INSTITUTE OF NUCLEAR RESEARCH
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STABLE AND RADIOACTIVE LEAD IN ENVIRONMENT AND HUMAN BODY

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1. INTRODUCTION

Great amounts of stable lead, of the order of several milion tons, have been introduced into the biosphere over the last several decades, as a result of the rapid growth of industrial activities and motorization. More recently, there has also been an increase in the level of radioactive lead in the biosphere, due to the nuclear test explosions. Epidemiological studies on intoxication with lead at the levels present in the natural environment are only in their initial stage. However existing data indicate, that chronic intoxications with small doses of stable lead can lead to neoplasmatic and other degenerative diseases, especially of the nervous system and to serious disturbances of development /WARREN 1961, KANCHEFF et al. 1964, EMRG et al. 1964/. Such chronic low-level intoxications with lead may afflict the whole population of highly developed countries /PATTERSON 1965/.

All the inhabitants of the Earth are subject to chronic contamination with small amounts of natural radioactive lead-210, a member of the uranium family. This radioactive lead contributes the highest radiation dose to the human body of all the "bone-seeking" natural radionuclides.

The hazards to which both types of lead may expose the human population, were recently recognized in a number of publications. However the problem has not so far been considered monographically. The present paper is a monograph of the stable and radioactive lead

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in the environment and human body. The aim of this study is to determine the level of stable and radioactive lead in typical components of the biosphere and in the organism, as well as to examine their sources and temporal and geographical distribution. The source of stable lead in the biosphere is different from that of radioactive lead. The former originates from the deposits in the Earth's crust whereas the latter comes predominantly from atmospheric fallout. However, in the living organism both these forms of lead are intermingled and run the same biological course. For this reason, in this monograph, both types of lead are dealt with jointly.

Over several years the present author carried out environmental and metabolic studies on stable and radioactive lead. The majority of his results concerning radioactive lead have already been published /JAWOROWSKI 1963 A, 1963 B, 1963 C, 1963 D, 1964, 1965 A, 1965 B, 1965 C, 1966, JAWOROWSKI et al. 1967 A, 1967 B, 1967 C, HILL et al. 1961/. The results of the author's studies on stable lead are published, for the first time, in the present monograph.

Because of the monographic character of this study, the author did not confine himself to the results of his own investigations, but included also the results of other authors, in an endeavour to achieve as reliable as possible world mean values and data on the geographic variations.

As investigations on the temporal distribution of stable and

radioactive lead have so far been performed only on a very limited scale, the present writer paid special attention to this problem, collecting large sets of samples, which enabled him to carry out this kind of study. Thus the results pertaining to the temporal distribution of stable and radioactive lead, presented in this monograph are based for the most part on the present writer investigations.

The materials used by the writer in this study consist of:

1. Recent human bones collected in the Warsaw and London regions; old human bones covering the period from XIII to XIX century, collected in underground crypts in different churches in Southern Poland; III century human bones collected in a cave near Kroczyce, province of Katowice, in Poland.
2. Recent human hair from Poland and Great Britain.
3. Recent red-deer antlers, collected in Białowieża and Popielno in Northern Poland; recent bovine bones from the Warsaw region.
4. Recent lichens of the family Cladoniaceae collected along the 20th Meridian /East/ in Spitsbergen, Sweden, Poland, Czechoslovakia, Hungary and Bulgaria.
5. Recent rainwater samples collected in Świerk near Warsaw; fossil rainwater samples collected from two glaciers in the Tatra mountains, Southern Poland; mineral water samples collected in Western Poland.
6. Recent wine from Hungary.
7. Air samples collected in Świerk, near Warsaw.

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8. Averaged coal and coal fly ash samples from a power station in Warsaw.

9. Mongrel dogs and rabbits used for metabolic studies.

The following persons and institutions kindly helped the writer in collecting the material used in this study: Professor K.Kowalski /Institute of Systematic Zoology, Polish Academy of Sci., Kraków/, Professor W.V.Mayneord /Physics Department, Institute of Cancer Research, London/,Metropolitan Curia of Warsaw and Diocesal Curia of Kraków, Professor R.W.Schramm /leader of the Polish 1965 Spitzbergen Expedition/, Professor Z.Jaczewski /Research Station of the Polish Academy of Sci. in Popielno/, Dr. S.Borowski /Mammal Research Institute, Białowieża/. Dr. J.Zielińska /Department of Botany, University of Warsaw/, Dr. M.Trepacz-Cabalska /Department of Archeology, Jagiellon University/, Dr. J.Nowak /Botanical Institute Kraków/, Professor T.Pocs and Dr. Klara Verseghy /Natural History Muzeum, Budapest/.

