

LEAD IN THE HYDROSPHERE

Tsaihwa J. Chow

Scripps Institution of Oceanography, La Jolla, California 92093, USA

Abstract - This paper describes sources of lead in the hydrosphere, the global and regional distribution of lead in continental and ocean waters, its occurrence in waste discharges as well as the chronological concentration profiles of lead in polar ice sheets, continental glacier, and aquatic sediments. The recent United States recommendation for the maximum permissible amount of lead in drinking water is mentioned.

INTRODUCTION

Lead was among the first of metals to be used by man in antiquity because of its easy isolation and low melting point. It is an element with no known beneficial function in human metabolism and has been known since the Roman times to be toxic; but, in spite of its recognition as a poison, it is so widely used in modern technology that it continues to be an environmental concern.

Because of the adverse effect of lead aerosols, the air quality criteria for lead have been examined by government agencies (Ref. 1). However, little is known about lead in the aquatic environment. The purpose of this paper is to summarize our current knowledge of lead in the hydrosphere.

The environmental pathways of lead are illustrated in Fig. 1.

SOURCES OF AQUATIC LEAD

Natural source

The only natural source of lead in waters is the earth's crust containing an average of 15 p.p.m. of lead, with which water comes in contact. Lead content of continental water during pre-historic times can be estimated as follows: the total flow of river water into the oceans is about 3.7×10^{16} liter per year, and lead compounds have entered the oceans over the last few hundred thousand years at an average rate of 1.7×10^{10} gram per year; therefore, the global average of natural lead level in surface water was about 0.5 $\mu\text{g/L}$. However, this natural lead level may vary greatly with locations, depending on the geochemistry of the drainage basins.

Anthropogenic source

High concentrations of lead in continental surface waters usually occur near sources of contamination due to human activities. Industrial discharge, fallout, washout and runoff are apparently the major contributors of lead to lake and river waters. Patterson (2) estimated that the lead content of surface fresh water increased from a pre-historic level of 0.5 $\mu\text{g/L}$ to 5 $\mu\text{g/L}$ in modern times. Livingstone (3) stated that the present lead content of lake and river waters on a world-wide basis is 1 to 10 $\mu\text{g/L}$.

The use of lead in human activities has significantly altered the natural distribution of lead in the aquatic environment. In 1975 the world production of primary ore lead was 3.9 million metric tons. The U.S. consumption of lead was 1.2 million metric tons, of which 0.19 million tons were used in manufacturing automotive antiknock additives (Ref. 1). Since the advent of lead alkyl additives in 1920's, a cumulative amount of more than 6.5 million tons of lead have been consumed as additives in the U.S. alone as shown in Fig. 2. This is the largest single contributor to aquatic lead pollution. The use of lead additives reached a peak in the early 1970's. As a result of legislative action in the U.S. with respect to the maximum permissible content of lead in gasoline, the production and use of lead additives has decreased in recent years and should continue to do so.

