· Article ·
Advances in Polar Science
doi: 10.13679/j.advps.2021.0044
June 2022 Vol. 33 No. 2: 156-166

Inventory of unintentional POPs emission from anthropogenic sources in Antarctica

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Received 16 August 2021; accepted 28 January 2022; published online 30 May 2022

Abstract In spite of remote location and very limited human activities, Antarctica is affected by persistent organic pollutants (POPs). POPs investigation in Antarctica has a comparatively long history, but there are still large knowledge gaps in assessment of their emission into environment. In the paper the results of the first inventory of unintentional POPs emission from anthropogenic sources in Antarctica for modern period and preliminary estimate for the late 1980s are presented. Assessment of dioxin/furans (PCDD/Fs) emission in different media, as well as polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) in air is based on methodology of emission factors and indicators of human activity. The following sources of POPs emission have been estimated: power generation and heating, waste incineration, mobile sources and open burning of waste (in the past). According to the data obtained, annual PCDD/Fs air emission for modern period comprises 60.74 mg toxic equivalent (TEQ), PCBs – 5.09 mg TEQ, and HCB – 457.6 mg. Additionally 2.5 mg TEQ of dioxin/furans is released to residues, so total PCDD/Fs emission is amounted 63.23 mg TEQ. Waste incineration makes the greatest contribution to POPs emission (96% of PCDD/Fs, 98% of PCBs and 36% of HCB air emission). In late 1980s open burning of waste was the major source of POPs. Retrospective assessment shows that over a 30-year period air emissions of PCDD/Fs decreased about 13 times, PCBs—15 times and HCB—57 times, primarily due to the prohibition of open burning of waste in compliance with the Protocol on Environmental Protection to the Antarctic Treaty requirements.

Highlights

- Waste incineration makes the greatest contribution to total POPs emission in Antarctica for modern period.
- Open burning of waste was the main anthropogenic source of POPs emission in Antarctica in late 1980s.
- Over a 30-year period air emissions of dioxins/furans, PCBs and HCB have decreased dramatically.

Keywords persistent organic pollutants, emission inventory, PCDD/Fs, PCBs, HCB

Citation: Kakareka S, Kukharchyk T. Inventory of unintentional POPs emission from anthropogenic sources in Antarctica. Adv Polar Sci, 2022, 33(2): 156-166, doi: 10.13679/j.advps.2021.0044

1 Introduction

Persistent organic pollutants (POPs) are a group of toxic chemical substances which are considered a global issue and are regulated by Stockholm Convention on POPs (Secretariat of the Stockholm Convention, 2020). Long range transport of POPs to polar regions via atmosphere is caused by ability of their volatilized fractions to undergo global fractionation along latitudinal temperature gradients (Wania and Mackay, 1996; Bengtson Nash, 2011; Grannas et al., 2013). There are also other pathways of POPs redistribution including oceans and birds (UNEP, 2002; Bargagli, 2005; Luek et al., 2017).

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In spite of its remote location and very limited human activities, Antarctica is affected by POPs which were detected in 1970s for the first time (Risebrough et al., 1976). Up to now POPs were recorded in different Antarctic media including snow (Vecchiato et al., 2015), atmospheric air (Lugar, 1993; Ockenden et al., 2001; Bargagli, 2005; Pozo et al., 2014; Bengtson Nash, 2017; Hao et al., 2019), soil and lake sediments (Negoita et al., 2003; Klánová et al., 2008; Wang et al., 2012), lichens and mosses (Focardi et al., 1991; Borghini et al., 2005), and marine environment (Tanabe et al., 1983; Lenihan, 1992; Kumar et al., 2002; Negri et al., 2006; Corsolini et al., 2011; Ko et al., 2018; Krasnobaev et al., 2020, etc). The above mentioned list of references is not exhaustive; it only confirms POPs contamination in Antarctica. This problem is also well demonstrated in some reviews (Bargagli, 2005; UNEP, 2002; SCAR, 2009, 2021; Bacaloni et al., 2011).

Regarding the sources of POPs in Antarctica, it should be noted that transboundary transport of these pollutants has been confirmed to be the most important for this polar region (Lohmann et al., 2001; SCAR, 2009; Li et al., 2012; Kallenborn et al., 2013; Piazza et al., 2013). However the influence of local anthropogenic sources of legacy and novel POPs within Antarctic scientific stations has been also reported (Risebrough et al., 1990; Lenihan, 1992; Lugar, 1993; Lugar et al., 1996; Kennicutt II et al., 2010; Wild et al., 2015; Bengtson Nash et al., 2021). Actually, the influence of both factors (long-range transport and local sources) is relevant for PCDD/Fs and PCBs. In contrast, the influence of mostly long-range transport is characteristic for chlorinated pesticides, while local sources–for polybrominated diphenyl ethers (Choi et al., 2008a; Bacaloni et al., 2011; Vecchiato et al., 2015). According to Bengtson Nash (2011) and Ko et al. (2018), the content of HCB in Antarctic environment is also caused by long range transport because it is usually considered as a consequence of pesticides application outside of Antarctica.

