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Spread of Lead Pollution over Remote Regions and Upper Troposphere: Glaciochemical Evidence from Polar Regions and Tibetan Plateau

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Metals captured in glaciers can be used as a tracer for air mass transportation, and in revealing the history and intensity of local or global pollution. Since 1970s, efforts on measurement of metals, mostly of Pb, have been made mainly for the polar snow and ice cores (Murozumi et al., 1969; Ng and Patterson, 1981; Wolff and Peel, 1985a; Boutron and Patterson, 1986; Peel, 1989; Wolff, 1990; Hong, 1994). During the past 30 years, Pb records recovered from polar regions have covered the whole industrial era and have found dramatically increasing Pb pollution since industrialization.

In Arctic, it is believed that real Pb concentration in ice layers for 1970s in Greenland is around 200×10^{-12}g.g^{-1}. However, investigations have shown that this concentration decreased in firn since 1980s. For instance, average Pb content in the snowfall at Dye 3 during 1983~84 was 28×10^{-12}g.g^{-1} (Wolff and Peel, 1988), while in central Greenland in 1987 it was 43×10^{-12}g.g^{-1}. The decrease in Pb since 1980s is the result of reduced utilization of Pb-containing gasoline, a policy initially promoted by USA and Canada, and subsequently by European countries.

In this paper, Pb content in snow of the early 1990s' from the Arctic, Antarctica and the Tibetan Plateau will be discussed, based on measurements along 1990 International Trans-Antarctic Expedition (ITAE) route (Qin et al., 1999), three glaciers on Tibetan Plateau as well as snowpack over the Canadian and central Arctic. We seek to highlight the human influence on atmosphere over the most remote regions and the upper troposphere (5000-7000m a.s.l.)

MATERIALS AND METHODS

All samples collected from the Canadian Arctic, Tibetan Plateau and the Antarctic Ice Sheet are snow pits samples (Figure 1). During March to May 1995, four snow pits were dug each at Hudson Bay, Point of Woods, Coinwallis Island and Ellesmere Island, NWT, Canada. Ten snow pits were excavated on pack ice between approximately 88°~90°N. During training at Ely, near USA and Canada border, samples in a 10cm-depth pit (single snowfall event) were collected on pack ice of White Iron Lake immediately after the end of the snowfall. Each pit

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was sampled at 5cm intervals down the pit wall. All 15 snow pits, except at Ely, cover the snowfall of one annual layer.

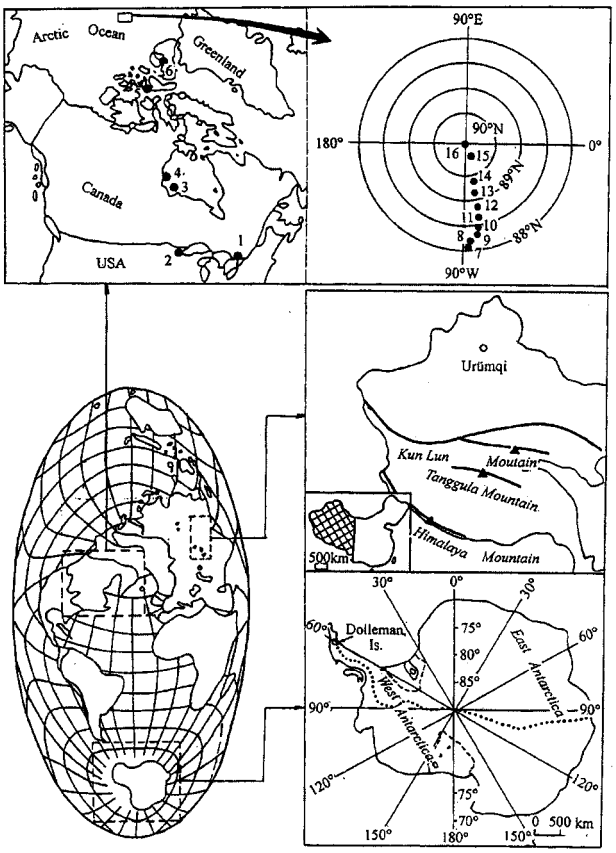

Figure 1. Locations of snow pits excavated in the Arctic (Site 2~16), the Tibetan Plateau (at three glaciers) and the Antarctic Ice Sheet (totally 93 sites along ITAE route)

Three snow pits were dug at each site at north, centre and south of the Tibetan Plateau. In August 1991, a 100cm-depth pit was dug on Kangwure Glacier (28°27′N, 85°45′E), middle Himalaya, at 6140m a.s.l. At Dongkemadi Glacier (33°04′N, 94°05′E) on Tanggula Mountain, the pit was 90cm deep excavated at 5700m a.s.l. in June 1993. The pit excavated at Meikuang Glacier (94°11′N 35°40′E) on East Kunlun Mountain in May 1994 was 60cm deep, at 5300m a.s.l. The three snow pits covered one annual layer of precipitation at each site. The samples were collected at 10cm intervals down each of the pit wall. Samples from Arctic also cover the snowfall of one annual layer.

