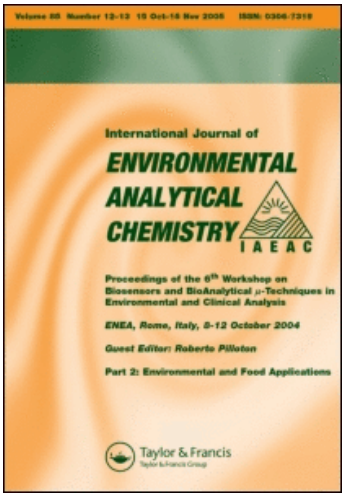

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Specific Activity of ^{210}Pb in the Environment

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KEY WORDS: Lead^{210}, biological samples, specific activity, aerosols.

A review of data on the specific activity of ^{210}Pb (pCi ^{210}Pb/mg stable Pb) in the environment is presented. Experimental results on aerosol, plant, animal and human samples from Finland are reported. Large variations (18–2450 pCi/mg Pb) were observed for aerosol samples taken from various locations and under different meteorological conditions. Results for the biological samples revealed a much narrower range. A considerable decrease of specific activity was observed along the food chain plant—animal—man, thus indicating contamination of the food chain by technological (low-activity) lead. Arctic values of specific activity were observed to be generally higher than "normal" values. Human samples from southern Finland contained about 20 pCi ^{210}Pb/mg Pb. Possibilities of utilizing specific activity measurements in lead pollution studies and in elucidating the origin of stable lead in the human diet are discussed.

INTRODUCTION

^{210}Pb ("radiolead") is a member of the natural ^{238}U-^{226}Ra decay series. Most ^{210}Pb found in the atmosphere and biosphere originates from the radioactive decay of atmospheric ^{222}Rn. Jaworowski[1] has estimated the annual natural atmospheric production of ^{210}Pb as approx. 0.62 MCi; Peirson *et al.*[2] have estimated the same figure to be about 1.0 MCi per year. Most of the stable lead in the human environment is apparently of technological origin.[3] "Technological lead," which is extracted from mines, normally has a very low ^{210}Pb content.[1] A major portion of atmospheric lead

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pollution originates from the burning of leaded gasoline by automobiles. In 1968, approx. 98% of the total amount of lead emitted to the atmosphere in the United States (estimated to be 184,000 tons) derived from gasoline combustion.[4]

Lead injected into the atmosphere as an aerosol becomes more or less "tagged" with radiolead. Thus, the "fallout" type of lead pollution can be generally expected to exhibit a relatively high specific activity of ^{210}Pb, whereas other types of lead pollution should have low or negligible specific activity. According to the world-wide survey undertaken by the Health and Safety Laboratory,[5] atmospheric stable lead concentrations (as monthly averages) vary within wide limits, the extremes being below 0.01 mcg/m^3 in arctic or oceanic areas and 3.4 mcg/m^3 in New York City. Since the radiolead concentration varies within much narrower limits,[6] the specific activity of atmospheric lead is also highly variable and can be expected to attain quite low values in urban centers with heavy lead pollution. A compilation of published data regarding the specific activity of lead in the environment is presented in Table I.

The aim of the present study was to examine the extent to which information concerning the specific activity of lead can help in understanding the sources, dispersion and accumulation of lead in the environment and along food chains. The specific activity of lead was therefore measured in environmental samples, beginning with aerosols and continuing on through plants and animals to man.

EXPERIMENTAL

^{210}Pb was determined in most cases by measuring the alpha activity of the daughter product, ^{210}Po, according to a method previously described[11] in which ^{209}Po was utilized as tracer. Air filter samples were also analyzed by counting the beta activity of ^{210}Bi, according to the method published by Jaworowski.[1] The samples were collected and analyzed during the course of several years. All radioactivity values were corrected to the time of sampling.

Aerosol samples from Helsinki were collected by filtering air on the roof of the Department of Biochemistry building, which is centrally located in the city. The sampling point is 13 m above street level. Samples from Nurmijärvi (Southern Finland) and Sodankylä (Lapland) were supplied by the Institute of Meteorology and were collected 1.5 m above ground level. Whatman G/F glass fiber was employed as filtering material. After treatment of the filters with HNO_3-$HClO_4$, the insoluble residue was removed by filtration and the ^{210}Po content was then determined. The filters had been allowed to stand sufficiently long to permit approximate equilibration between the ^{210}Po and the ^{210}Pb contents.