It is known that anthropogenic activity in Antarctica is connected with scientific station operation and maintenance, delivery of expeditions by sea and air, as well as tourism and fishing.

Unintentional POPs formation (first of all PCDD/Fs, PCBs and HCB) at polar region like in other regions with similar activities is possible during fuel combustion by stationary and mobile sources as well as waste incineration (Dioxin and Furan Inventories, 1999; Toolkit, 2013). For example, the measuring of PCDD/Fs at McMurdo Station in 1992–1994 showed that waste incinerator, power plants, vehicles and heating furnaces can contribute PCDD/Fs compounds to the air (Lugar et al., 1996).

Despite of long history of POPs investigation in Antarctica, no data on assessment of POPs emission into environment as well as their concentrations in flue gases or in residues has been revealed. There is lack of inventory and reporting on pollutants emission from anthropogenic sources at scientific stations. According to the recent horizon scan of priority challenges of POPs in Antarctica, local sources and emissions have been identified among the priority research to cover the gaps and inform the policy making in the framework of Madrid Protocol (SCAR, 2021).

The main purpose of this study is to assess PCDD/Fs, PCBs and HCB releases into environment from stationary and mobile ground-based sources in Antarctica for modern period (pre-Covid-19). In order to determine emission trends after the signature of the Protocol on Environmental Protection to the Antarctic Treaty, draft assessment of POPs emission for the late 1980s was also prepared. The following tasks were solved: identification of POPs emission sources in Antarctica; data collection of human activities characteristics; preparation of input activity data in required format; and selection of specific emission factors.

It should be noted that analysis of Covid-19 impact onto POPs emission sources in Antarctica was beyond the scope of the paper.

2 Methods and data

2.1 General methodology

At the preliminary stage of study, the main sources of POPs releases in Antarctica were identified. As it has been already mentioned, anthropogenic activity in Antarctica is very limited and mostly connected with scientific station operation and maintenance, so the list of POPs emission sources is comparatively short. Taking into account the international and national experience of unintentional POPs inventory (Dioxin and Furan Inventories, 1999; Toolkit, 2013), fuel combustion by diesel generators and boilers, motor vehicles as well as waste incineration were considered as modern sources of POPs emission for numerical evaluation. Shipping and aviation are not significant sources of POPs emissions at regional and national scale (Dioxin and Furan Inventories, 1999) and are rarely mentioned in national inventories (Secretariat of the Stockholm Convention, 2021); the corresponding sections are missing in Toolkit 2013. We took into account these sources to provide very preliminary estimates of POPs emission and the results were not included in total emission.

General methodology of POPs emission assessment used for this study is based on activity data and emission factors. Main emission calculation formula is as follows:

$$P_{i,n} = \sum V_{m,n} \times F_{i,m} \times 1000, \quad (1)$$

where $P_{i,n}$ – emission of substance i in the activity sector n, mg; $V_{m,n}$ – fuel (waste) m combustion in the activity sector n, TJ (t); $F_{i,m}$ – the specific emission factor of substance i from combustion of fuel (waste) m, $\mu g \cdot TJ^{-1}$ (t^{-1}).

Emissions of PCDD/Fs in air, residues and land, as well as PCBs and HCB in air from anthropogenic sources in Antarctica have been calculated. PCDD/Fs and PCBs emissions were expressed in mg toxic equivalent (mg TEQ), HCB - in mg.