It is a great pleasure to acknowledge the author's great indebtedness and thanks to all these persons and institutions.

During the years of his work the present author had the priviledge of discussing with the late Professor Edward Kowalski /Head of the Department of Radiobiology and Health Protection of the Institute of Nuclear Research, Warsaw/ many problems connected with this study. Professor Kowalski had ever been willing to give the writer every possible assistance. His continued advice and encouragement have been of the greatest value for the writers investigations.

2. HISTORICAL REVIEW

Lead has been known to man for several thousand years. It was used in Anau I 4200 B.C. and in the third millenium B.C. became usually a by-product of silver production /GILFILLAN, 1965/. The first mentions of this metal in the Bible were probably made about 750 B.C. /Job XIX, 23, 24/. Hesiod, who lived in Greece in VIII century B.C. used lead sheets for writing on /FRIEND, 1951/. In Egipt, lead was widely used at the time of the New Kingdom, i.e. about 1580 B.C. while in Troi it was used as early as 2200 B.C. /FRIEND, 1951/. In Greece, lead was produced in superabundance and hence used wholesale. It was estimated that 2.100.000 tons of lead were produced there over a period of 300 years /HOFMANN , 1885/. In Rome, this metal was used on a large scale for water piping and for kitchen utensiles and lead intoxication of the upper classes was very common there, which was also the case in Greece /GILFILLAN, 1965/. In Great Britain, lead was produced and used during the Roman invasion. In the early Middle Ages it was produced largely in Middle Europe, in Czechoslovakia and Poland mainly for export /HENSEL, 1965/.

The toxicity of lead has been known since antiquity, and it was used by the Egyptians as a homicidal poison /DE TREVILLE, 1963/. Widespread intoxication with lead was caused by the common practice of treating wine with metallic lead and by litharge or lead acetate as a "sweetener". In Germany in 1548 and 1577 decrees

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were issued invoking the death penalty for any one adulterating wine with lead /DE TRÉVILLE, 1963/.

At present lead is widely used in paints, crystal and optical glass, as typing metal, cable sheating, solder, in storage batteries, in insecticides, ceramics, manufacturing, construction industry, collapsible tubes, railroads, brass manufacturing, as tetraethyl lead in gasoline and in a thousand other things used in every day life. In 1960, the world lead production reached 2.623 thousand tons, /KAWIŃSKI et al., 1963/. In 1962, the United States total lead consumption was 1,075 thousand tons, a typical annual figure for the last years /ZIEGFELD, 1964/.

As an illustration of the time trends in the rate of lead production, the data pertaining to the lead resources of Southern Poland may be used /GAŁKIEWICZ, 1956, 1957, 1959/. Of a total of 4 million tons exploited in this region, 10% was produced from antiquity to the beginning of XIX century, 5% in the first half of XIX century, 30% in the second half of XIX century and about 55% in the first half of XX century. New applications for lead in modern civilisation are still on the increase, and though old lead is partially reused /as e.g. in the storage battery industry/, its global production steadily grows.

Since the beginning of his evolution man was subject to contact with minute amounts of stable and radioactive lead, present in his natural environment. In this monography it will be shown that as a result of human activities the stable lead

contamination of the environment has been drastically increased. It appears that the radioactive lead content in the biosphere has also increased during the last decade.

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3. ABUNDANCE AND ISOTOPIC COMPOSITION OF LEAD

The cosmic abundance of lead expressed in number of atoms per 10^6 atoms of silicon is < 2. Due to the pronounced differentiation of terrestial matter the abundance of lead in the Earth's crust is higher than the cosmic one and reaches a value of 54 in igneous rocks /RANKAMA, 1954/.

The absolute abundance of lead in the Earth's crust increses steadily due to its production by the three natural radioactive families i.e. uranium, actinium and thorium. Their ultimate stable decay products are respectively ^{206}Pb, ^{207}Pb and ^{208}Pb. The fourth stable isotope, ^{204}Pb is not known to be connected with any natural nuclear process.

The isotopic composition of stable lead may vary depending on its origin. /TABLE 1/.

TABLE 1. Percentage isotopic constitution of stable lead
/After RANKAMA, 1954/

Isotope	Greenland	unknown origine	Tetramethyl lead	Tetraethyl lead
^{204}Pb	1.55	1.37	1.31	1.36
^{206}Pb	22.51	25.15	26.79	25.42
^{207}Pb	22.60	21.11	20.55	21.11
^{208}Pb	53.34	52.38	51.35	52.10

As may be seen from this table the abundance rises from ^{204}Pb to ^{208}Pb along the increasing magic numbers of the isotopes, which is in accordance with the neutron-capture hypothesis of the formation of the nuclei. /RANKAMA, 1954/.