TABLE I

Review of published data on specific activity of ^{210}Pb in environmental samples

Type and location of sample	Specific activity (pCi ^{210}Pb/mg Pb) Average	Range	Reference
Aerosol samples[a]			
Manhattan, N.Y. (street)	7		7
Manhattan, N.Y. (roof)	13		7
Tuxedo, N.Y.	67		7
Rain water			
Argonne, Ill.	78	16–128	8
Rural area, Mich.	112	53–143	8
Rural area, Poland	40		1
Sea water (particulate samples)			
Pacific Ocean, surface waters	257		9
Pacific Ocean, deep waters	980	630–1300	9
Atlantic Ocean, surface waters	160	32–365[b]	9
Fish			
Fish protein concentrates	126	41–274	10
Samples of anchovy and saury	90	34–135	10
Terrestrial animals			
Bovine bones (Poland)	45		1
Bovine bones (U.K.)	80		1
Human samples			
Soft tissues (Poland)	3.9		1
Bones (Poland)	17.5		1
Hair (Poland)	22.2		1
Bones (U.K.)	4.2		1
Hair (U.K.)	2.1		1

[a] Annual mean for 1969.
[b] Specific activity increases with distance from coast.

Stable lead was determined by means of atomic absorption spectrophotometry. The lead content of the aerosol and plant samples was measured directly from the aqueous solution. For human tissue samples, a solvent extraction procedure was normally employed in which ammonium pyrrolidinediethyldithiocarbamate was used as chelating agent.

The majority of the lichen, plant and reindeer samples, as well as some of the other samples, were collected in connection with a study of polonium and radiolead in arctic food chains, and most of the radiolead determinations were performed during the progress of that work.[12]

RESULTS AND DISCUSSION

Table II presents the results of analysis for the aerosol samples. Due to the high level of stable lead, the specific activity of ^{210}Pb in the Helsinki samples is, in most cases, low (about 20 pCi/mg). The samples from Nurmijärvi and

TABLE II

Specific activity of ^{210}Pb in aerosol samples from three localities in Finland. Samples were collected over 24-hr periods during 1969–1970. Nurmijärvi is situated in a rural area of southern Finland; Sodankylä is in Lapland.

Locality	Date of collection	Weather conditions during collection	Stable lead (mcg/m³)	^{210}Pb (pCi/1000 m³)	Specific activity (pCi/mg)
Helsinki	October 7–8		0.37	10.2	28
Helsinki	October 28–29	Variable	0.20	4.2	21
Helsinki	December 4–5	Stable, cold	0.72	13.3	18
Helsinki	December 9–10	Unstable, cloudy	0.35	8.0	23
Helsinki	March 2–3	Slight snowfall, wind from SE	0.091	22.5	250
Nurmijärvi	January 18–19	Stable, cold (high pressure)	0.039	41.3	1060
Nurmijärvi	July 22–23	Stable, warm (high pressure)	0.014	10.7	760
Nurmijärvi	January 3–4	Unstable (low pressure)	0.029	6.5	220
Nurmijärvi	July 7–8	Unstable (low pressure)	0.12	3.5	29
Sodankylä	January 18–19	Stable, cold (high pressure)	0.016	39.1	2450
Sodankylä	July 28–29	Stable, warm (high pressure)	0.013	8.4	760
Sodankylä	January 4–5	Unstable (low pressure)	0.035	3.7	106
Sodankylä	July 6–7	Unstable (low pressure)	0.031	4.9	160

Sodankylä were especially selected to represent situations of high pressure (stable) or low pressure (unstable) weather conditions of several days' duration.‡ Weather obviously has a pronounced effect upon radiolead concentration: it appears that stable weather conditions result in the accumu-

‡ We are indebted to Mr. R. Mattson, Institute of Meteorology, for these aerosol samples.

lation of radon and its decay products in ground level air, thus causing the specific activity of ^{210}Pb to rise. The samples from Helsinki do not reveal the same pattern. This may be explained by the fact that, during stagnant weather, stable lead also accumulates in the urban atmosphere and thus the specific activity remains low during stable weather as well.

The average specific activity of lead "fallout" may presumably be found in lichen, which obtains its minerals primarily from the air and rain water. However, the slow growth of lichen permits some radioactive decay of its ^{210}Pb content. Table III presents the specific activity of lichen and plant

TABLE III

Specific activity of ^{210}Pb in lichen, plant and animal samples from Finland. Samples were collected between 1965 and 1969

