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Continental Scale Antarctic deposition of sulphur and black carbon from anthropogenic and volcanic sources

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Abstract

While Antarctica is often described as a pristine environment, the potential threats from local pollution sources including tourist ships and emissions associated with scientific activities have recently been raised. However, to date there has been no systematic attempt to model the impacts of such pollutants at the continental scale. Indeed, until very recently there was not even a sulphur emission budget available for Antarctica. Here we present the first comprehensive study of atmospheric pollution in Antarctica using a limited area chemistry climate model, and a monthly emissions inventory for sulphur from maintenance of research stations, ground and air traffic, shipping and the active volcano Mt. Erebus. We find that ship emissions, both sulphurous and black carbon, dominate anthropogenic pollution near the ground. These are likely to rise considerably if recent trends in tourism continue.

1 Introduction

The remote and mostly uninhabited continent of Antarctica was discovered in the late 18th Century but forsaken except for sporadic expeditions until the International Geophysical Year in 1957/1958 when a number of permanent research stations were established. Currently 37 year-round and 16 summer-only research bases operate in Antarctica, McMurdo (USA) being the largest with about 1000 people during the austral summer (November to March) and 250 during winter (April to October). The history of tourism in Antarctica also dates back to the 1950s. In 2000 an international organization was created (IAATO, www.iaato.org) to monitor and regulate tourist activity in the region. Their reports reveal a strongly increasing number of tourist visits from 12 248 to about 46 000 between the 2001/2002 and the 2007/2008 seasons, with an anticipation of further growth. Most of the tourists (98%) are ship borne.

A number of studies have been published concerning pollution due to research activity at stations. PM_{10} abundance measured in McMurdo during the austral summer

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routinely exceeds levels found in remote coastal regions of the continent (Lugar, 1993; Mazzera et al., 2001). Another study (Wolff and Cachier, 1998) conducted during 1992-95 measured BC at Halley research station and found a mean background level of approximately 1 ng BC per m^3 air, but spikes of several hundred ng BC per m^3 when the generators (350–400 m away) were downwind. It was suggested that the annual cycle of BC was indicative of control by the timing of biomass burning in the tropics, strongly modulated by the efficiency of transport to Antarctica. Other pollutant sources include combustion of fuel for transport and energy production, sewage burning (banned after 1997) and accidental oil spills (Barggali, 2008), for example from shipping accidents such as befell the Antarctic cruise vessel "Explorer" in 2007. All these studies concerned point measurements in the direct neighbourhood of research stations (for a comprehensive review see (Tin et al., 2009)), and we are unaware of any study of continental scale contamination. Only very recently was a first comprehensive emission inventory published for 2004/2005 (Shirsat and Graf, 2009).

Natural sources of pollution include Mt. Erebus (height 3794 m) on Ross Island, which has probably been degassing continuously from its active lava lake since at least its discovery in 1972 (Giggenbach et al., 1973). Between 1995 and 2002, the average of all SO_2 flux measurements for the volcano was 61 Mg d^{-1} (~1860 Mg $month^{-1}$). For December 2003, measurements of both SO_2 (74 Mg d^{-1}) and NO_2 (~5.2 Mg d^{-1}) were reported (Oppenheimer et al., 2005). Several sporadically active volcanoes are located in the outer South Sandwich and Shetland Islands, but their emissions have not been measured and they are not taken into account here.

Oceanic and coastal biota (mainly penguins) also contribute to atmospheric pollution as does the emission of NO_x from snow pack (Wang et al., 2008). Atmospheric measurements near several penguin colonies revealed 0.05–0.15 $\mu g\, m^{-3}$ of sulphate and 15–38 ng m^{-3} methanesulphonate (Minikin et al., 1998). Marine bioactivity leads to the emission of dimethylsulphate (DMS), which oxidises to SO_2 and, ultimately to SO_4 in the atmosphere. Estimates of source strength range between 1.4 and 2.7 Mt S per year for the Southern Ocean (Berresheim, 1987; Curran and Jones, 2000) but their trans-

formation into sulphur species and subsequent deposition is highly uncertain (Boucher et al., 2003). In any case these sources by far exceed anthropogenic emissions of sulphur species and mask the contribution from anthropogenic sources. In order to determine the patterns of anthropogenic and volcanic sulphur deposition we therefore exclude DMS sources from our study.