Along the 5896km ITAE route, 93 snow pits, each 25cm deep were dug (for details see Qin et al., 1999). Reliable Pb data in 91 among the 93 pits were obtained. Samples at 25cm along ITAE represented 0.5-3 annual snowfall depending on the accumulation rate along the route.

To avoid contamination, sampling tools and procedures follow international restrictions for polar trace metals (Wolff and Peel, 1985b). Samples from the Arctic and Tibetan Plateau were measured in Finnigan MAT, Sola ICP-MS (detection limit for Pb was $<0.6\times10^{-12}g.g^{-1}$). RSD for Pb was found to be between 10%~17%, the blank values for sample container and pure water was less than $2\times10^{-12}g.g^{-1}$. Al and Na from the same samples were used as conservative tracers for calculating crustal Pb and sea salt Pb. Al was analyzed simultaneously by the same instrument as Pb, with RSD<4.6%. Na was analyzed by Dionex-300 ion chromatography. The detection limit was $<0.1\times10^{-9}g.g^{-1}$. Samples from 1990 ITAE were analyzed in the Laser Excitation Atom Fluorescence Spectroscopy Analysis (LEAF, Qin et al., 1995).

RESULTS AND DISCUSSION

The data on Pb along with sampling sites and date are tabulated in Table 1. Mean Pb concentration in ten snowpits from the central Arctic, representing snowfall of 1994/95, was $105.7\times10^{-12}g.g^{-1}$. Although this value was much lower than that in snowfall of the 1970s, it was still much higher than Pb concentration in 1980s snowfall in Greenland (Wolff, 1990). Pb input into the Greenland Ice Sheet has been reduced since the pollution control action initiated by U.S.A. and Canada, and later by European countries. By the square, these regions are the major pathways for air masses going northwards to Greenland. Nevertheless, air masses entering the Central Arctic Ocean have more passways such as central Eurasia, western North America and/or Siberia (Iversen, 1996), As some countries have not reduced the amount of gasoline-additives, This may have been the cause of high Pb concentration.

The distribution of Pb over northern Canada indicates that the Northwest Territory (NWT) of Canada is not an efficient channel for Pb transport to central Arctic. Pb concentrations display a decreasing trend from Ely to NWT Canada (Figure 2). However, further northward in central Arctic (88°~90°N), much higher values is found. It is clear that the high Pb input comes from other sources (mainly Eurasia) instead of NWT Canada.

Meikuang, Dongkemadi and Kangwure Glaciers locates at north, center and south of the Tibetan Plateau, respectively. Pb concentrations at Meikuang and Kangwure are higher than that found at Dongkemadi. It is suggested that the

Table1. Concentrations (include their ranges) of Pb in snow pit samples from Arctic, Qinghai-Tibetan Plateau and Antarctica.

Sampling sites #		Position	sampling date (D/M/Y)	Pit depths * (cm)	Pb (10^{-12}g·g^{-1}) (range)
Arct.	2	47°52′N, 91°48′W	14/Apr./1995	10 (2)	223.3(117.5~329.1)
	3	58°46′N, 94°10′W	07/Apr./1995	18 (4)	58.1(16.0~111.9)
	4	58°50′N, 94°12′W	09/Apr./1995	67.5 (13)	41.9(21.6~95.9)
	5	74°43′N, 94°59′W	07/May/1995	20 (4)	43.6(26.9~76.8)
	6	80°N, 85°51′W	06/May/1995	25 (5)	30.1(22.2~49.3)
	7	88°01′N, 82°29′W	24/Apr./1995	14 (3)	100.7(34.1~207.9)
	8	88°06′N, 82°37′W	25/Apr./1995	35 (7)	137.6(82.3~242.4)
	9	88°17′N, 81°40′W	26/Apr./1995	9 (2)	169.9(97.5~242.2)
	10	88°30′N, 80°29′W	27/Apr./1995	15 (3)	183.9(17.8~429.7)
	11	88°41′N, 79°25′W	28/Apr./1995	22 (4)	102.4(22.2~147.2)
	12	88°53′N, 80°32′W	29/Apr./1995	20 (4)	155.6(25.2~224.2)
	13	89°12′N, 70°22′W	02/May/1995	55 (11)	25.6(16.4~43.8)
	14	89°25′N, 69°52′W	03/May/1995	45 (9)	47.2(24.2~78.6)
	15	89°49′N, 72°31′W	05/May/1995	42 (9)	60.2(20.4~143.0)
	16	90° N	06/May/1995	60 (12)	73.8(17.3~166.7)
T.Plt.	Kunlun	35°40′N, 94°11′E	21/May/1994	60 (6)	30.9(21.3~61.8)
	Tanggula	33°04′N, 94°05′E	18/Jun./1993	90 (9)	20.1(11.7~34.1)
	Himalaya	28°27′N, 85°45′E	09/Aug./1991	100 (4)	58.9(25.9~81.3)
Ant.	ITAE route (5896km)	A line from (65°05′S, 59°35′W) Via 90°S to (67°21′S, 93°26′E)	27/Jul./1989 to 25/Feb./1990	25 (Totally 93)	Average 13.68 (3.1~47.3)