Species	Locality	Stable lead (mcg/g dry wt)	^{210}Pb (pCi/g dry wt)	Specific activity (pCi/mg)
Lichen (*Cladonia alpestris*)	Enontekiö, Lapland	9.3	10.2	1100
Lichen (*Cladonia alpestris*)	Inari, Lapland	4.8	5.7	1180
Lichen (*Cladonia alpestris*)	Enontekiö, Lapland	11.2	6.6	600
Lichen (*Cladonia alpestris*)	Koli, E. Finland	13.4	6.4	490
Lichen (*Cladonia alpestris*)	Virolahti, S.E. Finland	20.8	6.7	330
Lichen (*Cladonia alpestris*)	Tuusula, S. Finland	20.8	7.0	340
Lichen (*Cladonia alpestris*)	Porvoo, S. Finland	19.0	5.5	290
Lichen (*Cladonia alpestris*)	Loppi, S. Finland	19.4	6.5	340
Clover (*Trifolium hybridum*)	Tuusula, S. Finland	11.5	0.83	72
Grass (*Deschampsia flexuosa*)	Inari, Lapland	3.2	0.41	130
Birch, leaves (*Betula verrucosa*)	Koli, E. Finland	2.6	0.46	180
Birch, leaves (*Betula verrucosa*)	Wessö, S. Finland	3.4	0.56	165
Mountain ash, leaves (*Sorbus aucuparia*)	Koli, E. Findland	2.9	0.28	95
Mountain ash, leaves (*Sorbus aucuparia*)	Wessö, S. Finland	3.4	0.35	110
		(mg/kg wet wt)	(pCi/kg wet wt)	
Reindeer liver, 10 animals	Salla, Lapland	1.27	927	730
Reindeer blood, 10 animals	Inari, Lapland	0.085	86	1010
Reindeer blood, 10 animals	Inari, Lapland	0.106	95	900
Reindeer meat, 10 animals	Inari, Lapland	0.051	(7.3)[a]	(140)
Ox meat, 5 animals	S. Finland	0.015	(0.62)[a]	(41)
Ox liver, 5 animals	S. Finland	0.085		

[a] Not same sample as that utilized for stable lead determination.

samples collected from various regions of Finland. The specific activity of lead "fallout" in Lapland appears to be approx. 1000 pCi ^{210}Pb/mg Pb, whereas in Southern Finland, as a consequence of higher stable lead concentrations, the corresponding values are only about 350 pCi/mg. The "natural" (prehistoric) specific activity of atmospheric lead, calculated upon the basis of estimates by Patterson,[3] was of the order of 20,000 pCi/mg.

Samples of green plants and leaves exhibit considerably lower specific activities (70–180 pCi/mg) than lichen. This difference may be due not only to the uptake of lower activity lead from the ground, but also, at least partly, to the higher specific activity of lead in the winter "fallout" collected by lichen. Mattsson,[13] for example, reports values five times higher in winter than in summer for the concentration of atmospheric ^{210}Pb in Sodankylä, Lapland.

Contradictory results have been reported concerning the uptake of lead in plants. Wilson and Cline[14] observed very low values for the uptake of ^{210}Pb added to soil as lead chloride, and Berger *et al.*[15] claim that lead is not taken up from the soil at all. On the other hand, Tso *et al.*[16] concluded that most of the ^{210}Pb contained in tobacco plants was taken up by the roots, and Lagerwerff[17] reports that approx. half of the lead in the green portion of the radish is obtained from the soil. These and other data were reviewed by the Committee on Biological Effects of Atmospheric Pollutants,[4] which arrived at the conclusion that lead is both absorbed through the roots from the soil as well as deposited upon the leafy portions of the plants from the air, but that very little atmospheric lead reaches the edible parts of plants, except edible leaves (lettuce, spinach).[18]

The above-mentioned contradictions may be partly due to the difficulty of defining soluble (available) lead in soil. No data appear to have been published regarding the specific activity of this soluble lead. In one measurement by Ter Haar *et al.*,[8] the specific acitivty of total lead in soil near Argonne, Illinois, was found to be about 75 pCi/mg. Considering the location and the rain water data by the same authors (Table I), corresponding values for unpolluted areas may be expected to be higher. Mention may also be made that an analysis of eight recent sediment samples from the Baltic yielded an average specific activity value of 170 pCi/mg.[19]

It appears, then, that the specific activity of environmental lead at the beginning of the food chains is of the order of 100 pCi/mg or somewhat higher. An exception is the arctic food chain starting with lichen, where the specific activity seems to be closer to 1000 pCi/mg. Samples of reindeer also exhibit very high specific activities, up to 1000 pCi/mg. This is obviously due to the fact that reindeer feed almost exclusively on lichen from October to May. The reindeer samples were taken in winter, predominantly in March.

The data presented in Tables I and III–V reveal that the specific activity

decreases along the food chain plant—animal—man by approx. one order of magnitude, the reported values for human samples of normal Western populations ranging from about 5 to 25 pCi/mg. Radioactive decay explains only a small portion of this decrease: the principal cause must be the addition of low activity ("technological") lead along the food chains. In order to decrease the specific activity by one order of magnitude, the original amount of environmental radiolead has to be diluted by a tenfold excess of inactive lead. Should this be the actual case, it is important to investigate the sources of this additional lead contamination.

