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Lead pollution in Antarctic surface snow revealed along the route of the International Trans-Antarctic Expedition

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ABSTRACT. This paper reports the lead concentration and flux (where accumulation rate is available) along the route of the 1990 International Trans-Antarctic Expedition. The lead concentration in Larsen Ice Shelf and Antarctic Peninsula, the western part of the route, was 7.4 ± 4.1 pg g^{-1}. The lead concentration in East Antarctic snow (South Pole to Mirny station) was 2–3 times higher than that in West Antarctica (Larsen Ice Shelf to South Pole). Taking into account the difference in site conditions, the difference between the above value over this area in 1989 and the value of 6.3 ± 3.3 pg g^{-1} at a site within this area in 1980 (Wolff and Peel, 1985) is not significant. Because the relative contribution of soil dust, volcanoes and the oceans to lead concentration in Antarctica is about 0.5 pg g^{-1} under modern climatic conditions (Boutron and Patterson, 1987), it is believed that the lead in Antarctic surface snow is dominated by pollution input. The lead-concentration increase from west to east over the trans-Antarctic route suggests that remote Antarctica has been impacted by anthropogenic activities. The lowest lead flux (0.064 ng cm^{-2} a^{-1}) was on the Antarctic Plateau, mainly reflecting the background global pollution. The mean flux of 0.273 ng cm^{-2} a^{-1} in the western part of the route (Larsen Ice Shelf to the Ellsworth Mountains) may result from the pollution input from the Southern Hemisphere. In addition to the influence of global and/or hemispheric pollution, local activities (notably the use of leaded gasolene) appear to have affected the region from Pionerskaya to Mirny.

INTRODUCTION

Considerable effort has been made during the past 30 years to measure the concentrations of heavy metals, especially the toxic metals lead, cadmium and mercury, in polar ice (Murozumi and others, 1969; Wolff and Peel, 1985; Boutron and others, 1991; Suttie and Wolff, 1992). Such studies offer a unique opportunity to reconstruct the temporal evolution of the atmospheric concentrations of these species. In recent years, special attention has been paid to the investigation of lead concentration in Antarctic snow and ice because it is a sensitive indicator of anthropogenic activity (Scarponi and others, 1997). Unfortunately, much of the pre-mid-1980s data in the literature is now considered unreliable (Peel, 1989; Wolff, 1990; Boutron and others, 1994). Peel (1989) analyzed the lead-concentration data taken before 1990 in ancient and present precipitation in Antarctic and Arctic regions, and believed that most of the data suffered from contamination. Consequently, reliable lead data from the Antarctic ice sheet are extremely rare. In order to obtain reliable data, extreme precautions must be taken throughout sampling, transportation and analysis procedures, and sensitive analytical techniques are necessary. To date, the reliable data largely cover only a few years (around 1980) at sites along the coast of Antarctica (Wolff and Peel, 1985; Boutron and

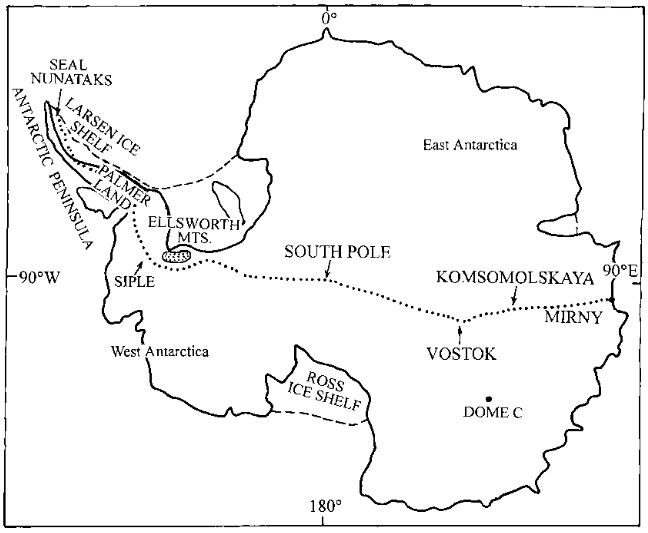

Fig. 1. Sketch map showing the sampling sites along the 1990 ITAE route. The expedition started on 28 July 1989 and ended on 3 March 1990, covering 5896 km in 220 days. It crossed major geographic zones of Antarctica, including the Larsen Ice Shelf, the Antarctic Peninsula, the Ellsworth Mountains and the Antarctic Plateau. Sampling sites were located along the route every 60 km.