2.2 Approaches for characterization of activity levels

Antarctica has a specific legislative status. Operations on its territory are regulated by the Antarctic Treaty (1959) and Protocol on Environmental Protection to the Antarctic Treaty (1991). Thirty countries from all over the world operate more than 70 year-round and seasonal stations in Antarctica, as well as several dozens of other facilities (airfields, depots, refugees, camps, etc.). According to the data of the Council of Managers of National Antarctic Programs (COMNAP) (2017), winter population in Antarctica amounted to about 1000 people while summer population – about 5000. Due to Antarctica's specific status there is no joint system of statistics collection (possibly, except for tourism). Therefore, there is no aggregated human activity data per sector for the whole continent, which is necessary as input data in POPs emission calculation. That is why a combination of approaches was applied to derive activity rates for Antarctica. For modern period available records on stations and national Antarctic programs were used including volumes of fuel and wastes burned, capacity of power installations, number of vehicles and stations population. For this purpose the following sources of information were taken into consideration: annual reports of the Parties to the Antarctic Treaty and Protocol on Environmental Protection (Secretariat of the Antarctic Treaty, 2021a, 2021b), results of inspections according to the Article VII of the Antarctic Treaty and Article 14 of the Protocol on Environmental Protection (Secretariat of the Antarctic Treaty, 2021d), initial and comprehensive environmental evaluation (Secretariat of the Antarctic Treaty, 2021c) and data of COMNAP (COMNAP, 2006, 2017). Information from scientific publications (Savatyugin, 2001, 2009; Potter, 2003; O'Brien et al., 2004; Bharti et al., 2015, etc.) and reports (NSF, 2019, etc.) was also collected.

Based on these resources volume of total fuel combustion, the share of fuel consumption by energy production, heating systems and ground vehicles as well as volume of waste incineration for modern period were obtained.

For the late 1980s previous estimates of fuel consumption in Antarctica by stationary and land-based mobile sources according to Boutron and Wolff (1989) were used. The total volume of diesel fuel (2.3×10^7 m^3) was distributed among energy production, heating and ground vehicles using modern share of these sources in total fuel consumption. Estimation of the volume of waste burning was made taking into account published data (NSF, 1991; Crockett and White, 1997; O'Brien et al., 2004).

All types of data were analyzed and prepared in required format; some assumptions were made in case of data absence or incompleteness (some details are shown below).

2.3 Retrieving input information for activity sectors

2.3.1 Energy production

In spite of introduction of renewable sources of energy like solar panels and wind generators, diesel generators are still the main source of electricity and heat in Antarctica and consume the main share of fuel. According to a recent assessment, total installed diesel generator capacity is 36400 kW, the average load is 10510 kW. Details of this study as well as assessment of fuel consumption in energy production are presented in Kakareka (2020). In the above mentioned assessment volumes of fuel used in heating systems were included in energy production; whereas for current estimates consumption of fuel in these sectors was separated.

2.3.2 Autonomous heating systems

Fuel-burning installations, such as boilers, stoves, etc. are often used especially at large Antarctic stations for heating and sometimes for water production. They usually use the same fuel as diesel power plants. Data on the use of heating systems was found for 16 stations including McMurdo, Palmer, Marambio, Syowa, Scott Base and some others. At another 28 stations such systems are most likely to be present as well. At eight stations there are definitely no such systems or they consume negligible fraction of all fuel. At the rest stations such systems are most likely to be absent.

Fuel consumption data for heating systems is available for McMurdo, Scott Base and Rothera stations. Based on these records as well as other available information the share of heating systems in the total fuel consumption for energy purposes (stationary installations) for those stations where they are available or their presence is very likely to be determined, equals 20%.

2.3.3 Waste incineration

Evidence of waste incinerators modern use at 29 Antarctic stations and high probability of waste incineration at 17 other stations was collected. In particular waste incineration is carried out at the stations of Argentina (Marambio, Carlini), Chile (Esperanza), Russian Federation (Novolazarevskaya, Progress, Bellingshausen), Australia (Davis, Casey, Mawson), United Kingdom (Rothera, Halley VI), Japan (Syowa), Korea (King Sejong), France (Dumont D'Urville) and some other countries. Some examples of incineration installations are the following: incinerators IN-50.02 of 20–40 $kg\cdot h^{-1}$ capacity are used at Progress II and Novolazarevskaya stations, incinerator SP-50 (designed for ships) of 50 $kg\cdot h^{-1}$ capacity is used at Molodezhnaya Station (Savatyugin, 2001; Report of the Russian Federation, 2006). A small wood-burning stove for waste incineration operates at Palmer Station (Khan et al., 2019). Specially designed incinerators have been made by Todaysure Matthews and adapted to operate in sub-zero temperatures at Rothera and Halley stations (Shofield, 2015). At some stations electric heat is used for incineration of human wastes (Incinolets).

For 16 stations including McMurdo, Amundsen-Scott, Troll, Neumayer III, Jang Bogo and some others, there is information that wastes are currently not incinerated. Most likely wastes are also not incinerated at 35 other stations and field bases in Antarctica.