The weather and climate of Antarctica are dominated by strong katabatic winds blowing from the continent's interior towards the open ocean and by strong westerlies with embedded synoptic storms encircling the continent between 60° S and 40° S, making the region extremely windy, cloudy and precipitation rich. Most human activity is concentrated in coastal areas with relatively easy access in summer, and these weather conditions tend to hinder transport of airborne pollutants towards the interior of the continent.

2 Emission inventory

We use a new emissions inventory (Shirsat and Graf, 2009) in a model simulation of transport, chemical decomposition and deposition of sulphur compounds and BC (from ship emissions only) for the year 2004/2005. The emissions from each source were tagged to allow comparison of the contribution from each source type of the inventory (station activity, ground transport, aviation, ships, and emissions from Mt. Erebus) to the total atmospheric concentration and deposition. Station activity including ground transport contributes 158 Mg of SO_2 for April 2004 to March 2005, very close to an earlier estimate (Boutron and Wolff, 1989) by 18 Mg. Aircraft operations contribute 56 Mg of SO_2 at varying altitudes but exclusively during the Antarctic summer.

By far the highest anthropogenic source during the 2004/2005 summer season are ships: 3873 Mg of SO_2 and 14.5 Mg of BC. This is 40 times more SO_2 than the estimated total shipping source south of 60° S in 1989 (Boutron and Wolff, 1989)! On the other hand, Bond et al. (2007) estimate the total BC emission from ships south of 60° S, including fishery activities, to 100 Mg per year, an amount exceeding our BC

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emission data, which are only based on tourist and scientific cruises, by a factor of ~7. The number of ship visits to Antarctica, including scientific missions and tourism, is rising. In the Antarctic summer (November to March) 2004/2005, 65% of all ships carried tourists (Shirsat and Graf, 2009) and their number rose from 210 trips in 2004/2005, our study period, to 301 trips in the 2007/2008 austral summer, an increase of 43% within three years.

Human activity on the continent is highly localised around bases, with the highest populations found on the Antarctic Peninsula, in the Ross Sea area (Ross Island and Terra Nova Bay), Queen Maud Land and around the Amery Ice Shelf. Even though the sulphur emission of Erebus volcano is more than six times higher than all anthropogenic emissions, the crater is ~3800 m above sea level allowing winds to dilute and disperse the plume over a much greater area than the near-ground emissions.

Estimates of total emissions of SO_2 from different source types (see Shirsat and Graf, 2009) are shown in Table 1. During the austral winter, station activity is reduced and ship operations and aviation cease. Station emissions were distributed to the base locations and, where known, to transport routes according to available information. In many cases, only the seasonal total of burned fuel was available. If one nation operates several stations, national total fuel use was then distributed on a *per capita* basis to the station locations. Ship routes and mooring times were obtained from a number of different sources, including the on-line ship tracking system (www.sailwx.info) and sea ice maps. Air operations peak in January and February when research activity is highest. They had to be estimated from rather crude information obtained from COMNAP and personal communications.

As seen from Table 1, ship emissions dominate anthropogenic sources during the Antarctic summer. The busiest month is December when, in 2004, SO_2 emission was comparable to volcanic emissions. Considering the dramatic >40% increase in tourist activity between 2004 and 2007 it is possible that December ship emissions today exceed those from Mt. Erebus.

3 Model simulations

To estimate the contribution of each source to the total sulphur deposition and atmospheric concentration, we ran the 20 layer limited area model REMOTE (Langmann, 2000) at 0.5° resolution from April 2004 to March 2005. The model meteorology was driven at the lateral boundaries every 6 h by ECMWF ERA40 reanalysis data (Uppala et al., 2005) after initialization at all grid points for 1 April 2004. Gas phase RADM2 (Stockwell et al., 1990) sulphur chemistry was included and wet (Walcek and Taylor, 1986) and dry (Wesley, 1989) depositions were calculated. We concentrate on anthropogenic effects from point and line sources (mainly stations and ships) and Erebus volcano and did not take into account the uncertain background emissions and transformation pathways of DMS in our model study. These emissions potentially lead to much higher atmospheric concentrations of sulphur compounds and would mask the anthropogenic and volcanic contributions if they were included in the model calculations. The model was applied over Antarctica for the first time and so we checked winds and temperatures against observations and reanalysis data. The results showed overall good agreement and made us confident that the model is a useful tool for our task.