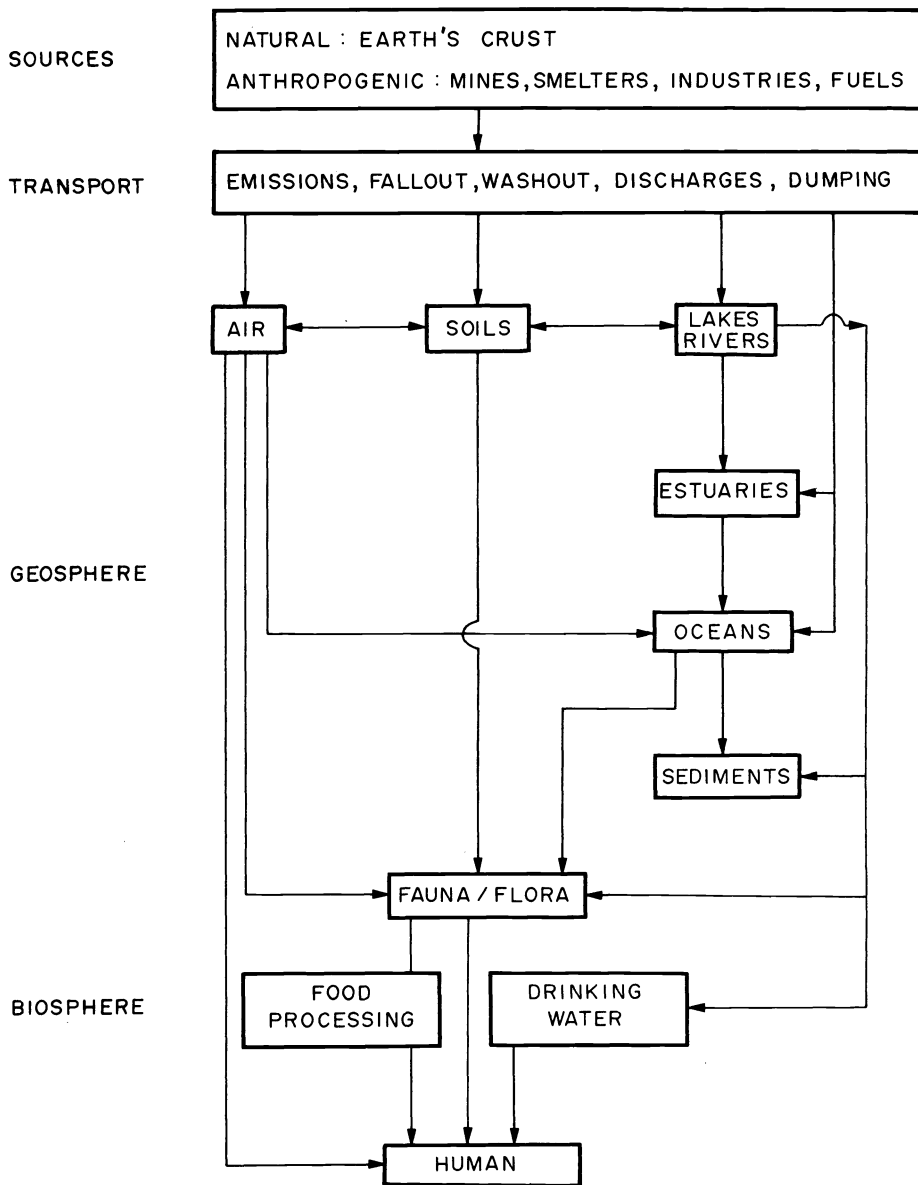


Fig. 1. Environmental pathways of lead.

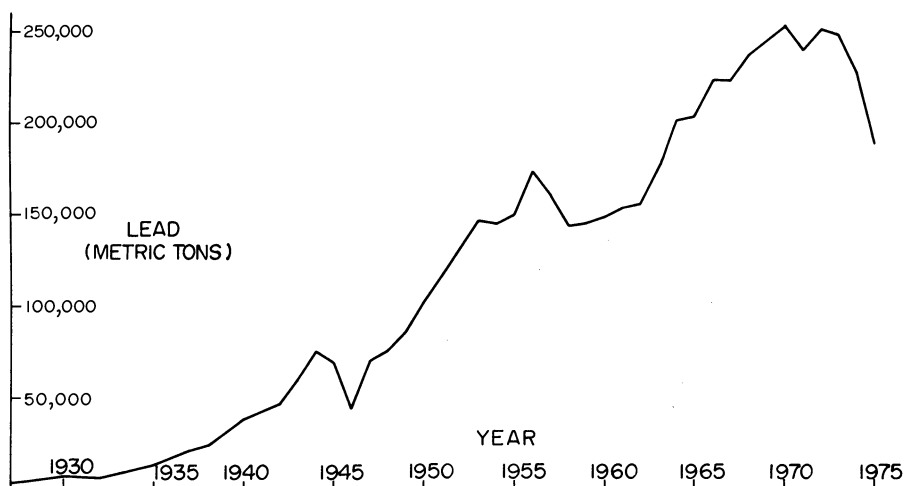


Fig. 2. Annual consumption of gasoline antiknock additives in the U.S.

The transport and distribution of lead from mobile and stationary sources are mainly via atmosphere. Lead aerosols emitted from leaded fuels are effectively dispersed into the ambient atmosphere of densely populated urban regions. Their sizes range from submicron to millimeter in diameters. These particulates are removed from the atmosphere by fallout and washout. The removal processes are mainly a function of particle size, shape and density, as well as ambient lead concentrations and meteorological conditions. The coarser fraction of lead aerosols settles along the roadside and is responsible for the high lead concentrations in soils. Lead aerosols of submicron sizes are carried in the air currents and spread around the globe.

DRINKING WATER

Maximum permissible lead

Drinking water is one of the major sources of lead intake by humans. Only few countries have established mandatory maximum permissible lead content for drinking water. The World Health Organization's European (4) and international (5) lead limit for drinking water is 100 $\mu\text{g/L}$; it specifies that in no instance should the lead content of drinking water exceed 300 $\mu\text{g/L}$ after 16 hours contact with the service pipes.

The Swedish permissible lead concentration in drinking water, which is purified according to the normal practice of chemical flocculation and slow filtering, is 20 $\mu\text{g/L}$ and that in waters much more extensively treated before use is 50 $\mu\text{g/L}$ (Ref. 6).

The USSR's maximum permissible lead concentration in drinking water is 100 $\mu\text{g/L}$ (Ref. 7); in addition, the USSR does not permit any amount of lead alkyls in its drinking water.

The Canadian Department of National Health and Welfare (8) has set a maximum permissible lead for drinking water of 50 $\mu\text{g/L}$.

In 1962 the U.S. Public Health Service (9) established a mandatory lead limit for drinking water as 50 $\mu\text{g/L}$. Most U.S. city building codes now forbid the use of lead pipes for potable purposes; however, lead pipes still exist in some older buildings.

In the United Kingdom, bye-laws issued by the Ministry of Housing and local government provide for the prohibition of lead pipes where necessary, but they do not compel the replacement of lead pipes already in use.

Plumbo-solvency

In some regions of soft water of slightly acidic pH where lead pipe is used in service and domestic lines, the drinking water can possess sufficient plumbo-solvency to result in appreciable concentration of dissolved lead. As high as 600 $\mu\text{g/L}$ of lead was found in lead-piped tap water in Cheshire, England, with the lead concentration decreasing to 3 $\mu\text{g/L}$ after the lead pipes were replaced (Ref. 10). The average lead content of

drinking water in Glasgow was 160 $\mu\text{g/L}$, with a range of 20 to 350 $\mu\text{g/L}$ (Ref. 11).

The U.S. Community Water Supply Study conducted in 1969 gave an average lead concentration of 13 $\mu\text{g/L}$, ranging from not detected to 640 $\mu\text{g/L}$ (Ref. 12). Of the 2,595 water samples surveyed, some 1.4 percent exceeded the 50 $\mu\text{g/L}$ limit of lead in drinking water. Some 76 percent of the tap water in Seattle, Washington exceeded the U.S. Public Health Service lead limit (Ref. 13). Similar results of Boston, Massachusetts indicated that 65 percent of tap water samples exceeded the 50 $\mu\text{g/L}$ limit for lead. In all cases, the lead content of drinking water was higher at the tap than at the treatment plant (Ref. 14).

