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Sixty-year trends in sulphur dioxide emissions from anthropogenic ground-based sources in Antarctica

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Abstract: Knowledge of trends of pollutants released into the environment is very important for interpreting the observed trends in pollution of natural environments and forecasting their development. This article reconstructs for the first time a series of sulphur dioxide emissions from the main categories of land-based sources of emissions in Antarctica (diesel generators, heating systems, vehicles and waste incineration) over 60 years: from the beginning of the intensive construction of scientific stations in 1960 until 2019. The trends in the sulphur content of fuels and the dynamics of fuel consumption by the main categories of emission sources are taken into account. According to the estimates obtained, total emissions of sulphur dioxide in Antarctica varied in the range of 28.6–161.3 tons. This paper establishes that the greatest levels of sulphur dioxide emissions occurred in the late 1980s–beginning of the 1990s. In subsequent years, there was a rapid reduction in emissions, primarily due to a reduction of the sulphur content of fuels. The rates of reduction of sulphur dioxide emissions for different areas of Antarctica are also shown.

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Key words: Antarctic stations, emission, sulphur dioxide, trend

Introduction

Sulphur oxide emissions, primarily sulphur dioxide (SO_2), are the main causes of environmental acidification in Europe, North America and East Asia. Sulphur dioxide emissions influence the level of acidification of freshwater ecosystems and soils (Stoddard *et al.* 1999, Schopp *et al.* 2003, Vestreng *et al.* 2007) and climate change (Haywood & Boucher 2000, Ramanathan *et al.* 2001) and have impacts on human health (WHO 2000). The acidification situation has been serious in large parts of northern Europe in the 1970s, mainly in the Fenno-Scandia region due to the slow weathering of soil and bedrock.

A number of international agreements have been signed to reduce sulphur emissions, in particular the 1979 Geneva Convention on Long-Range Transboundary Air Pollution, with subsequent 1985, 1994 and 1999 protocols (Reis *et al.* 2012, UNECE 2015). Much effort has been made regarding obtaining reliable SO_2 emission inventories as well as to establish an SO_2 ambient monitoring network.

The human impact on the natural environment of Antarctica may seem insignificant in absolute terms compared to the impacts in other, more inhabited areas of Earth. However, the Protocol on Environmental Protection to the Antarctic Treaty and its connected documents established strict requirements for conducting activities in Antarctica.

The impacts of SO_2 in Antarctica are much less well understood, partially due to the fragmental character of its ecosystems, freshwater bodies and soils. Most often studied are sulphur concentrations in snow and ice, including ice cores. However, considering the vulnerability of Antarctic ecosystems (Bargagli 2008, Tin *et al.* 2009), it can be expected that these impacts will not be negligible.

Regular and detailed SO_2 emission inventories, similarly to inventories of other pollutant emissions, are very important for Antarctica because they will form the scientific basis for the assessment of the impact of this pollutant on Antarctic ecosystems, including its cumulative impacts, and they will support intentions to keep the continent clean, as agreed according to in the Protocol on Environmental Protection to the Antarctic Treaty. However, despite emissions estimates being conducted by certain Antarctic programmes, only two continent-wide estimates of SO_2 emissions for specific years are available: Boutron & Wolff (1989) for 1987–1988 and Shirsat & Graf (2009) for 2004–2005.

The purpose of this article is to present yearly estimates of anthropogenic emissions of SO_2 in the Antarctic Treaty area from land-based sources (diesel generators, heating systems, off-road and road transport and waste incineration) over the whole period of the intensive exploration of the continent starting from 1960 until 2019 (i.e. a 60 year period), accounting for trends in activity levels and fuel quality parameters.

Methods and data

General methodology of emissions calculations

SO_2 emissions have been estimated using the traditional approach of using data on activities that have been assessed as significant emission sources for Antarctica: energy production, heating, waste incineration, ground vehicles and emission factors.

Annual SO_2 emissions have been estimated for every main source sector using a bottom-up approach based on data on activity in a sector (number of installations and their capacity, fuel use, waste incinerated) and emission factors of SO_2 per unit of activity.

The basic calculation formula is as follows:

$$R = \frac{r_{xy}}{r_{yy}} \tag{1}$$

where $E_{i,j}$ represents SO_2 emissions in sector i in year j, $A_{i,j}$ represents activity in sector i in year j and $F_{i,j}$ represents the emission factor of SO_2 in sector i in year j.

The annual activity in source sectors was estimated in three steps.

In the first stage, fuel consumption in terms of energy production for every year for every Antarctic station was estimated using data on capacities of installed diesel generators at Antarctic stations and number of days (or hours) of their operation per year, accounting for the type of station (seasonal or year-round) and hourly consumption of fuel (Kakareka 2020).

In the second stage, fuel consumption per station was estimated for heating and for ground vehicle use.

In the third stage, waste incineration per station was estimated.

Yearly SO_2 emission factors for energy, heating and vehicles were obtained from the sulphur content in fuels; for waste incineration, emission factors were taken from available emission inventory guidelines.

Assessment of activity in the main sectors and its dynamics

The assessment of fuel consumption and its dynamics was carried out on the basis of available data on fuel consumption, the capacity of fuel-burning units, the number of vehicles and other information.

Particular attention was paid to the identification of the stages of rapid change in Antarctic programmes, the opening and closing of stations and their reconstruction. The operation of the nuclear power plant at McMurdo Station in 1962–1972 was particularly taken into account.

