean = 51%), followed by SG1 (waste incineration) with 13% for PCDD/PCDF$_{total}$ and 17% for PCDD/PCDF$_{air}$. The production of minerals (SG4), transport (SG5), industry (SG7, chemical industry, pulp and paper, textile, etc.), and the source group of miscellaneous (SG8, including crematoria, tobacco smoking) do not contribute much to PCDD/PCDF release inventories.

High-income countries tend to have lowest average releases of PCDD/PCDF$_{total}$ and PCDD/PCDF$_{air}$ per capita (19 mg TEQ capita^{-1} yr^{-1} and 11 mg TEQ capita^{-1} yr^{-1}, resp.), whereas the lower middle-income countries have highest releases for PCDD/PCDF$_{total}$ and PCDD/PCDF$_{air}$ (50 mg TEQ capita^{-1} yr^{-1} and 23 mg TEQ capita^{-1} yr^{-1}, resp.; see Table 8). In the high- and upper middle-income countries, metal recycling processes (SG2) are the second highest emitters, numerically very close to the SG6, the largest emitter.

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For this review, I thank colleagues and coworkers, government representatives, students, and interns for assistance in the development of the Toolkit methodology and the wider discussions on pollutants' inventories. Special thanks go to Ms. Haosong Jiao for the preparation of the maps. With the present status, we have come a long way but remain curious and willing to improve further.

Glossary

Box and Whisker plot Graphically summarises the median, a measure of dispersion (first and third quartile) and the range for a sample or population.

GNI PPP Is gross national income (GNI) converted to international dollars using purchasing power parity rates. An international dollar has the same purchasing power over GNI as a U.S. dollar has in the United States. Gross national income is the sum of value added by all resident producers plus any product taxes (less subsidies) not included in the valuation of output plus net receipts of primary income (compensation of employees and property income) from abroad. PPP GNI was named formerly PPP GNP.

Income categories As defined by the World Bank; they are as follows: Low income (L), lower middle income (LM), upper middle income (UM), and high income (H).

ISO codes ISO 3166 is the International Standard for country codes and codes for their subdivisions. The purpose of ISO 3166 is to define internationally recognised codes of letters and/or numbers that we can use when we refer to countries and subdivisions. The ISO country codes are maintained by the ISO 3166 Maintenance Agency, Geneva, Switzerland, www.iso.org/iso/country_codes.

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