Health effect

Although acute lead poisoning due to drinking water is rare, chronic lead toxicity occurs even with low daily intake because of its accumulation in bone and tissue. There is a correlation between water lead content and the quantity of lead piping in the Glasgow water supply system and, in addition, that as this water lead concentration rose, so did the blood lead concentration of people resident in these areas (Ref. 15). Results of studies in the Boston area indicated that increase lead levels of blood occur in children when the water supply contains 50 to 100 $\mu\text{g/L}$ of lead.

In a recent study entitled 'Drinking Water and Health' conducted by the U.S. National Academy of Sciences (16), it is stated that "the inorganic contaminant [in drinking water] with the greatest potential for toxicity is lead. The present standard [for lead] may not provide an adequate margin of safety, especially for infants and young children..... The sum of the estimated absorption [of lead] from the various routes, 50-60 $\mu\text{g/day}$, is already at the maximum 'no-observed-adverse-health-effect' value of 50-60 $\mu\text{g/day}$ The World Health Organization recommendation of 5 μg of lead per day per kg of body weight as a safe total daily intake cannot be met for a 12 kg child when the water supply contains as much as 50 $\mu\text{g/L}$ of lead. It is concluded that the 'no-observed-adverse-health-effect' level cannot be set with assurance at any value greater than 25 $\mu\text{g/L}$ [of lead for drinking water]."

Water purification

Drinking waters are not specifically treated for removing lead, since the lead content of raw water supplies is seldom over the 50 $\mu\text{g/L}$ limit and the ordinary water treatment process has little effect on lead. Naylor and Dague (17) carried out laboratory experiments and evaluated the effectiveness of lime coagulation, lime, and lime-soda ash softening in the removal of lead from drinking water. The system effecting the most complete removal lead is the solid-contact softening process. Virtually all lead is removed from solutions containing an initial lead concentration of 2,000 $\mu\text{g/L}$ at contact solids concentrations of 1 percent and greater.

Coagulation and flocculation in water reduce the lead concentration to less than 50 $\mu\text{g/L}$ in the pH range of 7.7 to 8.8. Because the coagulation and flocculation process is usually operated at a pH of about 7, the maximum lead removal will not be obtained.

RAIN WATER

Large quantities of lead aerosols are continuously emitted into the atmosphere. Therefore, rain water may contain an appreciable amount of lead as a result of airborne lead being washed out in the precipitation process.

In the U.S., lead concentration in rain water samples collected at a nationwide network of 32 stations averaged 36 $\mu\text{g/L}$ (Ref. 18). In the cities studied, the lead content of rain water was correlated with local gasoline consumption. The average lead concentration of rain water collected at San Diego, California during 1967-1970 was 38 $\mu\text{g/L}$ with a range from 3 to over 300 $\mu\text{g/L}$, showing an inverse relationship between amount of precipitation per rainfall and lead content (Ref. 19). The lead concentration of rain water collected during 1973 at a rural station near Chadron, Nebraska averaged 4.3 $\mu\text{g/L}$ (Ref. 20).

In England, Peirson et al. (21) obtained an average lead content of 39 $\mu\text{g/L}$ for rain water samples collected at Windermere in 1971.

Drozdova and Makhon'ko (22) reported that the annual average lead concentration of rain water obtained during 1968-1969 at 16 stations in the European territory of the USSR was 5.5 $\mu\text{g/L}$, ranging from 1 to 48 $\mu\text{g/L}$.

SNOW AND ICE

Polar snow

One way to find out the anthropogenic contribution of lead to the environment with time is to measure the precipitated lead in annual successive layers of preserved snow. The quiescent ice sheets in the arid, perpetually frozen polar regions provide chronological layers of precipitation that are undisturbed by percolation and mechanical mixing.

Murozumi et al. (23) reported the analyses of lead aerosols, terrestrial dusts and sea-salts in annual ice layers from the interior of northern Greenland and of the Antarctic continent. The Greenland ice contained 10 p.p.b. of silicate dusts and 60 p.p.b. of sea salts, which contributed approximately 0.0002 μg of lead per kg of ice and this contribution did not change appreciably with time. As shown in Fig. 3, the observed 0.001 μg of lead per kg of the 800 B.C. Greenland ice is higher than the lead level expected from natural terrestrial dusts present in the ice. It can be seen that the lead concentration in the 1753 A.D. ice layer, which corresponds to the beginning of the European Industrial Revolution, was 0.011 $\mu\text{g}/\text{kg}$. This concentration is already more than twenty-five times higher than the natural level. Lead content apparently tripled in Greenland snow during the half-century from 1753 to 1815, and doubled again during the following century from 1815 to 1933. During the next three decades from 1933 to 1965, lead concentrations arose abruptly by a factor of about three to 0.20 $\mu\text{g}/\text{kg}$, which is well over five hundred times the natural level.

Lead concentrations were found to be much less in south polar snows than in the Greenland snows, confirming their industrial origin since most of the lead aerosols originated from the northern hemisphere. The lead content of the Antarctic snow samples later than 1916 was at least 0.01 $\mu\text{g}/\text{kg}$.

The increase of lead concentration with time in north polar snow is attributed to the coal-burning and lead smelteries before 1940, and to combusted lead alkyl additives in automotive fuels after 1940.