Usually the following forms of natural lead are distinguished.

1/ Original lead, which was formed when the elements were formed. Its proportion in a lead sample is indicated by the content of the ^{204}Pb isotope.

2/ Ordinary or common lead, i.e. the lead which is obtained from nonradioactive minerals. It is composed of original lead and varying small amounts of radiogenic lead.

3/ Radiogenic lead, consisting of ^{206}Pb, ^{207}Pb and ^{208}Pb is the stable end product of the natural radioactive families.

4/ Primary lead, i.e. a lead present in a mineral at the time of its formation.

Five radioactive isotopes of lead occur as members of the natural radioactive families. These are ^{214}Pb /RaB/ and ^{210}Pb /RaD/ in the uranium family, ^{211}Pb /AcB/ in the actinium family, ^{212}Pb /ThB/ in the thorium family, and ^{209}Pb in the neptunium series. Among them ^{210}Pb has a half-life of 20 years, the half-lives of the rest being in the order of minutes or hours. /NESMEYANOV, et al., 1956/.

Due to its long half-life lead-210 and its daughters bismuth-210 and polonium-210 can concentrate in various compartments of environment, exposing living organisms to their ionisating ra-

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diation /TABLE 2/.

The abundance of ^{210}Pb in ordinary lead was studied by several authors. Mc LENNAN in 1907 was the first who found that old common lead is less radioactive than the new one. RUTHERFORD et al. suggested in 1930 that while radium present in the lead ore would be efficiently separated from the lead in the smelting process, ^{210}Pb would remain. ANDERSON et al. /1964/ found the following ^{210}Pb concentrations in common lead produced from different ores: leads from Nebraska 40 pCi/g, from Peru 25 pCi/g, from Utah 25 pCi/g, from Canada 10 pCi/g, from Missouri, Illinois, Mexico and Australia less than 2 pCi/g. Similar concentrations ranging from 4 to 38 pCi/g were found by WELLER et al. /1965/ in common lead originating from the United States and other regions.

Besides the natural radioactive lead isotopes there are the following artificially made ones: ^{198}Pb, ^{199}Pb, ^{200}Pb, ^{201}Pb, ^{202}Pb, ^{203}Pb, ^{204}Pb, ^{205}Pb, ^{207}Pb. Except for professional intoxication these artificial radioleads are not regarded as contributing much to the radiation hazards of the population.

TABLE 2. The uranium family

Radioelement	Nuclide	Radiation	Half-life
Uranium I	U-238	α	4.50×10^9 y
Uranium XI	Th-234	$\beta-$	24.10 d
Uranium X2	Pa-234	$\beta-$	1.14 m
Uranium II	U-234	α	2.52×10^5 y
Ionium	Th-230	α	8.0×10^4 y
Radium	Ra-226	α	1622 y
Ra emanation, Radon, Niton	Rn-222	α	3.825 d
Radium A	Po-218	$\alpha, \beta-$	3.05 m
99.96% 0.04%			
Radium B	Pb-214	$\beta-$	26.8 m
Astatine	At-218	$\alpha, \beta-$	1.5 s - 2 s
99.99% ~0.01%			
Radon	Rn-218	α	0.019 s
Radium C	Bi-214	$\beta-, \alpha$	19.7 m
0.04% 99.96%			
Radium C'	Po-214	α	1.637×10^{-4} s
Radium C''	Tl-210	$\beta-$	1.32 m
Radium D	Pb-210	$\beta-$	25 y
Radium E	Bi-210		5.02 d
100% 5×10^{-5}%			
Radium F	Po-210	α	138.3 d
Thallium	Tl-206	$\beta-$	4.23 m
Radium G	Pb-206	Stable	-

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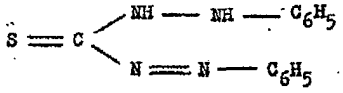
4. METHODS OF DETERMINATION OF STABLE AND RADIOACTIVE LEAD

4.1 METHODS OF DETERMINATION OF STABLE LEAD

During the past thirty year numerous methods for the determination of lead in different materials have been developed. Usually the separation of lead from the samples is performed by precipitation, exctraction, ion exchange or by electrolytic procedures. For determination of traces of lead the commonly used methods are: colorimetry, spectrography and polarography. The selection of a particular analytical procedure will depend upon the nature of the problem, available equipment and of coarse on personal preferences. A critical review of the analytical methods of lead determination in biological and related materials has been prepared by CHOLAK /1964A/. According to this author for environmental studies and determination of lead in the body the wet chemical methods emploing dithizone appear to be the methods of choice, mainly because of their convenience, economy and accuracy of analyses.