Table 1. Pb concentration and flux of 25 cm surface snow and interrelated data along the 1990 ITAE route

Sampling site	*Sampling date*			*Position*	*Accumulation*	*Pb concentration*	*Pb flux*
	dd	mm	yy		$\mathrm{g\,cm^{-2}\,a^{-1}}$	$\mathrm{pg\,g^{-1}}$	$\mathrm{ng\,cm^{2}\,a^{-1}}$
1	27	07	89	65°05′ S, 59°35′ W			
2	28	07	89	65°10′ S, 59°59′ W			
3	29	07	89	65°17′ S, 60°19′ W			
4	30	07	89	65°27′ S, 60°45′ W			
5	01	08	89	65°35′ S, 61°10′ W		19.4	
6	02	08	89	65°42′ S, 61°32′ W			
7	03	08	89	65°50′ S, 61°57′ W			
8	10	08	89	66°20′ S, 62°36′ W		7.7	
9	13	08	89	66°54′ S, 63°32′ W		4.1	
10	15	08	89	67°20′ S, 64°07′ W		4.6	
11	18	08	89	67°47′ S, 64°32′ W	60.0	4.0	0.237
12	23	08	89	68°25′ S, 65°15′ W		13.0	
13	27	08	89	68°43′ S, 65°26′ W		6.2	
14	28	08	89	68°48′ S, 65°22′ W			
15	29	08	89	68°57′ S, 65°26′ W			
16	30	08	89	69°04′ S, 65°20′ W			
17	31	08	89	69°10′ S, 65°18′ W		5.2	
18	05	09	89	69°41′S, 65 813′ W	16.4	7.5	0.124
19	09	09	89	70°25′ S, 64°44′ W	16.4	5.9	0.097
20	14	09	89	70°58′ S, 64°41′W		6.5	
21	15	09	89	71°17′ S, 65°43′ W		4.2	
22	17	09	89	71°56′ S, 65°18′ W		14.3	
23	18	09	89	72°14′ S, 65°29′ W		3.1	
24	19	09	89	72°33’ S, 65°55′ W		4.6	
25	23	09	89	73°14′ S, 66°48′ W		4.5	
26	30	09	89	73°56′ S, 67°30′ W	50.0		
27	06	10	89	74°05′ S, 69°57′ W	22.8	6.8	0.156
28	09	10	89	74°46′ S, 72°46′ W	25.1	9.6	0.242
29	11	10	89	74°51′S, 75°45′ W	26.5	6.4	0.169
30	15	10	89	75°26′ S, 78°29′ W	22.8	8.8	0.201
31	18	10	89	75°47′ S, 81°40′ W		7.5	
32	24	10	89	76°25′ S, 85°10′ W	34.4	19.7	0.678
33	26	10	89	76°56′ S, 86°15′ W		8.7	
34	28	10	89	77°39′S, 87°07′ W		5.2	
35	30	10	89	78°12′ S, 87°39′ W	40.0	14.8	0.592
36	01	11	89	78°49′ S, 87°14′ W	30.0	4.0	0.121
37	02	11	89	79°10′ S, 86°51′W		15.1	
38	04	11	89	79 ⩽ 45′ S, 85°18′ W	20.0	24.8	0.496
39	09	11	89	80°18′ S, 81°21′W		10.7	
40	11	11	89	80°45′ S, 81°15′ W		9.7	
41	14	11	89	81°12′ S, 82°00′ W		10.0	
42	16	11	89	81°48′ S, 82°50′ W			
43	17	11	89	81°55′ S, 83°20′ W		8.8	
44	19	11	89	82°41′S, 84°00′ W		7.7	
45	20	11	89	83°05° S, 84°45′ W		12.1	
46	22	11	89	83°47′ S, 87°25′ W	15.0	10.9	0.164
47	23	11	89	84°12′ S, 88°05′ W		17.0	
48	24	11	89	84°35′ S, 88°55′ W		18.8	
49	26	11	89	85°11′Σ, 88°58′ W		5.4	
50	29	11	89	85°53′ S, 88°10′ W		19.9	
51	30	11	89	86°12′ S, 88°25′ W	10.0	9.8	0.098
52	02	12	89	86°34′ S, 88°57′ W		14.9	
53	03	12	89	86°54′ S, 90°19′ W		10.7	