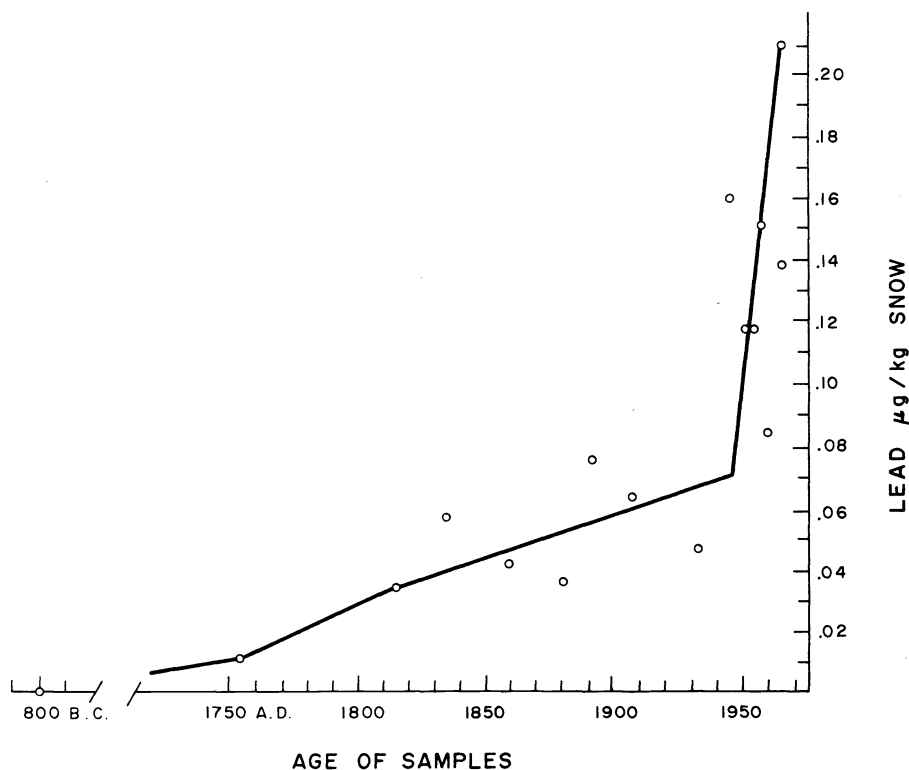


Fig. 3. Increase of industrial lead pollutants in Camp Century, Greenland snow with time since 800 B.C.
From Murozumi et al. (23)

Continental glacier

On the European continent, Jaworoski (24) found that lead concentrations in ice from two small glaciers in Poland have increased during the past century. The mean lead concentration of 75 $\mu\text{g}/\text{kg}$ for the 1960-1965 samples was about fifteen times greater than that for the 1861-1866 snow samples, in which the mean lead concentration was 5 $\mu\text{g}/\text{kg}$; but, it should be noted that some of the latest ice layers contained between 80 to 170 $\mu\text{g}/\text{kg}$ of lead. It is unlikely that such lead concentrations are typical of those in temperate latitude snows of the northern hemisphere. However, temperate glaciers are much dustier than polar ice sheets. The observed lead in polar ice and Polish glaciers probably represent the extremes of global and local significances, respectively.

Snow

An average lead content of 1.6 $\mu\text{g}/\text{kg}$ of fresh snow was found in Lassen Volcanic National Park, California (Ref. 25) and 0.59 $\mu\text{g}/\text{kg}$ for a snow pack in Thompson Canyon which is located in the crest of the High Sierra in California (Ref. 26).

In recent years, the snow falling in southern Norway was laminated with conspicuous grey bands which had a higher lead content, 30 to 40 $\mu\text{g}/\text{kg}$, than the white layers of snow. The amount of lead pollutant cannot be accounted for by the local industrial activities (Ref. 27). The lead content of new-falling snow collected at about one-half km from a stack of a lead smelter in a mountainous area of Zerjav, Yugoslavia contained about 19,000 μg of lead per kg of snow (Ref. 28). With increasing distance from the stack, the snow contained consistently decreasing quantities of lead. At a distance of 60 km, the lead content of snow was 20 $\mu\text{g}/\text{kg}$.

The lead concentrations in snow collected on the ground within 30 meter of roads in residential Columbus, Ohio contained between 50 to 1,090 $\mu\text{g}/\text{kg}$, with an average of 410 $\mu\text{g}/\text{kg}$ (Ref. 29). Roadside snow samples in Ottawa, Canada contained between 55 to 410 $\mu\text{g}/\text{kg}$ of lead, while the rural snow contained only 2 to 15 $\mu\text{g}/\text{kg}$ (Ref. 30 and 31). These high lead concentrations of roadside snow were caused by the combustion of leaded gasoline and decreased with increasing distance from the source of pollution.

SURFACE AND GROUND WATERS

Voluminous quantities of publications on the concentration of lead in river, lake, and ground waters for some regions and the lack of information for others makes it impractical to compile comparative literature. The lead concentration of selected natural surface and ground waters summarized in Table 1 is intended to be a reference source; for the detailed interpretation of those values, the original publications should be consulted. It should be cautioned that the validity of the earlier results is not easily assured because the analytical techniques then in use might not be sensitive enough to determine lead in the microgram or submicrogram per liter range.

In discussing the occurrence of lead in U.S. surface waters, Fleischer (32) estimated that more than 90 percent of the lead discharged into the rivers and lakes is attributed to man-made sources. The lead concentration of water bodies varies daily, seasonally and yearly as well as with water depths. The rate and volume of river flow also influence the lead concentration of its water.