Estimates of fuel consumption on the continent as a whole and by major sectors are available for 1987, 2004–2005 and for the current period (Boutron & Wolff 1989, Shirsat & Graf 2009, Kakareka 2020, Kakareka & Kukharchyk 2022). To produce a continuous series for fuel consumption since 1960 per Antarctic station, the following data were used:

1) Volumes of fuel consumption by station, expedition and programme: data are available for some years for Soviet (SAE), Russian (RAE) and Belarusian (BAE) Antarctic Expeditions, the United States Antarctic Program (USAP), the Australian Antarctic Program and Antarctica New Zealand. Analyses of data sources are given in Kakareka (2020) and Kakareka & Kukharchyk (2022).
2) Data on installed capacities of diesel generators for different years; these data were previously generalized for the modern period for all active year-round and most of seasonal Antarctic stations (Kakareka 2020). There are some seasonal stations (~10), for which data on their energy systems are lacking. However, these stations are small and operate in the summer only, and they do not make significant inputs to total emissions. According to our estimates, 90–95% of fuel consumption in Antarctica is due to the operation of year-round stations. In addition, even for stations lacking direct data on fuel consumption and installed diesel generators, we can make reasonable assumptions about fuel consumption parameters according to their populations because there is a strong correlation between population and fuel use. This approach was used in this paper to fill in any gaps in the data on fuel consumption. For the early 1960s, the data of Dubrovin & Petrov (1971) served as the basis for the characteristics of the installed capacities of diesel generators at stations. For the subsequent period, data from the monographs of Savatyugin (2001, 2009, 2019), Savatyugin & Preobrazhenskaya (1999, 2000) and Savatyugin & Geller (2021) and reports on inspections of Antarctic stations in accordance with Article VII of the Antarctic Treaty, available on the website of the Secretariat of the Antarctic Treaty (Secretariat of the Antarctic Treaty 2022b), were used.
3) Data on the number and structure of vehicle fleets at stations available from the Council of Managers of National Antarctic Programs (COMNAP) Catalogue of Antarctic Stations (COMNAP 2017a), in the annual reports of the Parties to the Electronic Information Exchange System (EIES), inspection reports and publications (Secretariat of the Antarctic Treaty 2022a).

In addition, station population data, which are available on the COMNAP website (COMNAP 2017b) and other sources, were used.

Diesel fuel (density 0.83–0.84 kg/l), which is a gas oil fraction adapted for operation in polar conditions as fuel for diesel generators, is used at most stations. It is called Arctic diesel (in the USSR, Russian Federation and

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Belarus), Antarctic (Arctic) diesel (in Australia), DFA (Diesel Fuel, Arctic grade), polar diesel (in European Union countries) and GOA (Gas Oil Antartico; in Chile, Argentina, Uruguay and some other Latin America countries). Diesel fuel was also used until 1989 at the US Antarctic stations McMurdo and Amundsen-Scott but was later replaced by kerosene-based aviation fuel (JP-8; National Science Foundation 1991). Similar fuel is used at Scott Base. At stations operated by some programmes, in particular by the British Antarctic Survey (BAS), Marine gas oil (MGO) is used (Secretariat of the Antarctic Treaty 2017). All of these fuels differ in their sulphur content.

At most stations where autonomous heating systems are installed, these are fuelled by the same fuel as diesel generators. This is also true of heavy vehicles and specialized equipment. For road vehicles and light off-road vehicles such as snowmobiles, motor gasoline of various grades is used (e.g. A-72, A-76, etc., at SAE Antarctic stations, Mogas at USAP stations). Gasoline has been unleaded in recent decades.

Energy production consumes the bulk of the fuel and occurs at almost all stations in Antarctica. Annual fuel consumption in the energy sector was calculated based on the capacity of installed diesel generators and their annual load (number of hours of operation per year; it is assumed that generally one diesel generator is running at a time at any one station; Kakareka 2020).

As shown above, this paper accounts for the fact that, in 1962–1972 at McMurdo Station, an atomic power plant with a PM-3A reactor was in operation. During its 10 year lifespan, the nuclear power station produced over 78 million kilowatt-hours of electricity and 13 million gallons (49.2 million litres) of fresh water using the excess steam in a desalination plant. Once fully operational, the role of the plant was twofold: to produce 1.8 MW of electricity for McMurdo Station as well as to produce steam to operate a desalination plant for the production of 14 000 gallons (53 000 l) of fresh drinking water per day (Shafer 1967).

Fuel consumption in other sectors (heating, transportation and other machinery, waste combustion) was determined based on their share of gross fuel consumption for stations with available data and according to expert assessment for the other stations.

Data on fuel consumption by vehicles are very patchy. More or less complete data are available for McMurdo, Scott Base and Australian stations. Based on these data, it is difficult to derive a unified indicator of, for example, fuel consumption per capita. Therefore, the stations were grouped into the following categories:

1) The largest station with a large number of vehicles (McMurdo). It is known that, in 1989–1990, 570 000 l of gasoline (unleaded) were delivered to McMurdo (National Science Foundation 1991). In 2019, fuel consumption by transport and other equipment amounted to 1 735 000 l (National Science Foundation 2019); fuel consumption by mobile equipment is 30–35% of the fuel consumption in energy production (excluding consumption at Amundsen-Scott and Palmer stations).
2) Stations with an average number of transport units, which are used as bases for resupply traverses (Mirny, Progress, Dumont d'Urville, Novolazarevskaya, Zhonzhang, Syowa): fuel consumption by transport is estimated here to be 20–30% of the fuel consumption in energy production.
3) Continental stations (East Antarctica): fuel consumption by transport is estimated here to be 10% of the consumption in energy production.
4) Large and medium-sized stations in West Antarctica: fuel consumption by transport is estimated here to be 1–5% of the consumption in energy production.