54	05	12	89	87°36 ’ S, 91°06′ W		12.8	
55	06	12	89	87°57′ S, 91°55′ W	8.2	18.5	0.152
56	08	12	89	88°38′ S, 92°26′ W	8.0	21.0	0.168
57	09	12	89	89°00′ S, 92°58′ W	7.6	8.6	0.065
58	10	12	89	89°22′ S, 91°39′ W	8.1	4.2	0.034
59	13	12	89	90°00′ S	8.5	6.7	0.057
60	16	12	89	89°53′ S, 114°22′ E	8.1	11.6	0.094
61	17	12	89	89°32′ S, 108°18′ E	7.4	8.2	0.061
62	18	12	89	89°11′S, 105°35′ E	6.8	9.8	0.067
63	20	12	89	88°26′ S, 104°27′ E	5.6	8.1	0.045
64	21	12	89	88°03′ S, 104°35′ E	5.0	22.4	0.112
65	22	12	89	87°42′ S, 104°39′ E	4.9	7.9	0.039
66	23	12	89	87°20′ S, 104°25′ E	4.8	30.6	0.147
67	25	12	89	86°36′ S, 104°57′ E	4.6	18.3	0.084
68	29	12	89	85°33′ S, 105°40′ E	4.3	15.1	0.065
69	30	12	89	85°13′ S, 105°49′ E	4.2	5.6	0.024
70	01	01	90	84°28′ S, 106°17′ E	4.0	7.7	0.031
71	02	01	90	84°07′ S, 106°17′ E	3.9	14.8	0.058
72	03	01	90	83°44′ S, 106°24′ E	3.8	8.8	0.033
73	05	01	90	83°00′ S, 106°12′ E	3.6	11.8	0.042
74	06	01	90	82°40′ S, 106°19′ E	3.5	11.6	0.041
75	09	01	90	81°50′ S, 106°28′ E	3.3	9.8	0.032
76	11	01	90	81°05′ S, 106°26′ E	3.1	14.0	0.043
77	12	01	90	80°42′ S, 106°12′ E	3.0	16.8	0.050
78	13	01	90	80°17′ S, 106°14′ E	2.9	18.4	0.053
79	14	01	90	79°42′ S, 106°04′ E	2.7	28.9	0.078
80	16	01	90	79°08′ S, 106°08′ E	2.5	8.1	0.020
81	17	01	90	78°46′ S, 106°41′E	2.3		
82	22	01	90	78°07′ S, 105°47′ E	2.3	12.7	0.029
83	23	01	90	77°43′ S, 104°47′ E	3.0	9.6	0.029
84	25	01	90	77°00′ S, 102°55′ E	3.3	29.8	0.098
85	26	01	90	76°36′ S, 102°00′ E	3.6	8.6	0.031
86	28	01	90	75°54′ S, 100°31′E	3.7	19.9	0.074
87	29	01	90	75°33′S, 100°31′E	3.7	25.5	0.094
88	31	01	90	74°44′ S, 98°41′E	3.7	9.8	0.036
89	01	02	90	74°21′S, 98°00′ E	6.6	33.9	0.224
90	04	02	90	73°41′S, 97°26′ E	6.1	37.1	0.226
91	05	02	90	73°18′ S, 97°09 ’ E	7.1	5.8	0.041
92	06	02	90	72°51′S, 96°59′ E	7.8	24.8	0.193
93	07	02	90	72°28′ S, 96°45′E	9.1	26.8	0.244
94	09	02	90	71°42′ S, 96°17′ E	12.3	10.4	0.128
95	10	02	90	71°20′ S, 96°01′E	12.3	28.1	0.346
96	11	02	90	70°57′ S, 95°54′ E	12.9	3.1	0.039
97	12	02	90	70°33′ S, 95°43′ E	12.6	29.4	0.370
98	14	02	90	70°11′S, 95°35′ E	16.0	26.7	0.427
99	15	02	90	69°46′ S, 95°22′ E	12.6		
100	18	02	90	69°26′ S, 95°07′ E	11.2	12.9	0.144
101	21	02	90	68°51′S, 94°37′ E	10.8	9.2	0.099
102	23	02	90	68°05′ S, 93°50′ E	18.9	35.7	0.675
103	24	02	90	67°42′ S, 93°39′ E	40.3	47.3	1.906
104	25	02	90	67°21′S, 93°26′ E	49.0	25.2	1.235