High concentrations of lead in surface waters usually occur near sources of contamination. Industrial discharges, rainfall and runoff are apparently the major contributors of lead in river and lake waters. The content of lead in rivers, which flow through populated regions, increases as runoff from precipitation rises. Lead concentrations are lower in water samples collected at remote sites than in samples from rivers below metropolitan and industrial locations. For example, lead contents as high as 120 $\mu\text{g}/\text{L}$ were occasionally observed in Frazer River waters below Vancouver as compared with a lead concentration range of 1 to 8 $\mu\text{g}/\text{L}$ at the upstreams of the river (Ref. 41).

Indications are that all continental surface waters show a varying degree of lead contamination. From the available data, it seems that the global mean of lead content for rivers and lakes lies between 1 and 10 $\mu\text{g}/\text{L}$.

Ground waters usually contain a higher lead concentration than the natural surface water. The chemical composition and temperature of the ground water as well as the lead content of surrounding rocks affect the lead content of ground water. The penetration of sulfide deposits by aerated ground water can cause local anomalies of lead in runoff; thus, the streams and springs in those regions may contain higher amounts of lead. For instance, some well waters situated near lead ore deposits in Harz Mountain, Germany contained as high as 3,400 $\mu\text{g}/\text{L}$ of lead (Ref. 43).

TABLE 1. Lead concentration in selected surface and ground waters

Location	Sample	Lead content, µg/L		Reference
		Range	Average	
USA	Surface water	<1-55	3.9	Ref. 33
Mississippi River, USA	Surface water		6.3	Ref. 34
Hudson River, USA	Surface water		7.5	Ref. 34
Susquehanna River, USA	Surface water		3.5	Ref. 34
Columbia River, USA	Surface water		3.2	Ref. 34
Colorado River, USA	Surface water		11	Ref. 34
Sacramento River, USA	Surface water		3.3	Ref. 34
Ohio River, USA	Surface water	n.d.-30		Ref. 35
Lake Tahoe, USA	Surface water		<1	Ref. 36
Maine, USA	Rivers and lakes		2.6	Ref. 37
Lake Michigan, USA	Surface water	0.4-2.0		Ref. 38
Lake Erie	Surface water		4	Ref. 39
Lake Ontario	Surface water		0.83	Ref. 40
Mackenzie River, Canada	Surface water		5.6	Ref. 34
Churchill River, Canada	Surface water		3.3	Ref. 34
Nelson River, Canada	Surface water		6.2	Ref. 34
Frazer River, Canada	Surface water	1-8		Ref. 41
St. Lawrence River, Canada	Surface water		3.9	Ref. 34
Rhine River, Germany	Surface water		6	Ref. 42
Elbe River, Germany	Surface water	2-6		Ref. 42
Söse River, Germany	Surface water	up to 15		Ref. 43
Sieber River, Germany	Surface water	up to 13		Ref. 43
Oder River, Germany	Surface water	up to 18		Ref. 43
Oker River, Germany	Surface water	up to 25		Ref. 43
Ecker River, Germany	Surface water	up to 180		Ref. 43
Saale River, Germany	Surface water	1.4-9.2	3.9	Ref. 44
Pripyat River, USSR	Surface water	2.3-13	3.3	Ref. 45
Dnieper River, USSR	Surface water	0.1-5.5		Ref. 46
N.W. Siberia, USSR	Surface & ground		0.48	Ref. 48
Siberia, USSR	Surface & ground	0.1-2,071	1.5	Ref. 49
Pripyat Basin, USSR	Ground water	1.2-4.3		Ref. 45
Armenia, USSR	Ground water		50	Ref. 47
Donets Basin, USSR	Ground water	4-96		Ref. 50
Derbyshire, England	Ground water	1-16		Ref. 51
Amsterdam, Holland	Ground water	5-124	27	Ref. 52
Tamagawa, Akita, Japan	Hot spring	20-1,800		Ref. 53
Aomori, Japan	Thermal well	10-50		Ref. 54
Hyogo, Kanagawa & Shizuoka	Thermal water	3.6-62		Ref. 55
Shibukuro, Japan	Thermal water		1,530	Ref. 56
Kotsuki River, Kyushu, Japan	Surface water	0.9-3.5		Ref. 75
Rivers entering Nagoya Bay	Surface water	0.2-20		Ref. 76
Sakawa, Kanryu, Kuji & Katashina River water		0.8-1.4		Ref. 77

WASTE DISCHARGES

Pending water pollution control legislation in the United States requires heavy metal removal, including lead, not only before industrial wastes are discharged into navigable waters, but also prior to ocean dumping or land disposal.

Storm runoff

Storm water runoff from urban and industrial areas was estimated to carry approximately 8×10^9 gram of lead per year, which is equivalent to an addition of 4 $\mu\text{g/L}$ of lead to the total river discharge of the United States (Ref. 2). The lead concentration in Durham, North Carolina storm water averaged 1,500 $\mu\text{g/L}$ (Ref. 57). Runoff from streets with high traffic volumes was found to contain as much as 5,500 $\mu\text{g/L}$ of lead in Oklahoma City, Oklahoma, with only 90 $\mu\text{g/L}$ from an open field (Ref. 58).

About 90 to 140 metric tons of lead from the Los Angeles Land Basin flow annually into the California coastal waters as storm runoff (Ref. 59).

Municipal sewage

Municipal sewage varies greatly in lead concentrations depending on the nature of the collection system and the industrial waste contribution to the system. Cities with few industrial sources of lead may expect less than 10 $\mu\text{g/L}$ of lead in sewage, while industrialized districts may find 100 to 500 $\mu\text{g/L}$ of lead. The average lead concentration in effluent from Los Angeles, California sewer discharge was 250 $\mu\text{g/L}$ (Ref. 59). The lead concentration in the influent, primary and secondary effluent of a treatment plant near Pittsburgh, Pennsylvania was 110, 55, and 22 $\mu\text{g/L}$, respectively (Ref. 60).