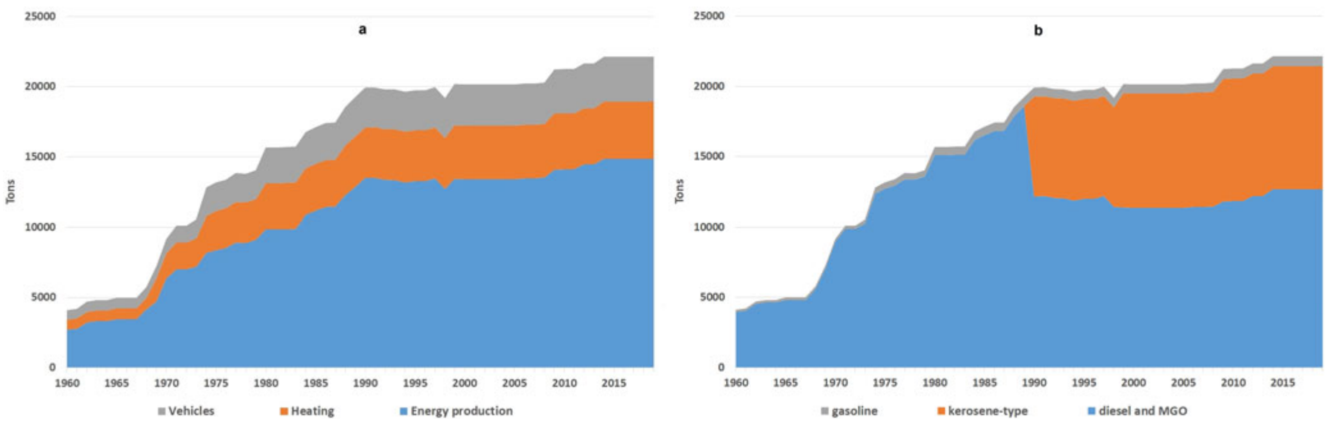

Figure 1. Trends of fuel consumption by land-based emission sources in Antarctica: **a.** by source category; **b.** by type of fuel. MGO = marine gas oil.

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5) Small stations in West Antarctica: fuel consumption by transport is estimated here to be < 1% of the consumption in energy production.

Gasoline is used at most stations for snowmobiles, motorboats and, less often, for road vehicles. It is assumed that the share of gasoline in the total fuel consumption by transport is as follows:

1) Stations of West Antarctic: 30% (McMurdo: 33%)
2) Inland stations: 5%
3) East Antarctic coastal stations: 10%

Reconstructed fuel consumption values for Antarctica are shown in Fig. 1. According to estimates, from 1960 to 1990 fuel consumption increased by more than fourfold (from 4100 to 19 900 tons); later level of fuel consumption was generally stable or showed only slight growth (in 2019: 22 100 tons). Shares of diesel fuel and MGO comprised 99.6% in 1960 and 57.3% in 2019, the kerosene-based fuel share comprised 0% in 1960 and 39.5% in 2019 and the gasoline fuel share comprised 3.9% in 1960 and 3.8% in 2019.

Waste incineration

Generally, waste incineration is not a significant contributor to total emissions of SO_2, but attention was given to the reconstruction of activity series in this sector over the 60 year period due to its important contribution to emissions of persistent organic pollutants and heavy metals, which are planned to be estimated in future.

No systematic continent-wide account exists of waste incineration in Antarctica. Approaches to assess volumes of waste incineration for the modern period and the late 1980s have been described previously (Kakareka & Kukharchyk 2022). Evidence was found of modern waste incinerator use at 29 Antarctic stations and of a high probability of waste incineration at 17 other stations; waste incineration volume data for certain years are available for eight stations (Casey, Davis, Mawson, Dumont d'Urville, Maitri, Syowa, Progress, Novolazarevskaya; COMNAP 2006, Australia State of Environment 2016, 2022, etc.).

Due to scarcity of direct data, waste incineration volumes for other Antarctic stations were calculated using mean ratios of waste incineration at the eight identified station for the maximum populations of these stations. Incineration volumes at the abovementioned eight stations range from 3.32 to 8.00 tons; the peak populations of these stations range from 50 to 170 people, with the amount of annual waste incineration equalling 110 kg/person on average. Based on this specific value, the total estimated volume of current waste incineration in Antarctica is estimated to be 287.2 tons/year.

For the earlier period, there are practically no quantitative estimates available: among those that are available are the calculations of waste incineration at McMurdo Station performed in the late 1980s (National Science Foundation 1991). There is information available regarding the construction of an incineration plant in the early 1970s at McMurdo Station and regarding the use of such installations at some other stations, but it is difficult to form a complete picture. The combined volume of open waste burned and combusted in incinerators in the late 1980s, including waste oil products, amounts to 2562 tons according to the estimates of Kakareka & Kukharhyk (2022).

In general, the available data are insufficient to trace the dynamics of waste incineration separately for open burning and incineration in installations. Therefore, in this paper, emissions from waste incineration were estimated without distinguishing between open burning and incineration in special installations. It can be reasonably assumed that the share of open burning decreased exponentially from 80–90% in 1989 to close to 0% values in 1999 and subsequent years.

Estimates of fuel combustion dynamics were used to construct the waste incineration series. At the same time, the entire 60 year period is divided into three stages: until 1987, 1988–1998 and 1999–2019.