Patterson, 1987; Scarponi and others, 1997). Snow–firn cores collected at D55, East Antarctica, reveal lead concentrations over the last two centuries (Boutron and Patterson, 1983) and the period 1940–80 (Görlach and Boutron, 1992). A longer record is available from Dome C and Vostok station, but with only sparse sampling (Boutron and Patterson, 1986; Boutron and others, 1988, 1990) over the past 27 ka BP and the last climatic cycle. Therefore, there is a gap in our understanding of lead-concentration distribution over the whole of Antarctica. Glaciological investigation along the International Trans-Antarctic Expedition (ITAE) route in 1990 offered a unique opportunity to study the geographic distribution and environmental significance of lead in surface snow over the Antarctic ice sheet.

EXPERIMENTAL PROCEDURES

Between 27 July 1989 and 3 March 1990, 25 cm surface snow samples at 97 sites along the 5896 km ITAE route were collected in polyethylene bottles (Fig. 1). The bottles were washed three times using Milli-Q-Water, and air-dried in a class 100 ultra-clean laboratory. The washed bottles were packed in clean bags in a clean room and opened only during sampling. To minimize contamination, the sample collector was dressed in full clean clothing (coverall, hood, boots and polyethylene gloves) throughout sampling. All samples were kept frozen during transport. Lead concentration was meas-