The detection limit of the dithizone colorimetry method of about 0.5 µg is usually satisfactory for such aims as the determination of lead in air, water, plants and animals.

Dithizone or diphelylthiocarbazone or phenylazone or phenylazothionoformic acid



first prepared in 1878 by Emil FI
proved to be highly valuable for
of some twenty heavy metals. This
been developed by Helmut FISCHER
were the first to develope the me
quantities of lead in biological
considerable number of procedures
for lead determination has been p
procedure described by JAWOROWSKI
a modified version of the method

Dithizone is a violet-black
solvents giving in dilute solutio
colour in more concentrated ones.
dithizone dissolves giving a yell
dithizone forms metal dithizonate
pletely dissociated /SANDELL, 195
soluble in chloroform and other o
nate gives in chloroform a cinnab
wave length of maximum absorption
/FISCHER et al., 1935/. The dithi
be readily dissociated and lead t
when the chloroform is shaken wit
solutions containing citrate and
extracted to chloroform, the only
bismuth, thalium and stannous tin

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cessary for the separation these three interferring metals from lead.

When extraction of lead dithisonate to the chloroform phase is made from the aqueous phase at pH 11.5, the recovery of lead is complete and the amount of excess dithizone left in chloroform is much reduced /SNYDER, 1947/. This enables colorimetric determinations to be made at a wave length of 5200 A without appreciable interference of dithizone, which has its maximum absorption wave length at 6200 Å.

This mixed color method depends to some extent on such factors as variation in dithizone content in the lead dithizonate solutions in chloroform, impurities in the dithizone used, degree of instability of dithizone solution and contamination from apparatus, reagents and atmosphere. In practice, the influence of these factors may be limited by running the blank analyses of each batch of samples and using the density of the blank samples for determination of the zero intercept of the absorption curve.

In material with high calcium magnesium and phosphate content which precipitate at higher pH, the separation of lead as dithizonate cannot be made from strongly basic aqueous solutions. Usually in such cases a preliminary separation is made by diethylammonium diethyldithiocarbamate extraction to chloroform from acidic solution, followed by the dithizone determination of lead.

The samples containing organic material must be ashed before analysis. This may be done by dry or wet ashing. The adventages of

dry ashing are: simplicity, freedom from blank corrections and the possibility of handling large samples. The advantages of wet ashing are: speed of oxidation, low temperature involved, and freedom from loss by retention of trace amounts of lead on solid material in the system. The disadvantage of dry ashing is loss of lead by volatilization /about 95 per cent when sodium chloride is present in the sample, GORSUCH 1959/. The errors in wet digestion are caused by co-precipitation of soluble lead compounds on sulphates formed during ashing. The studies of GORSUCH on the recovery of lead after destruction of organic matter revealed, however, that wet ashing methods are safer, recoveries being between 90 and 100 per cent.

Therefore a wet ashing method was included in the procedure used in this laboratory. The details of this procedure are given in APPENDIX I.

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4.2 METHODS OF DETERMINATION OF RADIOACTIVE LEAD

Direct measurements of radioactive lead -210 in biological material are usually not performed because of the poor detectibility of its radiation /beta max. 0.018 MeV, gamma 0,047 MeV/. However, indirect determinations can be made by measurements of the comparatively highly energic beta particles /beta max. 1.17 MeV/ of bismuth-210, the short living daughter of lead-210, or by measurements of alfa particle of the polonium-210, the next daughter of lead--210. This last method is used in the majority of studies on lead--210 in the biosphere. It has however a serious disadvantage, which is sometimes overlooked. Due to the comparatively long half-lives of lead-210 /20 years/ and polonium-210 /140 days/ determination of lead-210 with this method involves either assumptions concerning the state of equilibrium of these nuclides in the samples to be analysed, or careful examination of changes of activity in the samples, which have to be made over lenghty periods. Furthermore, characterisation of polonium-210 at background levels is no simple task and may require the application of highly-developed alpha--spectrometric techniques /MAYNEORD et al., 1959/. Since the advent of low background beta counting techniques the advantage of the low detection limits of alfa counters is no longer a sufficient reason for accepting the procedure of electrodeposition of polonium on silver or other metal foils, as the method of choise for determination of lead-210.