The following starting points were used to reconstruct the waste incineration series:

1) All waste incineration after 1998 was in closed installations. Before this period, both open burning and closed incineration were used.
2) Since 1999, waste incineration has been carried out at the same stations as at present.
3) Until 1998, waste incineration was carried out at all stations.

Calculation of basic sulphur dioxide emission factors and their vectors

The SO_2 emission factors for fuel combustion were calculated on the basis of the sulphur contents in the fuels. The series of SO_2 emission factors was constructed on the basis of the changes to the standards for sulphur contents in these fuels.

To construct the series of the sulphur contents in fuels (diesel, gasoline and others), the following sources of data were used:

1) International and national guidelines on emission inventories (AP-42 1996, EMEP/EEA 2019, GOST R 2019, etc.);
2) European Union (EU) and national standards on fuel quality from national environmental and standardization agencies and standards databases (Diesel Fuel Regulations 2002, UK Government 2014, Gas Oil Antartico 2018, Fuel Quality Standards 2019, EPA 2022, EU 2022, etc.);

3) Fuel quality surveys, reviews and databases (Infineum Insight 2000, 2018, Transportpolicy.net, diesel.net, Stratasadvisors.com, etc.).

The performed analysis showed that for the period up to the 1980s–1990s, few data were available on the sulphur contents in fuels, except for in some countries. Therefore, it was assumed that in the 1960s–1970s these contents were approximately the same in all countries, as measures to reduce these sulphur contents began to be taken mainly in the 1990s.

As the actual sulphur contents in the fuels may differ from the standards, it was assumed that the sulphur contents in the fuels in a particular year correspond to the upper limits set by the standards for this period of time if no other information is available.

It is known that at Antarctic stations usually the same diesel fuel is used for diesel engines, heating systems and vehicles (except for vehicles with sparkle ignitions).

Below is a description of the fuels used at some Antarctic stations.

Diesel fuel was used at US Antarctic stations until 1989–1990 (National Science Foundation 1991). The regulation of sulphur in fuels in the USA began in 1993; before that, the content reached 0.5%. According to AP-42 (1975), the SO_2 emission factor for stationary diesel engines was 3.74 g/l, which corresponds to a sulphur content of 1.87 g S/l (2250 ppm).

In the early 1990s, McMurdo and Amundsen-Scott stations switched to JP-8 military aviation fuel, which is largely the same as commercial Jet A-1 fuel; JP-8 is used both for aviation and for diesel generators, heating systems and off-road vehicles. The sulphur content limit for JP-8 (jet fuels) is 0.3% (3000 ppm; IPCC 1999). However, despite the fact that the sulphur limit for JP-8 (jet fuels) has not been changed, the real content of sulphur has reduced significantly and recent years, probably amounting to ~400–600 ppm (IPCC 1999).

At the same time, according to the National Science Foundation (2019), McMurdo Station's power generation emissions from burning 3.2464 million litres of fuel are projected to be 13.745 tonnes. The calculated SO_2 emission factor is 4.211 g/l or 5.2 g/kg, which is equivalent to 2600 ppm of sulphur, or close to the limit value.

The main fuel used at Scott Base (New Zealand) currently is JP-8, as it is at McMurdo Station. Taking