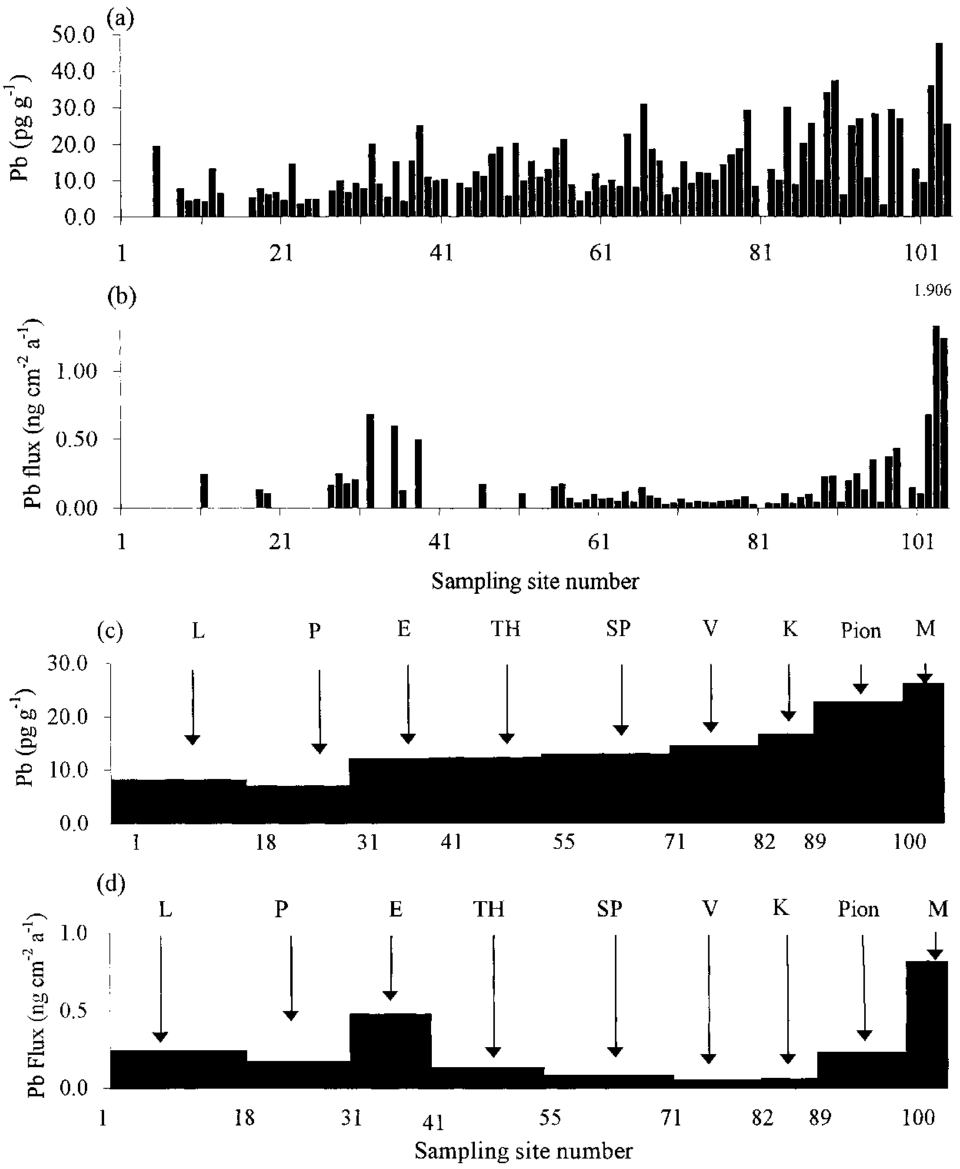

Fig. 2. Distribution of lead concentration and flux along the ITAE route. (a) Pb concentration (site by site); (b) Pb flux (site by site); (c) the mean Pb concentration of different geographic units; (d) the mean Pb flux of different geographic units. L, Larsen Ice Shelf; P, Antarctic Peninsula; E, Ellsworth Mountains; TH, Thiel Mountains;, SP, South Pole; V, Vostok; K, Komsomolskaya; Pion, Pionerskaya; M, Mirny.

ured by laser excitation atom fluorescence spectroscopy (LEAFS) analysis in a class 100 ultra-clean room in the Laser Single Atom Detection Laboratory, Department of Modern Applied Physics, Tsinghua University, Beijing, China. Each sample was measured seven times (discarding the maximum and minimum values, then averaging) and the standard was measured before and after sample measurements. The detectivity of this technique for lead is 0.2 pg g^{-1}, and the accuracy is about 10%. The laboratory blank value for lead is about 2 pg g^{-1}; the reported concentrations have the blank value subtracted (Qin Dahe and others, 1995; Xue Meng and others, 1997).

RESULTS

Lead concentrations at sites 1 and 7 (located on the Larsen Ice Shelf in the north of the Antarctic Peninsula) were about 1000 and 64 pg g^{-1}, respectively. These values are much higher than those for surrounding sites. An alloy knife was used for snow-pit sampling at sites 1–7 because the snow layer was hard in this area. This may have resulted in the elevated lead concentrations. Ninety-one reliable Pb samples were obtained, with a mean of 13.7 pg g^{-1}, a median of 10.4 pg g^{-1}, a range of 3.1–47.3 pg g^{-1} and a standard derivation of 9.0 pg g^{-1}. There were ten samples whose lead concentration was <5 pg g^{-1}, 35 samples between 5 and 10 pg g^{-1}, 29 samples between 10 and 20 pg g^{-1}, and 17 samples higher than 20 pg g^{-1}, accounting for 11%, 38%, 32% and 19% of all samples, respectively. The lead flux was calculated for those sampling sites where accumulation rate was available, by multiplying the lead concentration and the accumulation rate. Lead-concentration and flux data are reported in Table 1, and plotted in Figure 2a and b, respectively. In order to determine the regional values of lead, the ITAE route was divided into nine segments according to geographic character (Table 2). Mean concentrations of each segment are listed in Table 2 and plotted in Figure 2c. The mean lead concentration from the Ellsworth Mountains segment to the South Pole segment (12.3 pg g^{-1}) is about double that of the Larsen Ice Shelf and Antarctic Peninsula segments (7.4 pg g^{-1}), while the mean lead concentration of Pionerskaya and Mirny seg-

Table 2. Mean Pb concentration and flux in nine segments along ITAE route

Segment name	*Sampling-site Nos.*	*Pb* pg g^{-1}	*Number of data*[*]	*Pb flux* ng cm^{-2} a^{-1}	*Number of data*[†]
Larsen Ice Shelf	1–17 (1,7 crossed out)	8.0 ± 5.5	8	0.237	1
Antarctic Peninsula	18 –30	6.9 ± 3.0	12	0.165	6
Ellsworth Mountains	31 –40	12.0 ± 6.6	10	0.472	4
Thiel Mountains	41 –54	12.2 ± 4.3	13	0.131	2
South Pole	55 –70	12.8 ± 7.4	16	0.078	16
Vostok	71 –81	14.3 ± 6.1	10	0.045	10
Komsomolskaya	82 –88	16.6 ± 8.5	7	0.056	7
Pionerskaya	89 –99	22.6 ± 11.9	10	0.224	10
Mirny	100 –104	26.1 ± 15.8	5	0.812	5

[*] Used for calculating the segment mean concentration.
[†] Used for calculating the segment mean flux.

ments (24.4 pg g^{-1}) is three times that of the Larsen Ice Shelf and Antarctic Peninsula segments. It is clear that the lead concentration increases continuously from the Ellsworth Mountains to Mirny station.