An alternative approach to the determination of lead-210 is provided by its immediate daughter, the relatively short-lived bismuth-210 / T 1/2 = 5 days / which emits beta particles suitable for detection in any type of low background counter. The advantage of this method is the comparatively short time necessery to approach a partial radioactive equilibrium state between the parent lead-210 and daughter bismuth-210. After 15 days of build-up of bismuth-210 about 86 per cent of radioactive equilibrium will be reached in the lead-210-bismuth-210 system. This may be compared with about 59 per cent equilibrium reached in lead-210-polonium--210 system after 6 months. Furthermore, in bismuth-210 method identification of the short half-life of this nuclide presents no practical problem and can easily be made in a week. In the case of polonium-210 the same takes a few months. For determination of radioactive bismuth-210 it is necessery first to separate lead-210 together with stable lead and then isolate the radioactive bismuth--210. These two separations ensure the high radiochemical purity of the counting sample.

For these reasons the present author accepted for determination of lead-210 a procedure involving measurement of the bismuth-210 component. The separation of bismuth-210 from the bulk of stable lead containing also lead-210 may be made or by cupferron excraction of bismuth from acidic aqueous medium to chloroform /BODE et al., 1952/ followed by precipitation of bismuth as phosphate /JAWOROWSKI, 1963 A/ or by electrolytic deposition on pla-

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tinum cathode /JAWOROWSKI et al., 1967/. Both methods were applied successfully for determination of lead-210 in different materials and are described in detail in APPENDIX II.

The main advantages of the internal electrolysis method /JAWOROWSKI et al., 1967/ are the simplicity of operations and the lack of self-absorption in the counting sample. Carrier-free bismuth-210 can also be prepared in this way. This method may be used without preliminary dithizone separation of lead, in cases where the ashed sample can be dissolved directly in the electrolyte /e.g. hair or metallic lead/. As this method was used for the majority of determinations of lead-210 in this laboratory it will be described here in more detail.

The method is based on the procedure used by HOLLARD /1903/ for separation of zink from nickel, which was then modified by SAND /1930/ for separation of stable bismuth from lead. In this method bismuth dissolved in an electrolyte solution is deposited on a cathode in a galvanic cell without use of an external source of electric current. For this process SAND proposed the term "internal electrolysis". In our method we used as cathode 0.3 mm thick platinum foil of 17 mm diameter connected with 30 mm long platinum wire of 0.5 mm diameter. The free end of the wire was connected with the lead anode by an easily demountable screw link. The anode was enclosed in a glass pipe with a G3 sintered glass window. The electrodes were immersed in the electrolyte solution in a 50 ml beaker placed in a water bath. For the full procedure for determi-

nation of lead-210 in biological and other samples see APPENDIX II.

As may be seen from FIGURE 1: the best recoveries of bismuth-210 have been obtained by a 4 % concentration of nitric acid in the electrolyte solution.

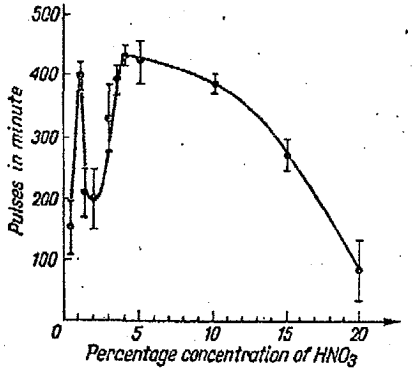


FIGURE 1. Influence of HNO_3 concentration in the electrolyte solution on bismuth-210 deposition on the Pt electrode.

The best deposition of bismuth-210 on the platinum cathode was found at a temperature between 50° and 70°C /FIGURE 2/. It was observed, however that at 70°D bismuth is more firmly attached to the platinum cathode than at lower temperatures. Satisfactory deposition of bismuth-210 was observed after 30 minute electrolysis /FIGURE 3/, however, the best reproducibility of results was found after 60 minute deposition. Therefore, as optimal working conditions are chosen: a nitric acid concentration in a electrolyte solution of 4%, a temperature of 70°C and a deposition time of 60 min.

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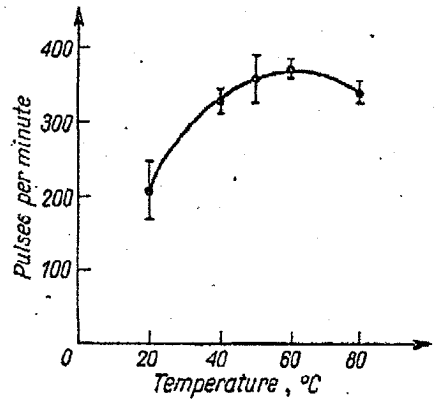


FIGURE 2. Influence of temperature on bismuth-210 deposition on the Pt electrode.