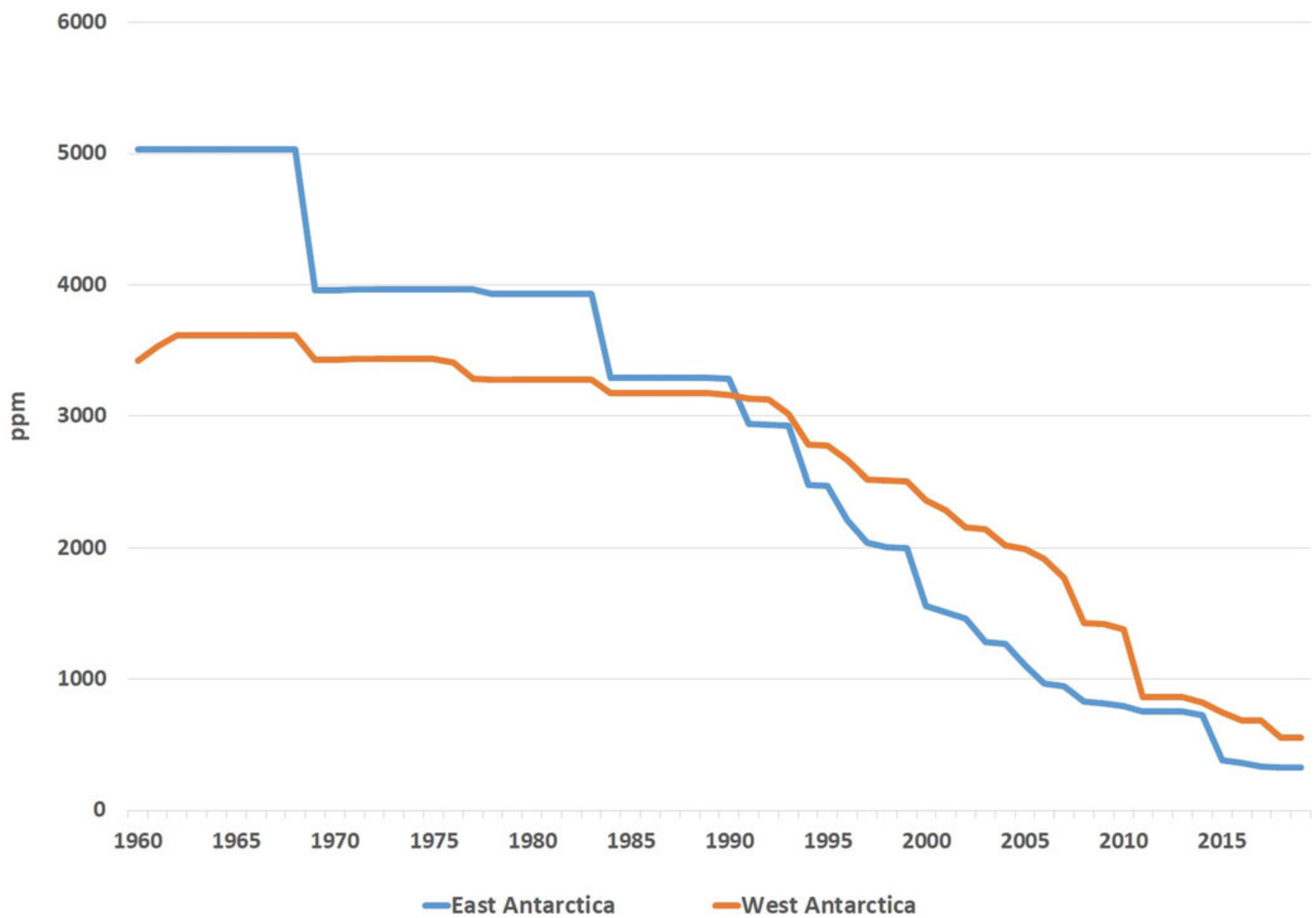

Figure 2. Estimated trends of averaged sulphur contents of fuels used by land-based emission sources in Antarctica.

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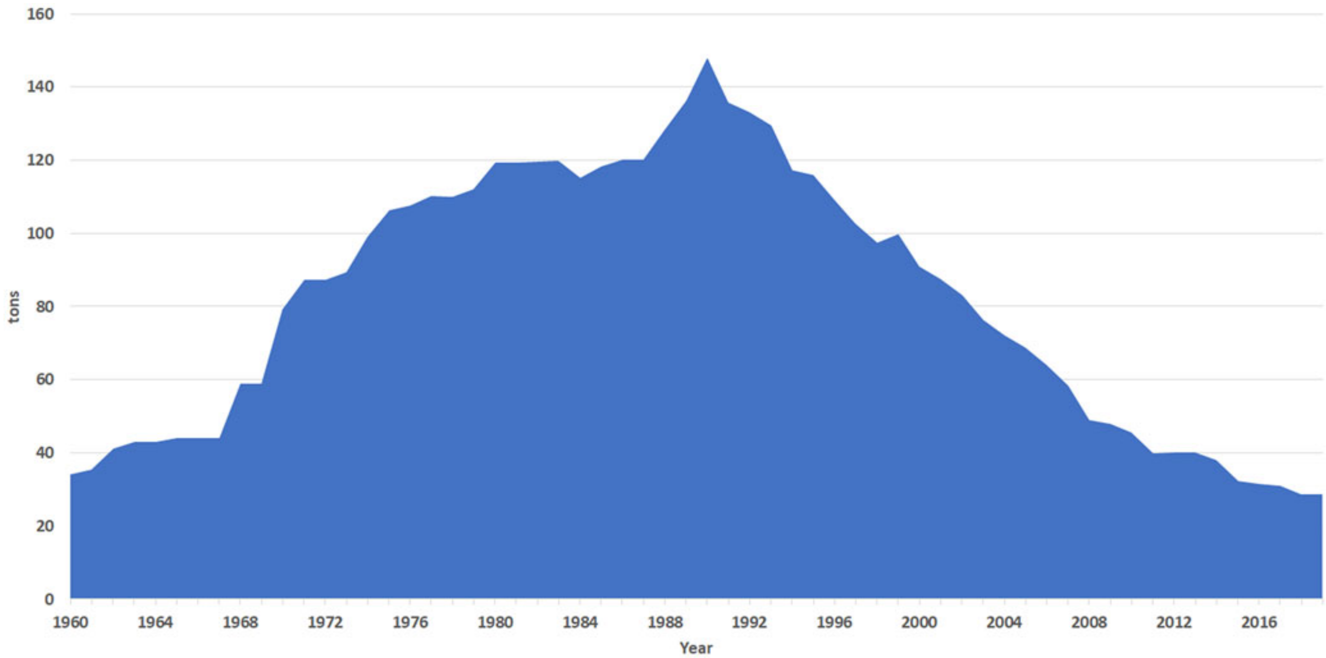

Figure 3. Trend of total sulphur dioxide emissions from anthropogenic land-based sources in Antarctica from 1960 to 2019.

into account the close logistical connections between these stations, both in the present and in the past, the trend of change in the sulphur content in the diesel fuel used is assumed to be the same for these stations.

The main diesel fuel used at the Australian Antarctic stations is Special Antarctic Blend (SAB). Its sulphur limits were 500 ppm on 31 December 2002, 50 ppm on 1 January 2006 and 10 ppm from 1 January 2009 (Fuel Quality Standards 2019).

At the stations of EU countries, mainly Antarctic (Arctic) diesel, DFA and polar diesel are used; the sulphur content currently complies with the Euro5 standard of 0.001% (10 ppm). Diesel and gasoline have been limited to 10 ppm of sulphur since 2009 for on-road vehicles and 2011 for off-road vehicles. At Norwegian Troll Station, Jet-A-1 with a 0.3% sulphur content is used (Secretariat of the Antarctic Treaty 2004).