Industrial sources

Discharges from lead mining, smelting and refining operations may cause severe lead pollution in local surface waters. Industries which use lead may also discharge wastes containing excessive concentration of lead. For example, plating waste with lead concentrations of 2,000 to 140,000 $\mu\text{g/L}$ were obtained (Ref. 61). The lead content of mine waste water from the Keno Hill and Galena Hill district of the Yukon Territory of Canada was as high as 1,000 $\mu\text{g/L}$ (Ref. 62). Concentrations of 1,000 $\mu\text{g/L}$ of lead was found in some settling ponds in recently developed lead mining districts of Missouri, USA (Ref. 63).

Mine and creek waters from mining areas in Kazakhstan, Caucasus, and Altai of USSR contained 7,000 to 9,000 $\mu\text{g/L}$ of lead (Ref. 74).

Lead in industrial waste waters can be removed by chemical precipitation, activated carbon adsorption, electrodeposition, solvent extraction, ultrafiltration, and ion exchange. Three papers on the removal of heavy metals from waste water and sludges will be presented later at this symposium.

OCEAN WATERS

It should be stated that much of the present knowledge concerning lead in ocean waters is tentative and that additional research is needed in order to understand the importance of lead in marine waters.

The lead concentration of sea water is extremely low. Because of difficulties associated with its sampling and chemical analysis, the distribution of lead in the open and deep ocean waters is little known. Prior to 1958, all analyses of lead in coastal surface water samples have shown a concentration range of 2 to 8 $\mu\text{g/L}$; these obviously too high concentrations were caused by insufficient precautions during water sampling and insensitive analytical methods. With the isotope dilution method, the lead content of Pacific waters off the Washington coast to a depth of 100 meters was determined in 1958 as less than 0.1 $\mu\text{g/L}$ (Ref. 65).

Until 1963 lead concentrations in deep ocean waters had not been determined. With the "unprotected" water sampler available at that time, the lead concentration profiles in the Atlantic, Pacific and Mediterranean ocean waters were obtained (Ref. 25, 66 and 67). Lead concentrations averaging about 0.05 $\mu\text{g/L}$, with some scatter, were obtained at four stations across the central Atlantic, and about 0.03 $\mu\text{g/L}$ was observed in deep waters of the Pacific as well as in the Mediterranean. In surface sea water off southern California, lead concentrations varied from 0.08 to 0.4 $\mu\text{g/L}$. For waters around Bermuda, which are relatively free from the continental influences, lead concentrations decreased

from 0.07 $\mu\text{g/L}$ at the surface of the ocean to about 0.04 $\mu\text{g/L}$ in the 3,000 meter deep water. These data interpreted at that time was that there were no essential differences in lead concentrations in deep waters below the 1,000 meter level of these oceans; but in the surface water, lead concentrations in the Pacific and Mediterranean waters were higher than those in the central Atlantic. These depth profiles of lead in the ocean waters may be interpreted as showing that lead is being introduced into the oceans at a rate far greater than the rate of natural weathering.

In recent years considerable emphasis has been placed on the study of marine environmental qualities. Many lead concentrations recently reported for various coastal waters, which were selected for their polluted conditions, were much higher than those of the open ocean waters.

Results of a "Lead Analysis Workshop" organized by Patterson (68) showed that of four southern California surface water samples investigated, none showed lead concentrations exceeding 0.08 $\mu\text{g/L}$, indicating that previously reported lead concentrations of several micrograms per liter in coastal waters may be taken as a grain of sea salt.

Patterson (68) also pointed out that deep ocean waters previously analyzed (Ref. 25, 66 and 67) were collected by lowering the cleaned water sampler through the surface water in an "unprotected" condition, and high lead concentrations may have been obtained as a consequence. If "protected" water sampler were used and sea water were analyzed by the isotope dilution method in a clean chemical laboratory, the lead concentration of deep ocean waters may be found much less than previously reported. Using a specially designed "protected" deep water sampler, recent results of Schaule and Patterson (69) indeed show that total lead concentrations in deep ocean water, collected 150 km off the California coast, are about 0.004 $\mu\text{g/L}$ at 1,000 meter depth, decreasing to about 0.002 $\mu\text{g/L}$ at 3,000 meter depth.

LEAD IN SEDIMENTS

Lead concentration profiles in sediment cores from inland lakes and marine waters can be used to evaluate the rate and extent of lead accumulation resulting from human activities. Using the records of lead concentration in sedimentary varves, the natural and anthropogenic lead influx to the southern California coastal waters over the past century were determined (Ref. 70). The lead concentration in the sediments increased with time from a pre-industrial level of about 10 p.p.m. to a range of 20 to 38 p.p.m. for recent years. The present anthropogenic lead influx into the Los Angeles Bight is 1.8 $\mu\text{g/cm}^2/\text{yr}$ compared to natural flux of 0.50 $\mu\text{g/cm}^2/\text{yr}$. The lead concentration profile in sediment cores of the western Baltic Sea also showed a four-fold increase to a present lead level of 80 p.p.m. within the last 70 years; the present rate of anthropogenic lead accumulation is 2.2 $\mu\text{g/cm}^2/\text{yr}$ for the Baltic (Ref. 71).

For inland lakes, the lead concentration profile for sediment cores from Lake Washington near Seattle, Washington has been reported (Ref. 72 and 73). The concentration of lead in these cores has increased from 25 p.p.m. to about 400 p.p.m. during the past century. This lead increase parallels the increase in population of the surrounding land.