The mean lifetime of SO_2 was estimated from the model calculations as 2 weeks in summer and 3 weeks in winter, comparable to estimates for the Arctic (Barrie and Hoff, 1984). Figure 1 shows December 2004 atmospheric mixing ratios of SO_2 in the lowest model level (ca. 50 m height above surface) originating from (a) station activity and ground traffic, (b) ships and (c) Erebus volcano. Clearly the highest abundance of SO_2 from research activity is found where most of the stations are situated, i.e., around the largest station (McMurdo) in the Ross Sea, and on Amery Ice Shelf and in Queen Maud Land, where clusters of bases operate. Weather conditions over the Peninsula, where further research bases are found, reduce the build up of SO_2 abundance. Most tourist ships are bound for the Peninsula region since its attractions are most easily accessible. SO_2 abundances from ship operation are thus especially high in this area, contributing at least ten times as much as research bases. Emissions from Erebus

volcano reveal high surface SO_2 abundance extending from the crater Island to adjacent areas of the Ross Sea and Transantarctic Mountains. Because of the high summit elevation, they are distributed more evenly and over a wider area than the near surface emissions from ships and station activity.

Clearly, even the highest simulated grid cell means of atmospheric SO_2 abundances from anthropogenic sources are well below observed local values elsewhere in the world, including the Arctic. In Nuuk, Greenland, between February 2002 and August 2004 mean SO_2 concentrations of 0.44 $\mu g\,m^{-3}$ were reported (Skov et al., 2006), an order of magnitude higher than the maximum values we simulated for Antarctica from emissions of research bases and ship operation (Peninsula 0.035$\mu g\,m^{-3}$ and Ross Island 0.057$\mu g\,m^{-3}$). Mt. Erebus contributes significantly to the SO_2 abundance in the Ross Sea area in the order of 0.020 $\mu g\,m^{-3}$. Aviation related atmospheric sulphur concentrations remain several orders of magnitude below all other sources. We have to keep in mind, however that sulphur from the oceanic DMS source is not included in the simulations and that the model results represent means for areas of about 2500 km^2 and cannot, therefore, capture local maxima, especially in the vicinity of strong point sources. Observations (Wolff and Cachier, 1998) revealed an average SO_2 concentration of 0.74 $\mu g\,m^3$ near Mc Murdo Station during the austral summers 1995/1996 and 1996/1997. This point measurement exceeds the Nuuk observations, and represents a much higher concentration than our model simulations yield for the area mean. Observed atmospheric concentrations representative of the scale of our model are not available, but since all relevant emission sources are point or line (cruising ships) sources, variations of one to two orders of magnitude can be expected within the model resolved grid cells.

Melted snow is the exclusive source of fresh water in Antarctica and therefore deposition of sulphur is important. SO_2 and SO_4 deposition may also serve as an indicator for the deposition of other, more toxic substances emitted simultaneously especially from ship diesel engines. During melting or by evaporation from the snow pack, any pollutants become more concentrated enhancing the potential to affect ecosystems.

The total modelled deposition of sulphur (from SO_2 and SO_4) and of BC from ship exhaust during the year 2004/2005 is plotted in Fig. 2. BC deposition reaches its highest rates at the outer Peninsula, where most shipping activity takes place. The maximum local concentrations of BC in snow just reach the order of 1 ng g^{-1} BC in snow. The reduction in albedo of snow is 1.5% for 10 ng g^{-1} of BC in snow (Hansen and Nazarenko, 2004), so the present pollution can only affect snow albedo marginally. However, with further increases in shipping in the Peninsula region, significant effects on snow albedo may ultimately arise. Most of the deposition of sulphur takes place into the Southern Ocean since the prevailing winds blow the pollutants away from the continent. However, maximum deposition rates exceeding 0.4 kg S km^{-2} are found along the coast of Ross Sea (0.54 kg S km^{-2}) and stretching from the tip of the Antarctic Peninsula towards the Southern Ocean (0.85 kg S km^{-2}). The western hemisphere experiences generally higher deposition rates and the interior of the continent remains nearly unaffected.