Waste incineration volumes are available for five stations (Dumont D'Urville, Maitri, Syowa, Progress, Novolazarevskaya) (COMNAP, 2006). Due to lack of data sets on incineration volumes, the ratio of waste incineration was calculated for the maximum population at the station: Incineration volumes at the above mentioned five stations equal from 3.32 to 8 t; peak population at these stations —from 50 to 170 people, therefore the amount of annual wastes incineration varies from 31 to 95 $kg{\cdot}person^{-1}$, or equals 66 $kg{\cdot}person^{-1}$ in average. Based on this specific value the total volume of waste incineration in Antarctica is estimated 167 $t{\cdot}a^{-1}$.

2.3.4 Mobile sources

A wide variety of ground-based motor vehicles is used in Antarctica ranging from snowmobiles to heavy tractors and special vehicles. Fuel consumption data for mobile equipment is very sketchy. More or less complete data is available for McMurdo, Scott Base and Australian stations. Based on these records it was difficult to derive a unified indicator, for example fuel consumption per capita. Therefore, grouping of stations was done as a first step. Stations were split into five groups according to the share of fuel consumption by motor vehicles to fuel consumption in energy production.

(1) Largest station with a lot of vehicles (McMurdo). Thus, in 2019 fuel consumption by transport and other equipment at McMurdo amounted to $1.735{\times}10^6$ L (NSF, 2019) which is 30%–35% of annual fuel consumption in energy production;

(2) Stations with an average number of transport units which are the basis for resupply traverses; the share of fuel consumption by transport according to approximate estimates makes up to 20%–30% of fuel consumption in energy production;

(3) Continental stations (East Antarctica): 10%;

(4) Large and medium-sized stations in West Antarctica: 5%;

(5) Small stations in West Antarctica: 1%.

Diesel predominates in the composition of motor fuels. Gasoline is used at most stations for snowmobiles, motor boats and less often for road vehicles. Based on the data available, it was assumed that leaded gasoline is not currently used by ground vehicles.

2.3.5 Open burning of waste

Open burning of waste was a widespread practice of waste management in Antarctica prior to the signing of the Environmental Protocol to the Antarctic Treaty. Waste problem was often solved quite simply: what could burn—was burned in an open way, the rest was buried in the snow or was dumped into the sea (O'Brien et al., 2004), though incinerators were installed at some stations. The practice of open burning of waste at certain Antarctic stations in the past is well described in literature (Lenihan, 1992; Crockett and White, 2003; Potter, 2003; Bargagli, 2008; Kennicutt II et al., 2010; Fryirs et al., 2013). For example, according to Crockett and White (1997), in 1956–1980 trash from McMurdo Station was piled on the steep shoreline of Winter Quarters Bay Landfill, doused with fuel and ignited.

However, quantitative estimates of the volume of waste burning across Antarctica are very rare. Thus, according to emission estimates for this sector made by Boutron and Wolff (1989), the amount of burned waste in Antarctica can be derived—it is about 750 $t{\cdot}a^{-1}$. According to NSF (1991), in late 1980s 512 t of waste (464.5 metric t) and 75 thousand gallons (2.839×10^5 L) of waste oil were burned at McMurdo Station annually. The maximum population of McMurdo at that time was 1073 people, thus the volume of burned waste was approximately 433 $kg{\cdot}capita^{-1}$. With the maximum population of Antarctica in 1987 (3400 people) (Boutron and Wolff, 1989), the volume of waste burned in the open and in incinerators amounts to 1472 t. Assuming that approximately 25% of all fuel was burned at McMurdo, the volume of waste oil incineration in Antarctica during this period will be 1.1356×10^6 L; assuming the density of waste oil products of 0.96, this equals 1090 t.

It is accepted that open burning of waste is not currently used, although there is occasional evidence to the contrary (Peter et al., 2013).

2.4 Emission factors

As it was mentioned above, POPs have not been measured in waste gases in Antarctica. Therefore possibilities to justify the selection of emission factors were very limited. In this study emission factors given in the Toolkit for Identification and Quantification of Releases of Dioxins, Furans and other unintentional POPs under Article 5 of the Stockholm Convention (Toolkit, 2013) and additions to it (Toolkit PCDD/PCDF, 2013; Toolkit HCB, 2019; Toolkit PCB, 2019) were used. These guidance documents were prepared specifically for obtaining harmonized POPs emission inventories across the world. Other international guidelines (AP-42, 1995; European Environment Agency, 2019), as well as national inventories (An Inventory of Sources, 2006; Secretariat of the Stockholm Convention, 2021) and scientific publications (Quaß et al., 2004; Gullet et al., 2010; Black et al., 2012; Gong et al., 2017; Zhang et al., 2017) were also taken in account.

It should be noted that data on POPs emission factors for liquid fuel combustion for vehicles and machinery are very limited, thus no large variability of available emission factors can be expected.