TABLE IV

Specific activity of ^{210}Pb in human samples from Lapland and southern Finland (samples were collected in 1966)

Type of sample	Locality	No. of persons sampled	Stable lead (mcg/ 100 ml)	^{210}Pb (pCi/ 100 ml)	Specific activity (pCi/mg)
Blood					
Lapps (reindeer breeders)	Inari, Lapland	70	13	0.65	50
Southern Finns, *average*	Helsinki	33	14	0.27	20
			(mg/kg wet wt)	(pCi/kg wet wt)	
Placenta					
Lapps	Inari and Utsjoki	10	0.073	1.9	26
Southern Finns	Helsinki	9	0.048	0.84	18
Bone					
Southern Finns	Helsinki	1	1.7	45	26
Average of all samples from southern Finland		43			20

Table V attempts to compare some widely varying data on the lead content of the human diet in the light of specific activity considerations. The first column of this table states the radiolead concentrations of various dietary items as reported by Morse and Welford.[20] The value of 1.2 pCi ^{210}Pb/day, obtained for the average New York diet in the above study, is somewhat lower than that reported by other authors. For example, Holtzman[21] and Hill[22] found 1.8 and 3.2 pCi/day, respectively, in excretion studies performed

with a limited number of subjects, and Magno *et al.*[6] reported 1.7 pCi/day based upon the analysis of a large number of U.S. diets. Replacing the figure of 1.2 pCi/day by 1.7 pCi/day would increase the corresponding specific activities by a factor of 1.4, but the overall picture would remain approximately the same.

The stable lead data presented in Table V are based upon three dietary surveys reported by Bogen.[23] These surveys give two differing sets of values for the average stable lead content of the diet in New York City. The first survey, undertaken during 1966–67, indicated an average daily intake of 285 mcg of lead, whereas the second and third surveys (1968–69 and 1970–71) revealed considerably lower dietary intakes of only about 85 mcg Pb/day. Bogen suspects contamination of the samples in the first survey.

TABLE V

Specific activity of ^{210}Pb in some components of the human diet, calculated upon the basis of radiolead data by Morse *et al.*[20] and stable lead data by Bogen[23]

Dietary item	^{210}Pb (pCi ^{210}Pb/kg food)	Specific activity (pCi ^{210}Pb/mg Pb)	
		Based on "high lead values"	Based on "low lead values"
Fresh vegetables	1.1	9	19
Canned vegetables	0.44	1.0	13
Fresh fruit	0.40	6	6
Bakery products	1.8	4.6	18
Flour	1.3	32	59
Meat	0.49	1.2	8
Fresh fish	0.39	2.4	8
Daily diet[a]	1.25 pCi	4.2	12.5
Daily absorption[b] to blood	0.21 pCi	7.0[c] 8.5[d]	15[c] 24[d]

[a] Including drinking water, 0.05 pCi ^{210}Pb and 15 mcg stable lead per day (refs. 20 and 4).
[b] Including inhalation. Retention of inhaled lead is calculated in the same way as in ref. 20.
[c] Assuming 1 mcg stable lead per m^3 inhaled air.
[d] Assuming 0.1 mcg stable lead per m^3 inhaled air.

Lead intake values of approx. 300 mcg/day have until now been generally accepted as valid,[24] and even higher values were recently published (e.g. Lehnert *et al.*[25] estimate the dietary intake of lead in Germany to be about 500 mcg/day). On the other hand, the "high level values" of Bogen[23] signify very low specific activities in some cases, such as 1.2 pCi/mg for meat and 1.0 pCi/mg for canned vegetables (Table V, second column). These low

activities are difficult to explain without assuming heavy contamination by technological lead. It should be noted that, in the first survey by Bogen, meat (including poultry) and canned vegetables alone accounted for almost half of the total lead in the diet, whereas the later surveys indicated eight times lower lead concentrations for the same items.

The specific activities presented in the third column of Table V are based upon the second and third surveys by Bogen[23] ("low lead values"). These activities appear to be more consistent with the values obtained by us for some food samples (Table III) and human samples (Table IV).

Additional analytical results are apparently necessary in order to resolve the discrepancy between the values for stable lead intake. Especially those items which have yielded very low specific activities should be investigated closer.

In conclusion, it is worth noting that, should the lower dietary values for stable lead be correct, then the proportion of atmospheric lead in the human intake of lead will be higher. In heavily polluted areas with more than approx. 1.5 mcg Pb/m^3 (cf. ref. 5), inhalation will be the major avenue of lead intake for man. Conversely, should the higher dietary stable lead values be correct, then the discrepancy between the specific activities of environmental and dietary lead becomes still greater, and consequently, more technological lead must be assumed to enter the food chains to "dilute" the ^{210}Pb activity of environmental lead.

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