"Arct.", "T. Plt." and "Ant." represents Arctic region, Tibetan Plateau and Antarctica, respectively. In the "Arct." column: site 1 locates at Toronto, Canada, no snow samples collected; site 2 locates nearby USA and Canada border, actually at mid-latitude other than Arctic region, but the results are listed for contrasting study. Sites 2~6 denote respectively: 2, Ely; 3, Hudson Bay; 4, Point of Woods; 5. Resolute; and 6, Eureka.

* Numbers in brackets represent sample numbers collected from each snow pit.

observed distribution of Pb concentration is mainly due to sources outside the plateau region. Industrial facilities and other pollution sources are practically non-existent in the plateau, while either side has strong pollution sources from two largest developing countries in the world, i.e., China and India. Highest Pb concentration in precipitation at Kangwure may come from pollutants from south Asian countries. Meteorological study revealed that air masses coming from south of the Himalaya pass over mountains and supply precipitation to northern slope. Another possible pollution source for the south of the plateau is from west Asia or Arabian countries (Wake et al., 1993). Westerly winds blocked by the plateau move round to the southern and the northern margin of the plateau and transport

pollution from these sources. Other pathways of air mass transportation to the Tibetan Plateau may be associated with the Mongolia high, and the East Asia monsoon. We have not enough evidence as yet to conclusively trace the dominant sources of Pb over the plateau.

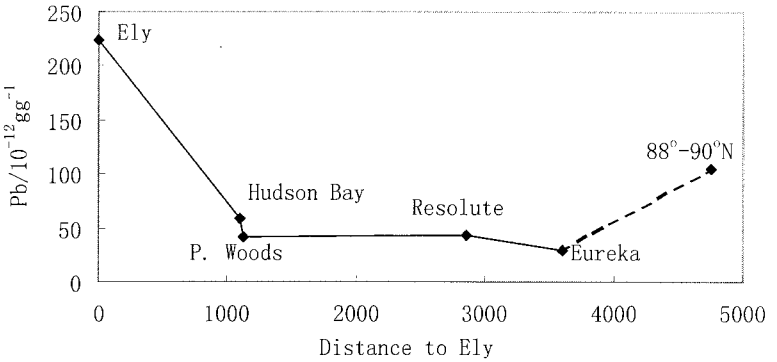

Figure 2. Distribution of Pb concentration in snow pits along transect from mid-latitude North America to North Pole.

Mean Pb concentrations are calculated for the 10 sites at central Arctic, 3 over Tibetan Plateau and 91 along the ITAE route. Among the three regions, the highest value is 105.7×10^{-12}g.g^{-1} for central Arctic, and 36.6×10^{-12}g.g^{-1} for Tibetan Plateau, and the lowest 13.7×10^{-12}g.g^{-1} for the Antarctic Ice Sheet. The central Arctic is surrounded by the continents of the Northern Hemisphere, and there are ample meteorological and atmospheric chemical evidence showing that industrial pollutants from these continents are transported to the central Arctic (Rahn, 1981; Barrie, 1986; Iversen, 1996). The Tibetan Plateau rises to the upper troposphere, and Pb input appears to decline with increasing elevation. Hence, although the plateau is located at mid-latitude and is surrounded by several big developing countries, including China and India, lower Pb concentrations are found in snow than those in the Arctic. This implies that the atmospheric Pb content over upper troposphere over mid-latitudes may be lower than that of the lower troposphere of remote Arctic regions. The Antarctic Ice Sheet, because of the long transport distance from other continents, and owing to the barrier effect of the Antarctic convergence zone, is the least polluted area on the globe.