UK Antarctic stations use MGO (equivalent to marine fuel oil, bunker fuel A, Fuel Oil No 2), with a sulphur content of 1%, as fuel for diesel generators, heating and transport (Secretariat of the Antarctic Treaty 2017). The International Maritime Organization (IMO) has set a limit of 0.50% m/m (mass by mass) for sulphur in fuel oil used on board of ships operating outside designated Emission Control Areas (Regulation 14 of MARPOL Annex VI), which came into effect on 1 January 2020.

At the Antarctic stations of the Russian Federation and Belarus, Diesel Winter (DW) and Diesel Arctic (DA) fuels are used. Diesel fuel quality standards have changed many times in the last 60 years. The following trend of sulphur contents in diesel fuels was adopted: before 1969 - 1%; 1969–1983 - 0.5%; 1984–2014 - 0.2%; after 2014 - 0.035% (Kakareka & Salivonchyk 2022).

The overall trend of sulphur contents in liquid fuels used in Antarctica since 1960 is shown in Fig. 2.

It was assumed that the sulphur contents in waste is the same as in Europe, and the same emission factor was used for all years according to EMEP/EEA (2019). It was also assumed that Antarctic waste can be considered as municipal waste in this respect.

Results

Modern sulphur dioxide emissions

Total SO_2 emissions in Antarctica in 2019 were estimated at 28.6 tons. In 2019, an average of 65% of emissions were due to the operation of diesel generators, 21.7% due to heating systems and 11.5% due to land vehicles; the contribution of waste incineration to total emissions was 1.8%.

Table I. Dynamics of sulphur dioxide emissions in Antarctica by source sector (tons).

Source sector	1960	1970	1980	1990	2000	2010	2019
Energy production	25.5	57.3	80.6	101.7	62.9	32.7	18.6
Heating	4.0	12.3	21.9	27.3	17.2	7.7	6.2
Road vehicles	0.3	0.5	1.2	1.3	1.2	0.3	0.1
Off-road vehicles and machinery	3.1	5.7	11.7	14.4	9.2	4.4	3.2
Waste incineration	1.2	3.4	4.0	3.3	0.5	0.5	0.5
Total	*34.1*	*79.2*	*119.4*	*147.9*	*91.0*	*45.6*	*28.6*

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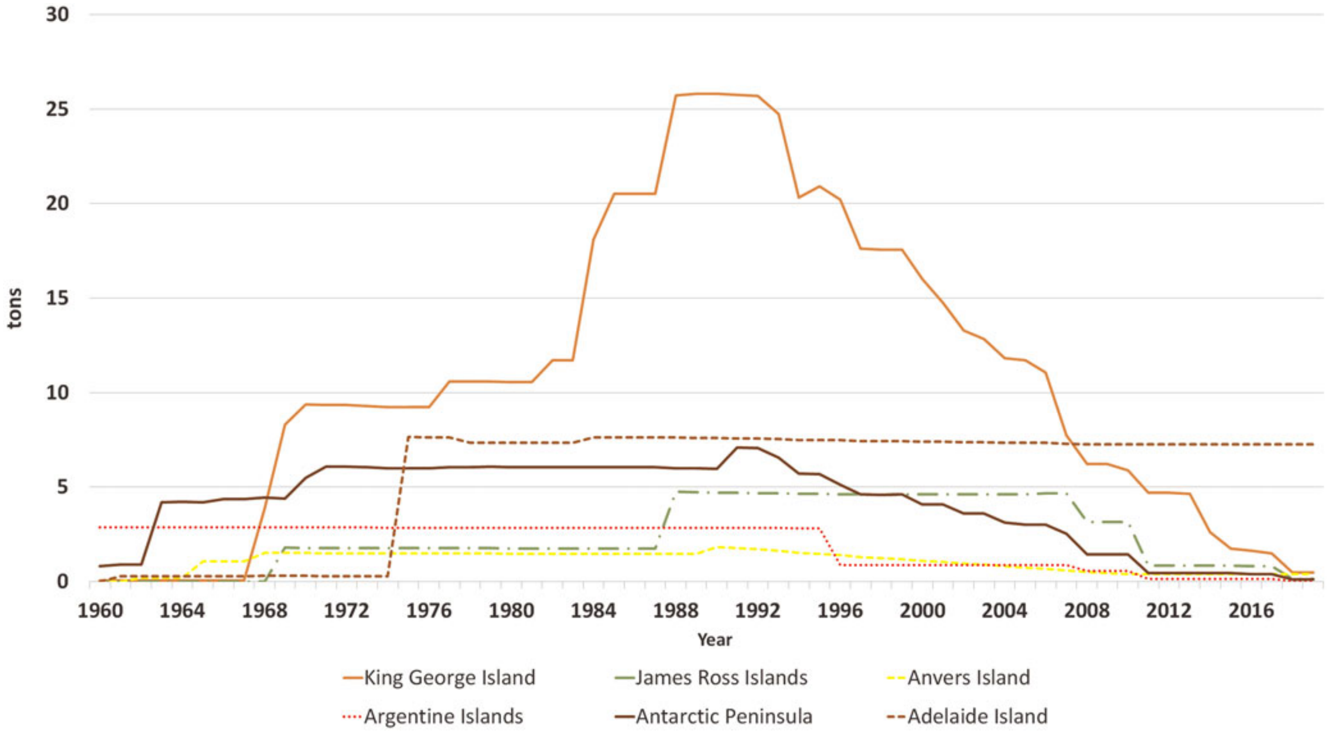

Figure 4. Trends of sulphur dioxide emissions from anthropogenic land-based sources from 1960 to 2019 in some geographical areas of West Antarctica.