The lead profile in the Lake Michigan sediment reflects the history of lead input to the lake, the source being primarily from the burning of coal and leaded gasoline since about 1830's and 1930's, respectively (Ref. 74). The lead content in sediments prior to 1830 was about 38 p.p.m. while that of recent sediment layers ranged from 120 to 164 p.p.m. The anthropogenic lead flux into the southern basin of Lake Michigan in 1972 was 1.3 $\mu\text{g/cm}^2/\text{yr}$ in comparison to the natural flux of 0.16 $\mu\text{g/cm}^2/\text{yr}$. An estimate of 230 tons of lead aerosols originating in the nearby industrial Chicago-Gary region deposited in Lake Michigan in 1972.

REGULATING FACTORS

At present, solubility considerations alone do not satisfactorily explain observed lead concentrations in natural waters. The adsorption of lead from aqueous solutions on clay minerals and other inorganics may play an important role in affecting the concentration of lead in natural waters. However, organic substances, both dissolved and suspended, are perhaps more important than the inorganics in complexing with lead.

Acknowledgement - An expanded version of this article will be published in Biogeochemistry of Lead (J. O. Nriagu, Ed.), Elsevier, Amsterdam, 1978. I thank Dr. Nriagu for his permission.

REFERENCES

1. U.S. Environmental Protection Agency, Air Quality Criteria for Lead, U.S. Government Printing Office, Washington, D.C. (1977).
2. C.C. Patterson, Arch. Environ. Health **11**, 344-363 (1965).
3. D.A. Livingstone, U.S. Geol. Survey Prof. Paper 440-G, 64 pp. (1963).
4. World Health Organization, European Standards for Drinking Water, 58 pp. Geneva (1970).
5. World Health Organization, International Standards for Drinking Water, 3rd ed., 67 pp., Geneva (1971).
6. National Swedish Environment Protection Board, Report on Water Quality Criteria for Swedish Surface Water, Solna, Sweden (1969).
7. D. Stöfen, Toxicology **1**, 187-195 (1973).
8. Canadian Department of National Health and Welfare, Canadian Drinking Water Standards and Objectives, Ottawa (1969).
9. U.S. Public Health Service, Drinking Water Standards, Public Health Service Publ. 956, 61 pp. (1962).
10. W.H. Parry, Lancet 1207-1208 (1967).
11. M.R. Moore, Science of the Total Environ. **7**, 109-115 (1977).
12. L.J. McCabe, J.M. Symons, R.D. Lee and G.G. Robeck, J. Am. Water Works Assoc. **62**, 670-687 (1970).
13. G.F. Craun and L.J. McCabe, J. Am. Water Works Assoc. **61**, 593-599 (1975).
14. P.C. Karalekas, Jr., G.F. Craun, A.F. Hammonds, C.R. Ryan and D.J. Worth, Proc. Am. Water Works Assoc. Annual Conf. **95**, 5-15 (1975).
15. A.D. Beattie, M.R. Moore, W.T. Devenay, A.R. Mills and A. Goldberg, Brit. Med. J. **1**, 491-493 (1972).
16. U.S. National Academy of Sciences, Drinking Water and Health, Washington, D.C. (1977).
17. L.M. Naylor and R.R. Dague, J. Am. Water Works Assoc. **67**, 560-565 (1975).
18. A.L. Lazrus, E. Lorange and J.P. Lodge, Jr., Environ. Sci. Technol. **4**, 55-58 (1970).
19. T.J. Chow and J.L. Earl, Science **169**, 577-580 (1970).
20. A.W. Struempfer, Atmos. Environ. **10**, 33-37 (1976).
21. D.H. Peirson, P.A. Cawse, L. Salmon and R.S. Cambray, Nature **241**, 252-256 (1973).
22. V.M. Drozdova and E.P. Makhon'ko, J. Geophys. Res. **75**, 3610-3612 (1970).
23. M. Murozumi, T.J. Chow and C.C. Patterson, Geochim. Cosmochim. Acta **33**, 1247-1294 (1969).
24. Z. Jaworoski, Nature **217**, 152-153 (1968).
25. M. Tatsumoto and C.C. Patterson, Nature **199**, 350-352 (1963).
26. Y. Hirao and C.C. Patterson, Science **184**, 989-992 (1974).
27. K. Elgmork, A. Hagen and A. Langeland, Environ. Pollut. **4**, 41-52 (1973).
28. Z. Kerin, Arch. Environ. Health **26**, 256-259 (1973).
29. W.L. Hamilton and J.E. Miller, Ohio J. Sci. **71**, 313-316 (1971).
30. I.R. Jonasson, Nature **241**, 447-448 (1973).
31. B.G. Oliver, J.B. Milne and N. LaBarre, J. Water Pollut. Control Fed. **46**, 766-771 (1974).
32. M. Fleischer, In: Cycling and Control of Metals, Proc. Environ. Resour. Conf., p. 3-10, National Environ. Res. Center, Cincinnati, Ohio (1973).
33. W.H. Durum, J.D. Hem and S.G. Heidel, U.S. Geol. Survey Circular 643, 49 pp. (1971).
34. W.H. Durum and J. Haffty, U.S. Geol. Survey Circular 445, 11 pp. (1961).
35. Ohio River Valley Sanitation Commission, Water Quality and Flow Variations, 1956-1957, 168 pp. Cincinnati, Ohio (1959).
36. C.R. Goldman and R.C. Carter, J. Water Pollut. Control Fed. **37**, 1044-1059 (1965).
37. K.K. Turekian and M.D. Kleinkopf, Bull. Geol. Soc. Am. **67**, 1129-1131 (1956).
38. H.V. Leland, S.S. Shukla and N.F. Shimp, In: Trace Metals and Metal-Organic Interactions in Natural Waters, Ann Arbor Science, Ann Arbor, Michigan (1973).
39. V.K. Chawla and Y.K. Chau, In: Proc. 12th Conf. Great Lakes Res., Internat. Assoc. Great Lakes Res., p. 760-765, Ann Arbor, Michigan (1969).
40. Y.K. Chau, V.K. Chawla, H.F. Nicholson and R.R. Vollenweider, In: Proc. 13th Conf. Great Lakes Res., p. 659-672, Internat. Assoc. Great Lakes Res., Ann Arbor, Michigan (1970).
41. K.J. Hall and K. Fletcher, In: Proc. Internat. Conf. Transport of Persistent chemicals in Aquatic Ecosystems, Vol. 1, p. I 83-87, Nat. Res. Coun. Canada, Ottawa (1974).
42. U. Föhrstner and G. Müller, Schwermetalle in Flüssen und Seen, Springer-Verlag, New York (1974).
43. H. Novak and F. Preul, Geol. Jahrb. **105**, 1-68 (1971).
44. F. Heide, H. Lertz and G. Böhm, Naturwiss. **44**, 441-442 (1957).
45. K.I. Lukashov, A.L. Zhukhositskaya and A.A. Zamyatkina, Dokl. Akad. Nauk Belorussk. SSR **9**, 183-186 (1965).
46. A.L. Zhukhositskaya, A.A. Zamyatkina and K.I. Lukashov, Dokl. Akad. Nauk Belorussk. SSR **10**, 891-893 (1966).
47. S.R. Krainov and M.K. Korolkova, Geokhim. 453-463 (1962).
48. A.E. Kontorovich, M.A. Sadikov and S.L. Shvartsev, Dokl. Akad. Nauk SSSR **149**, 179-180 (1963).
49. P.A. Udodov and Y.S. Parilov, Geokhim. 703-707 (1961).