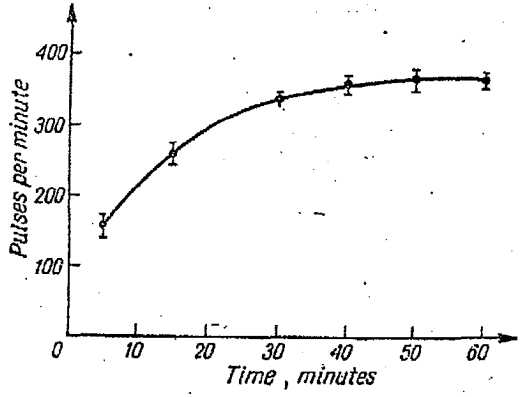


FIGURE 3. Influence of the electrolysis time on bismuth-210 deposition on the Pt electrode.

For determination of percentage recovery of bismuth-210 a "two tests method" was applied. In this method determination of a substance by a procedure is performed twice in the same sample. If the total amount of a substance in the sample is x, and the fraction of the substance recovered during analysis is y, then the amount recovered after the first determination, a, is $a = x.y$ and the amount recovered after the second determination, b, is $b = /x-a/.y$, assuming that in both determinations y is the same. The percentage recovery can be then calculated as

$$\text{percentage recovery} = \frac{a - b}{a} \quad 100$$

The recovery studies were performed in 16 distilled water samples containing 30 mg of stable lead and spiked with equal amounts of bismuth-210 in equilibrium with lead-210 and polonium-210. The results expressed as beta counts per minute of both sides of the platinum cathode are presented in TABLE 3. The average relative standard deviation of counting in this experiment is considered as $\pm$ 0.6 per cent for the first determination and $\pm$ 8 per cent for the second. The mean recovery of bismuth-210 was found to be 95.5 $\pm$ 2.5 per cent. The contribution of counting errors to the overall analytical error 1.5 per cent. A recovery of 95.5 $\pm$ 2.5 per cent has also been found in six samples containing 1 g of stable lead each. For recovery studies in bone, twelve 1 g

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bone samples were spiked with bismuth-210, wet ashed and dissolved in 25 ml portions of electrolyte solution in which bismuth-210 was determined starting with step 5. of the SEPARATION OF BISMUTH-210 BY INTERNAL ELECTROLYSIS /APPENDIX II./ The mean bismuth-210 recovery found in these samples was 92.0 ± 3.0 per cent.

The selectivity of the procedure was tested against ^{60}Co, ^{65}Zn, ^{90}Sr, ^{99}Tc, ^{106}Ru, ^{131}J, ^{137}Cs, ^{144}Ce, ^{160}Tb, and ^{204}Tl. None of these radionuclides were deposited on the cathode in detectable amounts when samples spiked with doses of nuclides corresponding to about 1000 beta counts per minute each were used in the experiment.

The total amount of stable lead in the reagents used throughout the analysis was 1.2 µg. During 60 minute electrolysis approximately 600 µg of the anodic lead was dissolved in the electrolyte solution. One gram of stable lead contains about 12 pCi of radioactive lead-210 /WELLER et al., 1965/, so the bismuth-210 contribution from both these sources to the activity deposited on the platinum cathode is some 0.007 pCi and can be neglected. Therefore it is not necessary to use an aged lead as anodic material even for low-level studies.

Measurements of radioactivity of Bismuth-210

For determination of radioactive lead-210 and other radionuclides naturally present in biological and related materials it is essential to have a counting method suitable for detecting the

Table 3. Recovery of bismuth-210 on the platinum cathode from the distilled water samples spiked with equal amounts of bismuth-210.

Beta counts per minute of both sides of the cathode		Percentage recovery $/\frac{a-b}{a} \cdot 100/$
First determination /a/	Second determination /b/	
836.8	51.1	93.9
780.9	35.9	95.4
828.8	43.2	94.8
826.1	80.5	90.3
848.0	16.7	98.0
759.2	34.8	95.4
809.5	54.6	93.3
839.8	20.8	97.5
876.6	57.9	93.4
863.5	14.5	98.3
813.0	21.4	97.4
842.1	37.2	95.6
823.5	51.3	93.8
817.2	24.1	97.0
807.4	32.1	96.0
823.4	37.0	95.5 ± 2.5

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levels of activity in the order of several desintegrations per minute. The sensitivity of a counting unit suitable for such a method is limited by the absolute value of background counts and by its statistical fluctuations. Therefore the background should be reduced to a minimum. Reasonable attenuation of the background have been succesfully achieved by many workers by application of heavy iron, lead or chalk shields and anticoincidence systems and by using the Geiger-Mueller tubes or proportional counters as detectors /WADD et al., 1962, ALBERIGI-QUARANTA et al., 1961, GAT et al., 1960/. The background of these units is about 1.0 count per minute. Recently low-background beta-counting units have been described, in which the anticoincidence umbrella is not applied and a thin plastic scintillation counter is used as detector /HARLEY et al., 1962, JAWOROWSKI, 1963 B/. The background of these units is usually of the order of 0.3 pulses per minute. Their outstanding advantage is high counting effectiveness and simplicity of design, these aparatuses being especially suitable for low- - level studies of beta emiting radionuclides, with the exeption or those with low energy emission.