Emission trends

Obtained trends in SO_2 emissions for the period from 1960 to 2019 are shown in Fig. 3.

According to the estimates obtained, total emissions of SO_2 in Antarctica varied in the range of 28.6–161.3 tons. The minimum emissions are typical for 1960–1961 (34.1–35.4 tons) and 2018–2019 (28.6 tons). The 60 year period can be split into two distinct periods: from 1960 to 1990, mainly showing increasing emissions; and from 1991 to 2019, mainly showing decreasing emissions. Since 2013, the level of SO_2 emissions has been < 40 tons/year, which corresponds to the level of emissions from the early 1960s. It should be noted that if the growth in SO_2 emissions in the 1960s–1990s was due to an increase in fuel consumption, then the subsequent reductions in emissions were due to decreases in the sulphur contents in the fuels used.

In the early 1960s, energy accounted for 75% of emissions, heating systems for 11%, transportation for 10% and waste incineration for 4%. The modern period is characterized by greater contributions from heating and transportation and reduced contributions from energy production and waste incineration (Table I).

Additionally, emission trends were calculated for the different geographical regions of Antarctica. The obtained trends in SO_2 emissions for the period from 1960 to 2019 by geographical region of Antarctica are shown in Fig. 4 & Table II.

SO_2 emissions in West Antarctica (including the Transantarctic Mountains) over most of the period represented more than two-thirds of total ground-based emissions on the continent. Today, the stations in this region contribute ~70% of total SO_2 emissions; from 1962 to 1983, the contribution of West Antarctica to total emission was < 60%, and from 1968 to 1974 it was < 50%.

Emission estimates were made by two main types of station represented in Antarctica: seasonal and year-round. The whole period is dominated by the contribution of year-round stations: their share generally exceed 90%, and only in some years (e.g. in 1970) was their contribution below this level.

In 1960, the distribution of stations by input to total SO_2 emissions was as follows: half of the stations (13 out of 26) accounted for 95% of emissions and 23% of stations accounted for ~80% of emission. A similar ratio remained by 1970: 51% of stations accounted for 95%

Table II. Dynamics of sulphur dioxide emissions by region of Antarctica (tons).

Region	1960	1970	1980	1990	2000	2010	2019
East Antarctica	12.2	44.1	54.8	56.4	30.2	14.8	8.7
West Antarctica	21.9	35.1	64.6	91.5	60.8	25.4	19.9
Total	*34.1*	*79.2*	*119.4*	*147.9*	*91.0*	*45.6*	*28.6*

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of emissions and 32% of stations accounted for 78% of emissions. By 1980, the distribution had become more even: 33% of stations accounted for 80% of emissions and 60% of stations accounted for 96% of emissions. By 1990, the distribution had become less even: 30% of stations accounted for 80% of emissions and 42% of stations accounted for 91% of emissions.

In subsequent years, the uneven contribution of stations to SO_2 emissions constantly increased: in 2000, 20% of stations accounted for 76% of emission; in 2010, 15% of stations accounted for 81% of emissions; and in 2019, 5.4% of stations accounted for 79% of emissions. This is primarily due to the uneven rates of reduction of sulphur contents in the fuels used.

Discussion

Comparison with previous estimates for Antarctica

According to Boutron & Wolff (1989), sulphur emissions from land-based sources in Antarctica were 55.5 tons in the late 1980s, including 54 tons from diesel fuel combustion.

According to our estimates, SO_2 emissions from land-based sources in the late 1980s were ~120 tons. Current estimates account for the spatial and temporal variability of the sulphur contents of the fuels used in Antarctica.

According to Shirsat & Graf (2009), the total estimated yearly emissions of SO_2 from power generation and vehicles during April 2004–March 2005 were ~158 Mg/year (not include waste incineration), which is approximately twice as high as current estimates. Our estimates take into account the significant drop in sulphur contents of fuels in the 1990s–early 2000s and the large differences in sulphur contents of the fuels combusted across Antarctic stations.

Comparison with global and regional sulphur dioxide emission trends

For comparison, the Emissions Database for Global Atmospheric Research (EDGAR) global estimates (Crippa *et al.* 2016, 2019), Community Emissions Data System (CEDS) data (Hoesly *et al.* 2018), Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) data (WebDab 2022) and US Environmental Protection Agency (EPA) data (US EPA 2022) were used.

The absolute values of global and Antarctic emission are not comparable, so only trends were compared.

Antarctic trends can compared to European, American and global ones. A characteristic feature is the growth in SO_2 emissions in Antarctica until 1990, after which there has been a gradual reduction in emissions. There are also peculiarities in emission trends in the various Antarctic regions due to the use of fuels with varying sulphur contents.

Other sources of sulphur dioxide emissions

The main anthropogenic source of SO_2 emission in Antarctica (excluding the adjacent seas of the Southern Ocean) not covered by this study is aviation.

According to Botron & Wolff (1989), SO_2 emissions from aviation in Antarctica in 1987 amounted to 14.1 tons. The estimates of Shirsat & Graf (2009) indicate that the total SO_2 emission flux rate during taxi, climb-out, approach and cruising in aviation from November 2004 to March 2005 was 56 tons. Trends of SO_2 emissions in this sector have not yet been studied.

It should be mentioned that there are other anthropogenic sources of SO_2 emissions from the combustion of some other fuels not accounted for in this assessment, such as hard coal, which was used for heating and cooking during certain stages of Antarctic exploration.

Long-range atmospheric transport of anthropogenic and natural sulphur compounds also provides a significant contribution to sulphate deposition in Antarctica.