The lowest lead flux (0.045 ng cm^{-2} a^{-1}) appeared in the Vostok segment of the route. The lead flux in the regions from Pionerskaya to Mirny station is around double that of the region from Larsen Ice Shelf to the Ellsworth Mountains (Table 2; Fig. 2d).

DISCUSSION AND CONCLUSION

Natural and anthropogenic sources can contribute to the lead concentration present in Antarctic snow. Boutron and Patterson (1987) reported that the relative contribution of soil dust, volcanoes and the oceans to measured lead in Antarctica is about 0.5 pg g^{-1}. If this is the case for the whole of Antarctica, our lead data indicate that most of the lead in the Antarctic precipitation is a result of human activity.

Previously published lead data for Antarctic surface snow are listed in Table 3. Boutron and Patterson (1987) collected large blocks of surface snow at South Pole and Terre Adélie, East Antarctica, and reported lead concentrations of 6.6, 7.4, 2.3 and 6.3 pg g^{-1} for sites D40, D47, D80 and South Pole, respectively. A 3 m snow pit was dug by Scarponi and others (1997) on the high plateau of Victoria Land, East Antarctica. They reported a lead-concentration decrease from 8.4 pg g^{-1} in 1986 to 2.5 pg g^{-1} in 1991 and a corresponding fallout-flux decrease from 0.184 to 0.034 ng cm^{-2} a^{-1}. The decreasing trend was assumed to be related to the decrease in the consumption of leaded gasolene in the Southern Hemisphere. Wolff and Peel (1985) reported a lead level of 6.3 ± 3.3 pg g^{-1} in present precipitation, probably representing just one snowfall in late January 1980 in southern Palmer Land. Suttie and Wolff (1992) obtained 2 year seasonal variations of lead in Antarctic snow through the detailed analysis of a 1.7 m snow pit recovered from Dolleman Island on the east coast of the Antarctic Peninsula in December 1985. The lead concentration was in the range 1–9 pg g^{-1} with a mean of 4.0 pg g^{-1}. Significant seasonality with an autumn/winter maximum is in accordance with variations in marine and crustal aerosol input ascribed to enhanced transport of atmospheric lead from the other continents to Antarctica during winter. In this work, the mean lead concentration for winter snow on the Larsen Ice Shelf and Antarctic Peninsula was 7.4 ± 4.1 pg g^{-1}. If seasonal variation and location are considered, the mean lead concentration for segments of Larsen Ice Shelf and Antarctic Peninsula is very close to the value of 6.3 ± 3.3 pg g^{-1} at southern Palmer Land in 1980. There are no obvious differences in lead concentration in the precipitation from 1980 to 1989 in the Larsen Ice Shelf and Antarctic Peninsula regions.

The reported lead concentrations at several different sites in Antarctica are quite similar (Table 3). All of the reported samples, except from South Pole, were collected from coastal regions. The lead concentration in coastal regions may not reflect the situation for the inland Antarctic Plateau.