In regard to waste incineration, it was assumed that waste is separated before incineration and no items, which are prohibited by the Annex III of the Protocol to

Environmental Protection to the Antarctic Treaty and national legislation, are incinerated. Waste of different composition is incinerated in small-scale incinerators which are usually not equipped with flue gases control. Only a few examples of abatement equipment are known: at Dumont D'Urville Station (a ceramic diesel particulate filter), Zhongshan Station (a cyclonic filtering system), and Commandante Ferraz Station (without specification) (COMNAP, 2006; Le Calvez, 2006; Secretariat of the Antarctic Treaty, 2021d). No data on implementation of special measures to reduce POPs emissions has been found.

Incinerated wastes in Antarctica usually include wood, paper, cardboard, kitchen waste, poultry waste, and in some cases–waste from the wastewater screening process, wastewater treatment sludge, textile, medical and others (Potter, 2003; O'Brien et al., 2004; Le Calvez, 2006; Khan et al., 2019). That's way to select specific POPs emission for waste incinerators, they were considered as corresponding to Class 2 of the Source category Waste incineration–Controlled combustion, minimal air pollution control systems of the Toolkit (2013). Selected PCDD/Fs air emission factor (350 μg TEQ·t^{-1}) is close to the factor obtained recently from measurements at small waste incinerators in China—327 μg TEQ·t^{-1} (Zhang et al., 2019).

Since the capture of fly ash in incinerators in Antarctica is sporadic, POPs emission into fly ash was not calculated separately. For emission into residues PCDD/Fs emission factor for bottom ash 15 μg TEQ·t^{-1} from Toolkit (2013) was used; this amount also includes emission with fly ash where appropriate.

For PCDD/Fs and PCBs emission assessment from open burning of wastes and waste oil combustion (for retrospective assessment) emission factors as for landfill fires were applied from the Toolkit (Toolkit PCDD/PCDF, 2013; Toolkit PCB, 2019). For HCB emission assessment emission factor for the category Open burning of domestic waste was taken from Toolkit HCB (2019). Emission factor of PCDD/Fs is close to the factor used in the USA for dioxin/furan emission inventory (U.S. EPA, 2013) and it is in the range of factors obtained as a result of measurements performed in simulated landfill fires (Gullet et al., 2010; Zhang et al., 2017).

Emission factors used in PCDD/Fs emission inventory into air, residues and land as well as PCBs and HCB in air for Antarctica are shown in Table 1.

Table 1 Emission factors, which were applied in POPs emission inventory in Antarctica (Toolkit, 2013; Toolkit PCDD/PCDF, 2013; Toolkit HCB, 2019; Toolkit PCB, 2019)

Pollutant	Energy production	Heating	Ground vehicles	Waste incineration		Open burning of waste	
	Air	Air	Air	Air	Residues	Air	Land
PCDD/Fs	0.5 μg TEQ/TJ	10.0 TEQ/TJ	0.1 μg TEQ·t^{-1}	350 μg TEQ·t^{-1}	15 μg TEQ·t^{-1}	300 μg TEQ·t^{-1}	10 μg TEQ·t^{-1}
HCB	50.0 μg·t^{-1}	50.0 μg·t^{-1}	100.0 μg·t^{-1}	1000.0 μg·t^{-1}	–	10000 μg·t^{-1}	–
PCBs	0.1 TEQ/TJ	0.1 TEQ/TJ	0.0001 μg TEQ·t^{-1}	30.0 μg TEQ·t^{-1}	–	30 μg TEQ·t^{-1}	–

2.5 Comparison of PCDD/Fs emissions

Comparison of PCDD/Fs emissions in Antarctica with emissions in gateway countries (Argentina, Chile, South Africa, Australia, New Zealand) was fulfilled. Gateway countries were selected due to their close location to Antarctica and possibility of greatest direct air impact. Besides, these countries are active in Antarctica (about 1/3 stations and other facilities in Antarctica belong to them). It was also accounted that most of other countries use ports of gateway countries to entry Antarctica, including staff and cargo transfer. National reports to the Stockholm convention (4th round) were used (Secretariat of the Stockholm Convention, 2021) as sources of POPs emission data. Generally the methodology of POPs emission inventories under the Stockholm convention is based on emission factors and source sectors classifier according to UNEP (2005) and Toolkit (2013).