There are large differences between winter and summer deposition patterns resulting from the seasonal contribution from anthropogenic sources, but also due to seasonal differences in weather conditions and sulphur photochemistry (Fig. 3). Sulphur deposition is due mainly to dry deposition of SO_2 and wet deposition of SO_4. From near surface sources (ships and stations) SO_2 is deposited closer to the source and the amounts follow the cycle of activity with very low values in winter and higher ones in summer. Erebus emissions are distributed over a much wider area, but over the Royal Society mountain range considerable amounts of volcanic sulphur are deposited on snow by dry and wet processes. Since oxidation of SO_2 to SO_4 is reduced at low temperatures and in the absence of sunlight, in winter, SO_4 from Erebus is barely formed before it reaches latitudes north of 60° S, where it is then rapidly rained out over the open ocean. During summer, ship emissions clearly dominate the deposition of sulphur both in the Ross Sea area and to the north of the Peninsula as well as over large parts of the western Southern Ocean, wet deposition being the main process. We find two maxima, one over the Ross Sea and adjacent parts of the Southern Ocean, which originates from ship emissions and from Mt. Erebus, and another stretching from the

northern tip of the Peninsula into the Southern Ocean, where 98% results from ships and station activity. Aircraft do not contribute in any measurable way to surface deposition of sulphur species.

Figure 4 summarizes contributions from all sources to the total sulphur deposition at the outer Peninsula (65° S, 65° W), Ross Island (77° S, 168° E), Queen Maud Land (70° S, 11° E), Amery Ice Shelf (69° S, 71° E), and South Pole for winter and summer. While in winter, Erebus is dominant at Peninsula, Ross Island and Amery Ice Shelf, it has practically no effect at South Pole and Queen Maud Land, where station emissions determine the very small deposition rates of sulphur. In summer, except for South Pole, ship emissions make by far the biggest contribution to sulphur deposition, with station emissions also a relevant source. Erebus, the biggest single sulphur source is of relative importance only at South Pole, where deposition rates are very small, and on Ross Island. Overall, pollution levels in Antarctica do not yet exceed any critical thresholds, as is already the case in the Arctic, but the rapid growth of human activity, especially mass tourism, may change this in the foreseeable future.

4 Conclusions

Model simulations for the year 2004/2005 indicate ships as the most significant source of sulphur and black carbon contamination during the austral summer, exceeding the natural source, Mt. Erebus. Most of these ships are tourism related and the most affected areas are the outer Antarctic Peninsula, a main destination of tourist cruises, and the Ross Sea, where the biggest research station operates. The simulated deposition patterns will help determine locations where snow samples can be collected to analyze large scale pollution in more detail.

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Table 1. Emissions from sulfur inventory (6) in Mg SO_2 month^{-1}.

Source	Station	Ship	Aircraft	Erebus
Apr	4	–		2229
May	4	–		2229
Jun	4	–		2229
Jul	4	–		2229
Aug	4	–		2229
Sep	4	–		2229
Oct	4	–		2229
Nov	26	223	6.4	2229
Dec	26	1971	5.6	2229
Jan	26	769	21.3	2229
Feb	26	693	21.2	2229
Mar	26	217	1.3	2229