The large seasonal changes in solar insolation reaching

Table 3. Available reliable Pb data for Antarctic snow

	Sampling site	*Location*	*Sampling date*	*Distance from coast* km	*Altitude* m	*Accumulation rate* g cm^{-2} a^{-1}	*Pb conc.* pg g^{-1}	*Source*
Antarctic Peninsula	Dolleman Island	70°35′ S, 60°56′ W	Dec. 1985	0	about 0	88	4.0	Suttie and Wolff (1992)
Antarctic Peninsula	Spaatz Island	72°53′ S, 74°41′ W	Jan. 1980	15	408	115	6.7 ± 2.4	Wolff and Peel (1985)
Antarctic Peninsula	Gomez Nunatak	74° S, 70° W	Feb. 1980	150	1100	88	6.3 ± 3.3	Wolff and Peel (1985)
Victoria Land	Hercules Névé	73°06′ S, 165°28′ E	Summer 1993–94	90	2960	17	6.1	Scarponi and others (1997)
							5.6 ± 2.1	Scarponi and others (1997)
Terre Adélie	Stake D40		3 Jan. 1983	33	848	63	6.6	Boutron and Patterson (1987)
Terre Adélie	Stake D47		5 Jan. 1983	103	1500	26	7.4	Boutron and Patterson (1987)
Terre Adélie	Stake D55	68°00′ S, 137°46′ E	Late Jan. 1980	180	2000	8	5.4	Görlach and Boutron (1992)
Terre Adélie	Stake D80	70°02′ S, 134°50′ E	14 Jan. 1983	433	2525	24	2.3	Boutron and Patterson (1987)
	South Pole	90°00′ S	18 Jan. 1984	1274	2880	8.5	6.3	Boutron and Patterson (1987)
Larsen Ice Shelf							8.0	This study
Antarctic Peninsula							6.9	This study
Ellsworth Mountains							12.0	This study
Thiel Mountains							12.2	This study
South Pole							12.8	This study
Vostok							14.3	This study
Komsomolskaya							16.6	This study
Pionerskaya							22.6	This study
Mirny							26.1	This study

the Antarctic ice surface modulate the intensity of the katabatic wind regime and thus the resulting mean meridional circulation between the Antarctic and subpolar latitudes. The mean meridional circulation is well defined throughout the year by (i) intense, shallow outflow presumably associated with the katabatic wind regime below approximately 700 hPa, (ii) rising motion just to the north of the continent at 60–65° S, (iii) a broad return branch from approximately 400 hPa to well into the stratosphere, and (iv) subsidence over the Antarctic. The mass redistribution extends to the subtropics of the Southern Hemisphere during austral autumn and spring transition periods (Parish and Bromwich, 1997). A series of aerosol measurements (Hogan, 1979) over Antarctica show that the most frequent transport route of aerosols to the polar plateau is near the lower troposphere to mid-troposphere, especially those layers below 400 hPa. In addition, there is a weak front over the Transantarctic Mountains at approximately 85° S, which divides extremely stable continental Antarctic cold air from peripheral Antarctic air. For coastal stations, katabatic winds tend to drive local pollution away from the continent, so few of these emissions should reach the continental interior, except when cyclonic systems penetrate the interior. On the high plateau, the presence of cold high pressures dramatically reduces the influence of oceanic air masses. The atmosphere over the high interior plateau may be considered to be a background atmosphere that is supplied by long-range transport of impurities from extra-Antarctic sites.

Scarponi and others (1997) reported that the lead concentration in a snow pit from Victoria Land (150 km from the coast) decreased during the period 1986–91. Based on the atmospheric circulation mentioned above, it is possible that the pollutants emitted from the Southern Hemisphere outside of the Antarctic reached the continent by meridional transport, especially through the lower to mid-troposphere. This may be the case for the region from the Larsen Ice Shelf to the Ellsworth Mountains. Lowest lead concentrations and high lead fluxes may be explained by the high accumulation rates in this region, because precipitation events may scavenge the impurities in the atmosphere effectively and dilute the concentration.

The region from the Thiel Mountains to Komsomolskaya is controlled by stable continental air, where oceanic air mass input is rare. When strong subsidence is present, the impurities in the upper troposphere and stratosphere may reach the surface, supplied by long-range transport. This may explain why the lead flux is very low (0.064 ng $cm^{-2} a^{-1}$) in this area, although the lead concentration is higher than in West Antarctica (from the Larsen Ice Shelf to the Ellsworth Mountains). The lead concentration increases from Vostok to Mirny and there is an accompanying dramatic increase of lead flux. It is well known that the 1450 km passage from Vostok to Mirny was an important land and air supply line for Soviet Antarctic expeditions. The use of gasolene and diesel oil in large motor vehicles, generators and airplanes results in enhanced lead concentration in this region. Moreover, the lead content in the gasolene utilized by the Soviet Union was 2.5 times higher than standard for Western countries. The atmospheric high-pressure system over the Vostok and Pionerskaya area favours the transport of local emissions into downwind areas. We suggest that the area from Vostok to Mirny has been polluted by local anthropogenic impacts more seriously than other areas.

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