Among natural sources, volcanic emissions (Radke 1982, Kyle 1990, Liss *et al.* 1997, Sweeny *et al.* 2008, Obryk *et al.* 2018, Goto-Azuma *et al.* 2019) are of great importance. According to Sweeny *et al.* (2008), the mean SO_2 emission rate from Mount Erebus is 61 Mg/day, whereas Radke (1982) reported 35 tons of SO_2 emissions per day. According to Kyle (1990), following a 4 month period of sustained strombolian activity in late 1984, these SO_2 emissions declined from 230 Mg/day in 1983 to 25 Mg/day, and then slowly increased from 16 Mg/day in 1985 to 51 Mg/day in 1987.

Biogenic emissions of sulphate precursors from the oceans, especially dimethylsulfide (DMS), contribute significantly to the overall input of sulphur into Antarctica (Liss *et al.* 1997, Goto-Azuma *et al.* 2019).

Sources of uncertainty

The uncertainty in estimates of SO_2 emissions is due to the uncertainty in activity data (fuel and waste combustion) and the uncertainty in SO_2 emission factors.

Fuel consumption varies due to differences in the duration of the period of operation of stations, changes of capacities of installed diesel generators and the number and type of vehicles used, as well as year-to-year variations in waste incineration. The uncertainty of this type can be quantified from the continuous series of fuel consumption and waste incineration data available for some stations, particular the Australian stations. For the first 20 years of the study period, when data on the use of diesel generators and other equipment are patchy, this uncertainty is significantly greater.

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The uncertainty of specific emissions of SO_2 is associated primarily with the limited data on the sulphur contents of the fuels used, especially for the 1960s–1970s. Indicators characterizing the sulphur contents of these fuels were taken according to the upper threshold of the sulphur content standards at force at the time in the territory of the country that operates the station. However, several standards were often simultaneously applicable in the territory of the country. The actual sulphur contents of the fuels could deviate from these standards in one direction or another. Data on the actual sulphur contents of the fuels imported to Antarctica are largely absent in the literature.

It appears that the second group of factors represent the main contributor to the overall uncertainty in estimates of SO_2 emissions.

One of the main results of the current assessment is that the overall situation regarding SO_2 emissions in Antarctica is far more heterogeneous spatially and temporally than it was previously believed. This is understandable if we take into account the fact that 29 countries from five continents operate Antarctic stations and that variations in fuel quality parameters across stations are very significant.

It should also be assumed that the overall uncertainty regarding emission estimates from Antarctic sources is greater than for other parts of the world due to the lack of such studies having been conducted in the region. Every such study contributes to reducing the uncertainty of emission estimates. Therefore, taking into account the fact that this is only the third estimate of SO_2 emissions for the whole of Antarctica - a large, sparsely populated continent – the overall uncertainty in the estimates can be considered acceptable.

Air pollution impacts

The data show that the seasonal average SO_2 levels at two monitoring locations near McMurdo Station were less than the 0.1 ppb analyser detection limit (Lugar 1993, 1994). Average monthly SO_2 concentrations at Hut Point (based on hourly average data) ranged from below the detection limit of 0.1 ppb for November and December to 0.1 ppb for January and February. The single maximum hourly average SO_2 concentration for the season was 5.6 ppb, recorded in January.

Spectrophotometric measurements of SO_2 are made at a few Antarctic stations. Thus, measurements of SO_2 at Indian station Maitri have been conducted since 2000 (Kulandaivelu & Peshin 2003). According to Mariano *et al.* (2010), atmospheric SO_2 measurements are conducted at the Brazilian Antarctic station Comandante Ferraz, where a Brewer Spectrophotometer is used. The total column of SO_2 was measured in spring from 2003 to 2009. It was observed that the total column of SO_2 did not show any difference over the time of the development of the ozone hole as compared to other periods. In the next a few days, however, the SO_2 total column exceeded the value considered normal for remote regions.

Observations (Wolff & Cachier 1998) revealed an average SO_2 concentration of 0.74 $\mu g/m^3$ (~250 pptv) near McMurdo Station during the summers of 1995/1996 and 1996/1997. According to Graff *et al.* (2010), measurements at the South Pole reported SO_2 concentrations of ~10 pptv.

Conclusion

Based on a comprehensive analysis of the available data, a series of SO_2 emissions over 60 years from the main categories of land-based sources in Antarctica has been constructed. It is shown that emission level trends show significant differences from the observed trends in other parts of the world. The period before 1990 is characterized by a predominance of growth in emissions due to the construction of new stations and the commissioning of greater capacities at power plants. Since the early 1990s, the prevailing trend of a reduction of SO_2 emissions is observed. This tendency is mainly due to the introduction of the stricter limits of the sulphur contents of liquid fuels globally. Such measures, aimed primarily at reducing SO_2 emissions in populated mainland areas, has led to reductions in SO_2 emissions in Antarctica.

It was found that emission reduction rates in Antarctica are not uniform; they are greater in areas (stations) where regular diesel fuel is used and significantly lower where aviation kerosene or MGO is used, as the latter fuels have not been subject to a tightening of regulatory requirements for sulphur content until recently.

As the next steps for SO_2 emission estimates, the following improvements may be considered:

1) Clarification of the emission series, accounting for minor types of fuel that were used at certain stages of Antarctic exploration (e.g. hard coal)
2) Direct in-stack measurements of SO_2 emissions
3) Emission projections for different scenarios of implementation of SO_2 emission reduction measures and Antarctic station development.

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Competing interests

The author declares none.

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