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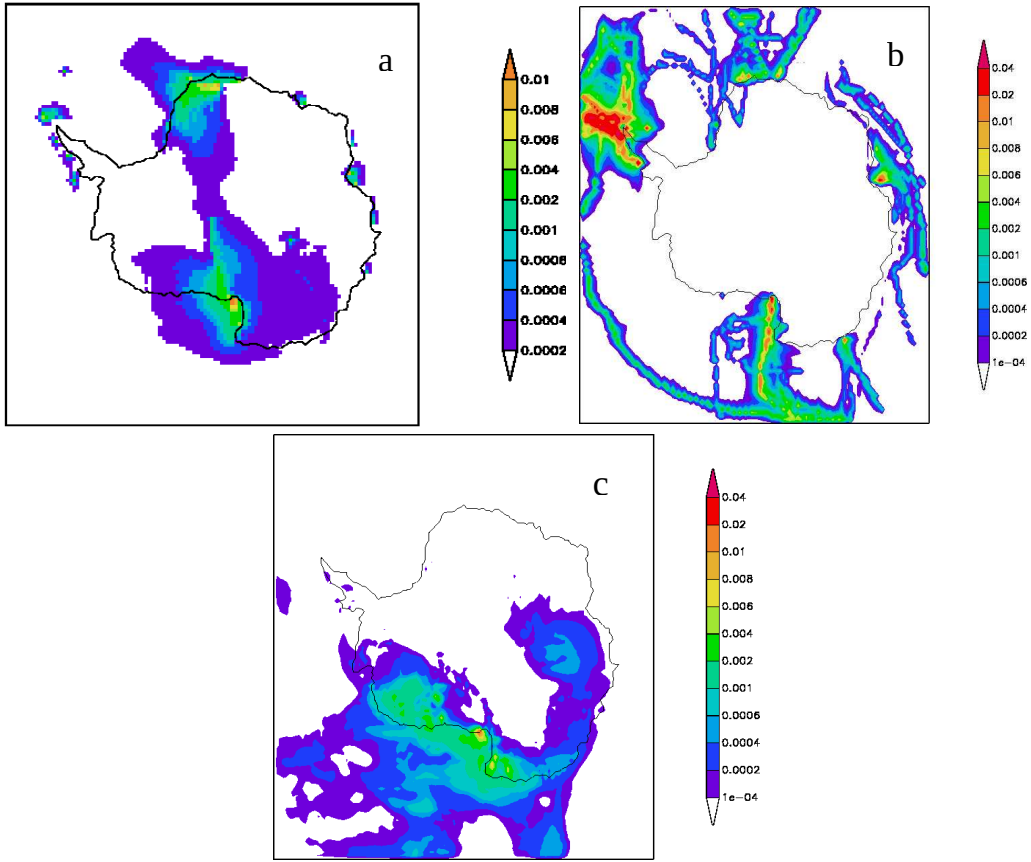

Fig. 1. Monthly mean December 2004 SO_2 mixing ratio in ppbv at lowest model level from: **(a)** power generation and vehicle operation at and between stations, **(b)** ship emissions and **(c)** volcanic emissions.

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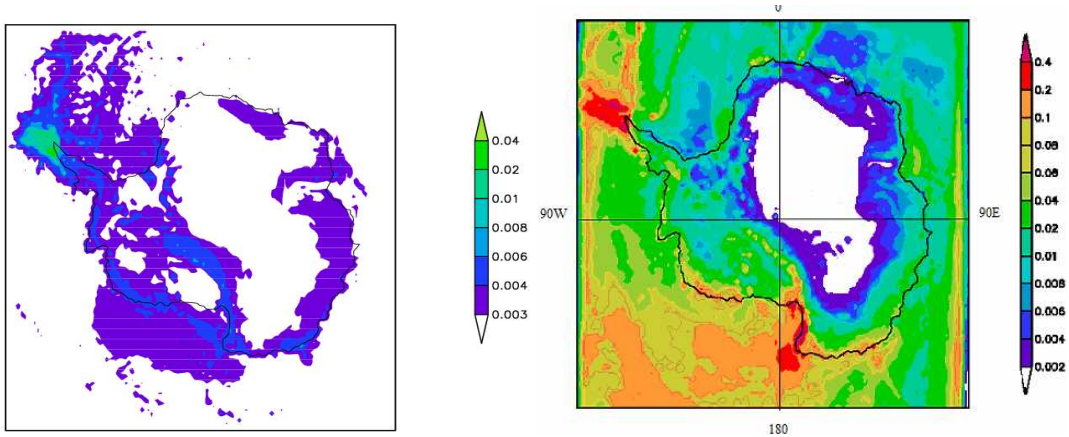

Fig. 2. Annual deposition in kg km^{-2} of Black Carbon (BC) from ships (left) and of Sulfur from stations, volcano and ships (right).