50. O.O. Falovskii, L.O. Davidyuk and N.S. Chervinko, Geol. Zh. Akad. Nauk Ukr. RSR **26**, 45-52 (1966).
51. W.M. Edmunds, Inst. Geol. Sci. London, Rept. 71/77, 52 pp. (1971).
52. F. J. Brinkmann, Geol. Mijbouw **53**, 157-161 (1974).
53. E. Minami, G. Sato and K. Watanuki, J. Chem. Soc. Japan, Pure Chem. Sect. **78**, 1096-1100 (1957).
54. K. Noguchi and R. Nakagawa, Nippon Kagaku Zasshi **91**, 127-131 (1970).
55. K. Kuroda, Bull. Chem. Soc. Japan **15**, 153-155 (1940).
56. B. Takano and K. Watanuki, Geochem. J. **6**, 1-9 (1972).
57. E.H. Bryan, J. Water Pollut. Control Fed. **46**, 2419-2431 (1974).
58. C.D. Newton, W.W. Shepard and M.S. Coleman, J. Water Pollut. Control Fed. **46**, 999-1000 (1974).
59. D.R. Young, C.S. Young and G.E. Hlavka, In: Cycling and Control of Metals, Proc. Environ. Resour. Conf., p. 21-39, Nat. Environ. Res. Center, Cincinnati, Ohio.
60. J.A. Davis, III and J. Jacknow, J. Water Pollut. Control Fed. **47**, 2292-2297 (1975).
61. B.B. Ewing and J.E. Pearson, In: Adv. Environ. Sci. Technol., Vol. 3, p. 1-126, Wiley, New York (1974).
62. R.W. Boyle, Canadian Dept. Mines and Tech. Surveys, Geol. Survey Bull. **111**, 302 pp. (1965).
63. P.D. Proctor, G. Kisvarsanyi, E. Garrison and A. Williams, In: Trace Substances in Environmental Health VII, University of Missouri, Columbia (1974).
64. M.M. Konstantinov, Dokl. Akad. Nauk SSSR **2**, 65-80 (1954).
65. T.J. Chow, J. Mar. Res. **17**, 120-127 (1958).
66. M. Tatsumoto and C.C. Patterson, In: Earth Science and Meteoritics, p. 74-89, North Holland Publ., Amsterdam (1963).
67. T.J. Chow and C.C. Patterson, Earth Planet. Sci. Lett. **1**, 397-400 (1966).
68. C.C. Patterson, Mar. Chem. **2**, 69-84 (1974).
69. B. Schaule and C.C. Patterson, Personal Communication (1977).
70. T.J. Chow, K.W. Bruland, K. Bertine, A. Soutar, M. Koide and E.D. Goldberg, Science **181**, 551-552 (1973).
71. H. Erlenseuser, E. Suess and H. Willkomm, Geochim. Cosmochim. Acta **38**, 823-842 (1974).
72. R.S. Barnes and W.R. Schell, In: Cycling and Control of Metals, Proc. Environ. Resour. Conf., p. 45-53, Nat. Environ. Res. Center, Cincinnati, Ohio (1973).
73. E. Crecelius and D. Piper, Environ. Sci. Technol. **7**, 1053-1055 (1973).
74. D.N. Edgington and J.A. Robbins, Environ. Sci. Technol. **10**, 266-274 (1976).
75. M. Kamada, T. Onishi and H. Sakamoto, J. Chem. Soc. Japan, Chem. & Indus. Chem. **1**, 35-47 (1977).
76. Y. Kitano, K. Ito and S. Kojima, J. Oceanogr. Soc. Japan, Special Volume, p. 114-122, (1975).
77. Y. Ide, Y. Morita and H. Fukushima, Indus. Pollut. **8**, 661-668 (1972).