In order to distinguish between natural and anthropogenic Pb in the three regions, three typical sites—North Pole at center Arctic, Tanggula Mt. at central Tibetan Plateau, and Dolleman Island near Antarctic Peninsula—were selected (Figure 3). The sites have been chosen to represent the average atmospheric chemical situation of the central Arctic, the Tibetan Plateau and Antarctica (Yao et al., 1991; Qin et al., 1999. Suttie and Wolff, 1992).

Generally the natural Pb comprises crustal and marine Pb. Using Al and Na^+ as the conservative tracers of crustal mineral and sea-salt, we calculated anthropogenic Pb by subtracting the crustal and marine Pb contributions (Suttie and Wolff, 1992). Marine Pb at Tanggula could not be calculated

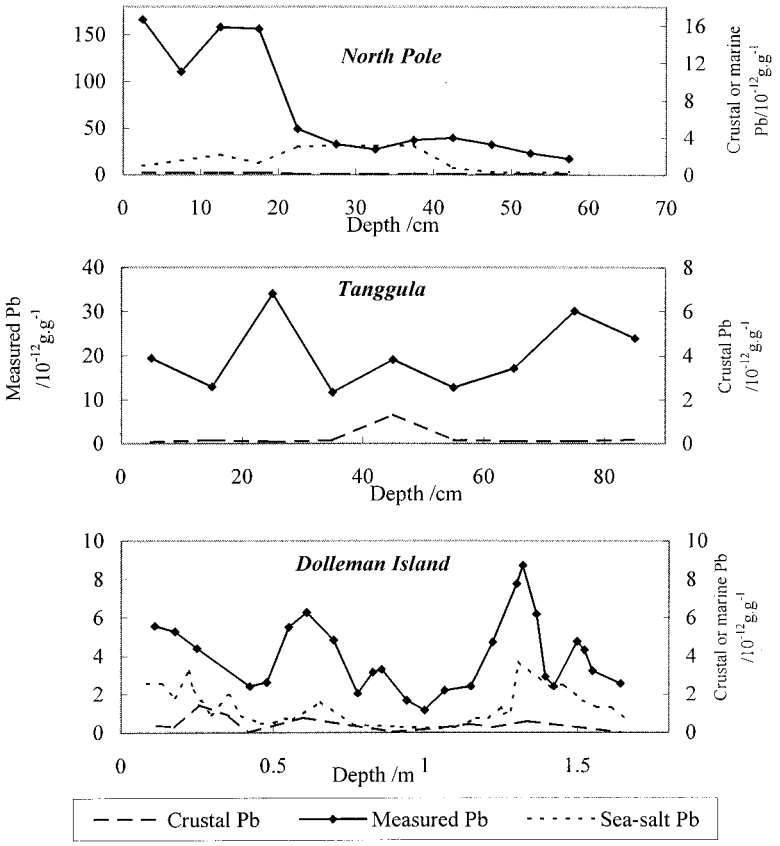

Figure 3. Contrast of Pb contents of different origin at central Arctic, Tibetan Plateau and Antarctica. The study on Dolleman Island is adopted from Suttie and Wolff (1992).

as there was no reliable conservative tracer, but we estimate that crustal contribution does not exceed marine Pb. The results are reported in Table 2. It is easy to see that natural lead at the three sites is roughly equal, i.e., less than 3×10^{-12}g.g^{-1}. However, the percentage of natural Pb varies: being negligible at the north pole and at Tanggula but reaching nearly 50% at Dolleman Island. In other words, anthropogenic Pb comprises over 97% of measured Pb at the north pole and Tanggula, and is over 50% at Dolleman Island. This confirms that Pb

pollution has spread into the most remote regions and probably the whole troposphere (at least 0~7000m a.s.l.).

Table 2. Contents and percentages of lead in different origin (natural or anthropogenic) at central Arctic,central Tibetan Plateau and Antarctica Peninsula

Sites	Background (10^{-12}g·g^{-1}) marine	crustal	%	Anthropogenic (10^{-12}g·g^{-1})	%
North Pole	1.70	0.15	2.5	71.95	97.5
Tanggula	≤0.26	0.26	≤2.6	≥19.61	≥97.4
Dolleman Island*	1.42	0.44	45.9	3.16	54.1

*Calculation based on Suttie and Wolff, 1992.

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