3 Results and discussion

3.1 POPs emission estimates

The calculation results for unintentional POPs emission by source sector for modern period are presented in Table 2. According to estimates, PCDD/Fs air emission comprises 60.74 mg TEQ, PCBs emission—5.09 mg TEQ, HCB emission—457.6 mg. Additionally 2.50 mg TEQ of PCDD/Fs are emitted annually to bottom ash, so total PCDD/Fs releases into environment amounts to 63.24 mg TEQ. Waste incineration makes the greatest contribution into the total POPs emission (96% of PCDD/Fs, 98% of PCBs and 36% of HCB air emission) (Figure 1). Vehicles provide 55% of total HCB emission.

The data obtained for POPs emission estimate for the late 1980s should be considered as only indicative, but it is obvious that POPs emissions were significantly higher which is mainly caused by open burning of waste (Table 3).

Annual PCDD/Fs emissions for the late 1980s amounted to 770.7 mg TEQ to air and 25.62 mg TEQ to land, so total PCDD/Fs emission amounted 796.3 mg TEQ. Burning of waste (mostly open) was the major source of POPs releases for that time. There is evidence of environmental effects of such practices for a long time (Lenihan, 1992; Savatyugin, 2001; Kennicutt II et al., 2010). After the Protocol on Environmental Protection to the Antarctic Treaty had been signed in 1991 and had entered into force in 1998, construction of incinerators at different

Table 2 Annual POPs emissions from anthropogenic sources in Antarctica (modern period)

Sector	Activity rates/(×10^3 t)*	Pollutants emission				
		PCDD/Fs/(mg TEQ)			PCBs/(mg TEQ)	HCB/mg
		Air	Residues	Total	Air	Air
Energy production	14.88	0.317	–	0.317	0.063	31.69
Heating	4.1	1.746	–	1.746	0.018	8.73
Ground vehicles	2.5	0.250	–	0.250	0.0003	250.24
Waste incineration	0.17	58.43	2.5	60.93	5.01	166.94
Total	–	60.74	2.5	63.24	5.09	457.6

Note: *, as estimated in the framework of this inventory

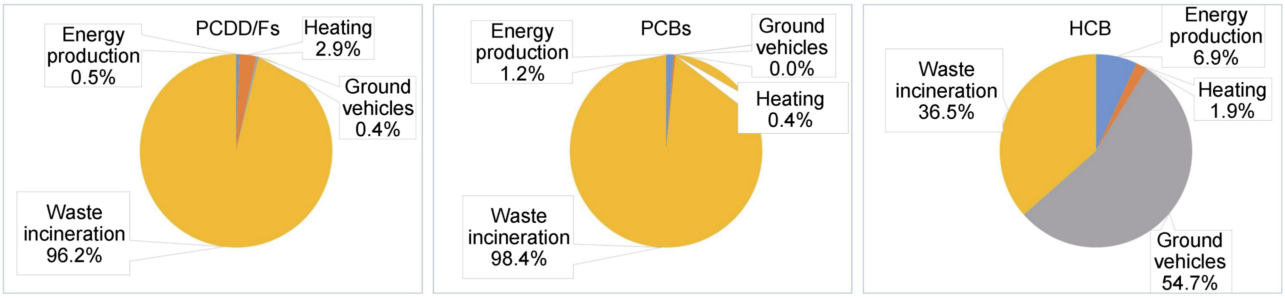

Figure 1 Contribution of source sectors into POPs air emission in Antarctica.

Table 3 Retrospective assessment of annual POPs emission at Antarctic stations (for late 1980s)

Sector	Activity rates/(×10^3 t)*	Pollutants emission				
		PCDD/Fs/(mg TEQ)			PCBs/(mg TEQ)	HCB/mg
		Air	Land	Total	Air	Air
Energy production	13.38	0.285	–	0.285	0.057	28.50
Heating	3.69	1.571	–	1.571	0.016	7.85
Ground vehicles	2.25	0.225	–	0.225	0.0002	225.1
Waste burning**	2.56	768.6	25.62	794.2	76.86	25621
Total	–	770.7	25.62	796.3	76.9	25882

Notes: *, as applied in the framework of this inventory; **, open burning and incineration combined.

stations and implementation of other measures were undertaken. It resulted in sufficient decreasing of POPs emission into environment of Antarctica: over a 30-year period air emission of dioxin/furans decreased about 13 times, PCBs—15 times and HCB—57 times.

Some other sources can also contribute to POPs unintentional releases into environment of Antarctica both in modern and past periods. For example, fires occur from time to time at Antarctic stations and cover a wide range of burning substrates. The fire at Commandante Ferraz Station in 2012 is a well-known example (Secretariat of the Antarctic Treaty, 2012). However, fires at Antarctic stations do not occur annually and it is difficult to assess their POPs releases on a yearly basis.