Since 1961 a unit based on this principle is used by the present author in his studies on lead-210. The detailed description of this unit has been previously published /JAWOROWSKI, 1963 B/. Few general informations pertaining its characteristics and performance are given below.

This counter is derived from an EKCO scintillation counter

type N 664 A. The pulses from the phototube pass first to the amplifier and then to the EKCO scaler type N 529 A.

As a detector, a NE 102 scintillating plastic disc was used, 17 mm diameter, 0.3 mm thick, optically coupled with an EMI type 9514 S photomultiplier tube. The shielding consists of a lead castle and chalk layers. The samples in the counting trays or platinum electrodes are mounted on a scintillating plastic disk so that close contact between the sample and detector is ensured.

The background and counting efficiency of this apparatus depends on the phototube potential, amplification gain, discrimination level, environmental patterns and mass of scintillating plastic used.

In 1000 V phototube potential and 45 V discrimination level the average background, measured in 17 hour periods, was found to be 0.4 $\pm$ 0.1 counts per minute, and the counting efficiency as measured against a Nuclear Chcago RT-3 model standard of lead-210 /in equilibrium with biemuth-210/ was 33.5 per cent. The detection limit of the counter was taken to be 0.36 pCi.

Depending on the radioactivity of the sample measurements were made over a period of 1 - 16 hours. The overall uncertainties caused by counting statistics and analytical procedures in studies on environmental and body levels of lead-210 are considered to be in the limits of 20 per cent relative standard deviation at the 90 per cent level of confidence.

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5. STABLE AND RADIOACTIVE LEAD IN ENVIRONMENT

5.1. ORIGIN AND LEVELS OF STABLE LEAD IN ENVIRONMENT

The stable lead present in such important constituents of human environment as the ambient atmosphere, water, soil and food, may have both a natural and artificial origin.

Atmosphere

The natural lead in the atmosphere is introduced there by volcanic activity and meteoric matter, by the wind transfer of soil and sea water and by smoke from forest fires. It is difficult now to establish experimentally this natural level of lead in the atmospheric air, which has been largely polluted by industrial activities, fuel combustion and motorisation.

An estimation of the natural level of lead in the air has been recently made by PATTERSON /1965/. For this estimation contributions from silicate dust were obtained from measurements of dust in rural air /ANONYMOUS, 1963 A/ and from the concentration of lead in silicates. The contribution from volcanic halogen aerosols and volcanic silicate smoke had been estimated /CHOW et al., 1962/ and 10% of this was assigned to an atmospheric reservoir. Of the existing timber acreage of United States less then 0.1% is burned each year by lightning /ANONYMOUS, 1963 A/. Taking this percentage as valid for the total terrestial biosphere and accepting the mean

concentration of lead in terrestial vegetation as about 80 parts per million parts of calcium, PATTERSON /1965/ estimated the upper limit of contribution from that source. The contribution from aerosolic sea salts was calculated accepting a concentration of 0.007 ppm Pb in sea salt and 6×10^{14} g of sea salts in the atmosphere per year /POLDEVAART, 1955/ from which 10% was assigned to an atmospheric reservoir. The imput of meteoric and meteoritic dust to the Earth is about 1×10^{12} g per year /WRIGHT, 1963/ and concentration of lead in this material is about 1×10^{-7} ppm /REED et al., 1960 and PATTERSON, 1955/. Of this imput 10% was assigned to an atmospheric resevoir. The results of these estimations are listed in TABLE 4.

TABLE 4. Estimated amounts of lead in the atmosphere from natural sources /After PATTERSON, 1965/.

Source	Amount $\mu g\ Pb/m^3$
Silicate dust	5×10^{-4}
Volcanic halogen aerosols	3×10^{-5}
Volcanic silicate smoke	6×10^{-6}
Forest fire smoke	6×10^{-6}
Aerosolic sea salts	1×10^{-6}
Meteoritic and meteoric smoke	2×10^{-9}
All sources	5.43×10^{-4}

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The lead concentrations presently existing in the atmospheric air are considerably higher than the estimated natural level of 5.43×10^{-4} µg Pb/m^3. The amount of lead in air depends on the locality, and is lower in rural than in urban regions. The mean concentrations of lead found during the two last decades in different localities are presented in TABLE 5. It will be seen that the mean existing levels of lead in the air are 3 to 4 orders of magnitude higher than the estimated natural levels.