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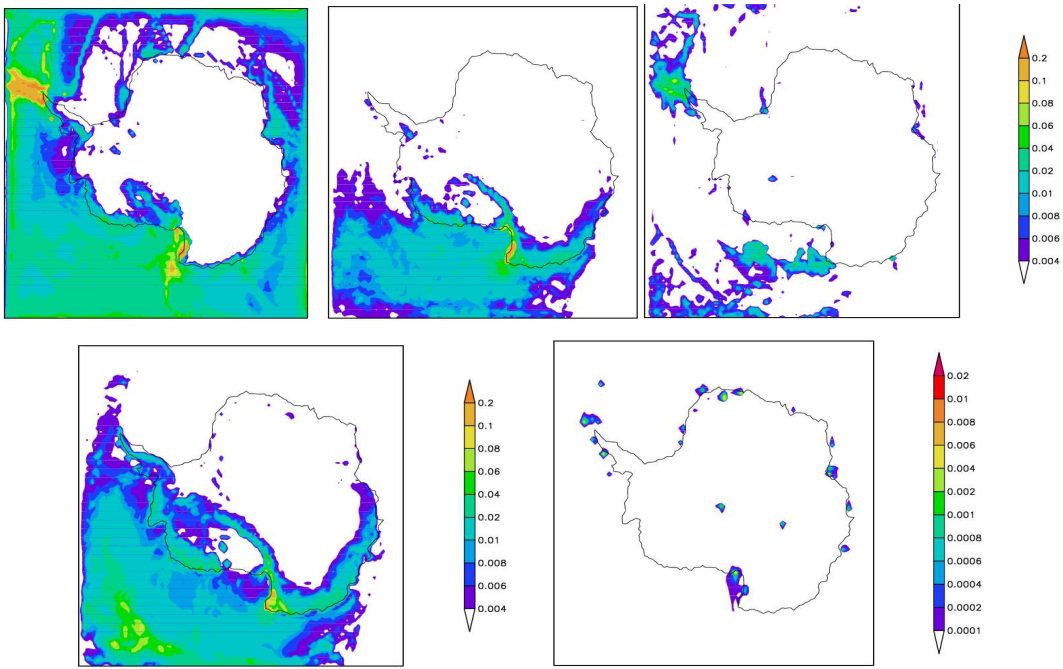

Fig. 3. (top row) Sulfur deposition (wet + dry) in kg km^{-2} from Ships (left), Erebus volcano (middle) and station activity (right) in kg S km^{-2} during Antarctic summer. (bottom row) Sulfur deposition (wet + dry) in kg km^{-2} from Erebus volcano (left) and station activity (right) in kg S km^{-2} during Antarctic winter.

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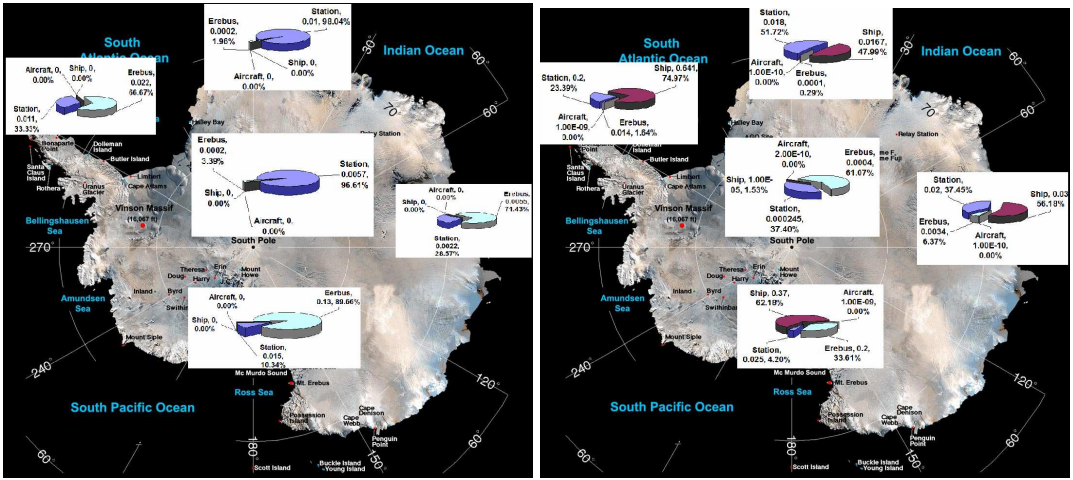

Fig. 4. Relative contribution of each emission source towards total (excluding DMS contribution) S deposition during winter (left) and summer (right) season of 2004/2005.

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