Certain amounts of POPs may release to water from old dumps and landfills as well as from waste water but they can hardly be estimated due to limited data. Situation improved significantly after the Protocol on Environmental Protection to the Antarctic Treaty entered into force.

Aviation and shipping contribute into POPs emission in the Antarctic, but they are beyond the scope of this study. In order to obtain indicative estimates of POPs emission from these sources the following results were used: CO_2 inventory in Antarctica (Shirsat and Graf, 2009; Farreny et.al., 2011), IPCC Guidelines (IPCC, 2006), emission factor of PCDD/Fs for aviation (Statistics Norway, 2017) as well as emission factors of PCDD/Fs and HCB for shipping (Cooper, 2004, 2005; European Environment Agency, 2019). Emission values that were obtained are as follows: PCDD/Fs and HCB emissions from shipping are 5.257 mg

TEQ and 3235.4 mg respectively, PCDD/Fs emission from aviation is 1.259 mg TEQ.

Considering the impact of POPs emission on the Antarctic environment it should be emphasized that the low stack height of diesel generators and incinerators at Antarctic stations as well as motor vehicles provide the dispersion of emitted POPs mostly in the surface layer of the atmosphere near the sources. Unfortunately, there is only one documented evidence of air impact of local emission of PCDD/Fs from waste incinerator up to now—for McMurdo Station (Lugar, 1993; Lugar et al., 1996). According to investigation made in 1992–1994 the most frequent and highest levels of PCDDs and PCDFs were measured at the location up to 500 m downwind of the station with concentrations of total PCDDs in the range of 0.12–1.8 $pg \cdot m^{-3}$ and PCDFs–0.02–2.77 $pg \cdot m^{-3}$. It was shown that PCDD/Fs were not detected at remote area. Since then no other measurements of dioxins/furans in air have been made.

Therefore research development and measurements of PCDD/Fs and other POPs in flue gases from waste incineration installations and other sources of emission in Antarctica are very important because the results will provide the basis to improve emission inventory as well as to model pollutants dispersion.

Ash handling processes from waste incineration should be in focus of further investigations in spite of its removal from the Antarctic Treaty area according to the Protocol on Environmental Protection requirements. But these residues may be a source of soil pollution by POPs within Antarctic stations during processes of their operation (ash removal from incinerator, its storage and transportation).

A certain amount of POPs may enter soil, water bodies, and marine environment with the leakage of petroleum products. According to Cooper (2005), the content of PCDD in residual oil are in the range of 0.008–0.16 μg WHO-TEQ$\cdot kg^{-1}$, HCB—<0.12 $\mu g \cdot kg^{-1}$, PCB—0.0003–0.0011 μg WHO- TEQ$\cdot kg^{-1}$. It is shown however that these data represent a limited number of samples with poor reproducibility therefore assessing the scale of such processes is a topic for a separate study.

3.2 Comparison of PCDD/Fs emissions in Antarctica with emission in the gateway countries

Comparison of dioxin/furans emission is presented in Table 4 (National reports contain no data on other unintentional POPs emission in the gateway countries). As expected the values of dioxin/furans emission in Antarctica compared to emission in the gateway countries is very low —currently it is in the range from 0.008% to 0.2% of emission in a particular country.

However the calculation of PCDD/Fs annual air emission per capita in Antarctica (maximum population at Antarctic stations was accounted) and gateway countries has proved that specific PCDD/Fs emission value in Antarctica (12.81 μg TEQ$\cdot person^{-1}$) is within the limits of this value for the gateway countries (1.11–14.68 μg TEQ$\cdot person^{-1}$) (Figure 2). Historical emission exceeds these limits significantly.

The reduction of POPs emissions in Antarctica is in harmony with global trends (Dopico and Gómez, 2015). This however does not diminish the need to obtain more accurate estimates of emissions and consequences for vulnerable polar ecosystems.

3.3 Sources of uncertainty

A lot of factors affect overall accuracy of POPs emission estimates. Among source sectors waste incineration should be considered as having the most uncertain POPs air emission levels. Emissions are impacted by composition and properties of waste, conditions of combustion, frequency and volume of loading and many other factors. High variability of PCDD/Fs content in flue gases of small-scale incinerators has been confirmed in different countries. For instance, values from 0.05 to 609.27 ng I-TEQ$\cdot Nm^{-3}$ were measured in Korea (Choi et al., 2008b). During combustion of different types of non-chlorine containing waste in Japan, PCDD/Fs concentration varied from 205 to 3080 ng$\cdot Nm^{-3}$ (Nakao et al., 2006). Uncertainty of PCDD/Fs content in flue gases leads to uncertainty in emission factors.

Table 4 Comparative values of PCDD/Fs air emission in Antarctica and in the gateway countries according to Secretariat of the Stockholm Convention (2021) (Unit: g TEQ)

Sector	Antarctica (authors' estimates)		Gateway countries (Secretariat of the Stockholm Convention, 2021)				
	Modern period	Late 1980s	Argentina (2014)	Chile (2015)	South Africa (2006)	Australia (2017)	New Zealand (2012)
Waste incineration	0.0584	–	26.0	0.04	24.43		0.794
Ferrous and non-ferrous metal production	–	–	26.842	2.84	75.49	3.2	0.404
Heat and power generation	0.002063	0.001856	15.839	14.37	441.87	15	3.329
Production of mineral products	–	–	11.0	0.49	4.04	7.5	0.066
Transportation	0.00025	0.000225	1.445	3.93	14.4		0.66
Open burning processes	–	0.7686	349.403	11.73	150.96		2.439
Production of chemicals and consumer goods	–	–	0.347	6.55	0.23	0.2	0.054
Waste disposal				34.9	0	0.1	0
Miscellaneous			11.042	0.08	0.13	0	0.191
Total	0.06074	0.7707	441.927	74.93	711.55	26.0	7.937

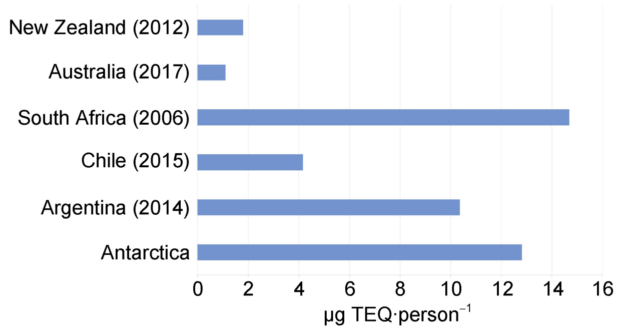

Figure 2 PCDD/Fs air emission per capita in Antarctica (modern period) and in gateway countries.

There are also significant uncertainties in assessment of PCDD/Fs content in bottom ash, due to the gaps in investigation of this substrate and numerous factors which could influence accumulation of PCDD/Fs. In some cases the samples of mixed ash (fly and bottom) were analyzed (U.S. EPA, 2019). According to the data available, emission factors of PCDD/Fs into bottom ash of different municipal waste incinerators vary by an order of magnitude (UNEP, 2005; Chung et al., 2010; Zhang et al., 2019). Lack of direct measurement of POPs in flue gases and residues from anthropogenic sources in Antarctica also influences the accuracy of emission estimates significantly.

Gaps in activity data especially on waste combustion also reduce accuracy of estimates, therefore improvement of emission sources data collection would be effective. On the whole, estimates of PCDD/Fs emission can be considered less uncertain than PCBs and HCB due to more complete study of PCDD/Fs emission globally.

Further assessments will be aimed at reduction of emission inventory uncertainties, for which improved account of waste incineration practices and incinerated volumes, as well as direct measurements of POPs in flue gases are essential.

4 Conclusions

The first estimates of PCDD/Fs, PCBs and HCB emissions from land-based sources in Antarctica have been obtained. According to estimates, dioxin/furans air emission comprises 60.74 mg TEQ, PCBs emission—5.09 mg TEQ, HCB emission—457.6 mg. Additionally, 2.5 mg PCDD/Fs are emitted annually to residues, so total PCDD/Fs releases into environment amounted to 63.24 mg TEQ. Waste incineration makes the greatest contribution into the POPs emission (96% of PCDD/Fs, 98% of PCBs, and 36% of HCB air emission). Retrospective assessment shows that over a 30-year period air emissions of PCDD/Fs decreased 13 times, PCBs—15 times and HCB—57 times, primarily due to the prohibition of open burning of waste and overall reduction of waste burning. This demonstrates the effectiveness of the Protocol on Environmental Protection.

Further measures should be aimed at improving incineration practices, the use of afterburners, dust and gas collection equipment. Establishment of emission reporting in the framework of Antarctic Treaty System is an important step towards better management of POPs emission sources in Antarctica.

Acknowledgements This work was implemented in the framework of the National Antarctic Program of Belarus for 2021-2025 and inspired by SCAR IMPACT Working Group as well as by SCAR Ant-ICON Scientific Research Programme activities. Authors thank COMNAP and Secretariat of the Antarctic Treaty as well as Secretariat of the Stockholm Convention for possibilities to use their information resources. We appreciate two anonymous reviewers, and Associate Editor Prof. Zhouqing Xie for their constructive comments that have further improved the manuscript.

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