TABLE 5. The existing lead content of the air.

Location	Mean concentration µg/m^3	Source of information
Cincinnati, Ohio	1.2	/KEHOE, 1961 A/
Los Angeles	5.2	/KEHOE, 1961 A/
San Bernardino /Calif/	1.5	/KEHOE, 1961 A/
WASHINGTON, DC	2.5	/KEHOE, 1961 A/
Philadelphia	9.5	/ANONYMOUS, 1959/
Minneapolis	0.05	/ANONYMOUS, 1959/
nonurban area	0.1	/ANONYMOUS, 1959/
Zurich 1949/50	1.4	/ANONYMOUS, 1961/
Zurich 1960/61	4.5	/ANONYMOUS, 1961/
Small town in Germany	0.12	/ANONYMOUS, 1961/
Countryside near Warsaw	0.09	/JAWOROWSKI, 1963 C/
Average urban	6.0	
" nonurban	0.1	

The principal sources of industrial lead in urban air are lead alkyls, lead paint dust and combustion of coal. According to PATTERSON /1965/ in the urban atmosphere of United States the contribution of lead alkyls to the average lead content in ground level air is some 0.6 µg Pb/m^3, and the contribution from lead paint dust is 0.001 µg Pb/m^3. PATTERSON /1965/ estimated the coal fly ash contribution as 0.001 µg Pb/m^3 in the ground level air of urban areas in the United States. This value was calculated accepting that 0.067 percent of the coal mass leaves the boilers for the atmosphere as aerosols, less than a few microns in diameter, and that the Pb concentration in silicate material of ash is 0.01 per cent, which corresponds to 0.002 per cent Pb in coal. According to PATTERSON the contribution from coarse fly ash is about four times higher. These values calculated for coal contribution seem to be too low as the fraction of burned coal mass transferred to the atmosphere is rather 2.0 percent /ZIENKIEWICZ, 1967/ and not 0.27 per cent as accepted by PATTERSON for coarse fly ash, and in addition the average concentration of lead in coal is higher than 0.002 per cent.

This last value ranges from 0.001 to 0.40 per cent in the British coals /MONKHOUSE, 1950; REYNOLDS, 1948/, and was found to be 0.015 per cent in the Polish coals /JAWOROWSKI, 1967/. The average world concentration probably may be accepted as about 0.02 per cent. From these values an average coal contribution may be inferred as about 0.4 µg Pb/m^3 of ground level air in industrial

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urban areas. During the coal burning, a part of its lead will be volatilised and pass to the atmosphere directly. By the ashing of organic matter about one third of the lead content may be lost by volatilising even at relatively low temperatures /GORSUCH, 1959/. The present author found that in the ash from a great power station the concentration of lead was only about 1/10 of that in coal burned in the boilers /JAWOROWSKI, 1967/. The contribution from this volatilised lead was not, however, included in the calculation.

The above estimations are subject to doubt as information on lead concentration in coal and its fraction passing to the atmosphere is scarce as well as information on the fate of combusted lead alkyls. It may however be concluded that the contribution from coal and lead alkyls to the atmospheric air are now probably of the same order of magnitude. In the past, the major contribution was certainly from coal burning. In countries with moderate motor vehicle traffic the relative contributions from coal and lead alkyls will differ from those in countries with heavy traffic. In Poland, the amounts of lead which were, in 1962, introduced to the atmosphere from coal and the combustion of lead alkyls were respectively 1.0×10^9 g and 5.0×10^7 g, which contrasts with the respective values for the United States of 3.7×10^9 g [x/] and 8.0×10^{10} g

x/ Calculated assuming that 1.0×10^{11} kg of coal and 2.5×10^8 kg of petrol is consumed yearly in Poland and $3,7 \times 10^{11}$ kg of coal in United States /ANONYMOUS, 1963 B/.

/ZIEGFELD, 1964/. The lead a
culated on the assumption th
charged in the exhaust gas /
considerable part of the bur
is deposited immediately on
diminishing the amount of le
During the past two decades
rapidly increased but in the
introduced into the atmosphe
of dust collecting devices.
centration in the urban atmo
change much between the year
contrary to speculative pred
crease in the number of moto
hensive information on the v
in various parts of a given
the land masses and seas, an
scale. The majority of inves
lieved to be of immediate hy
cerned themselves with the g
makes an estimation of the e
societes to chronic intoxica

Surface water

The natural lead content
lated by PATTERSON /1965/ as

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1.7×10^{10} g Pb/year entered the oceans during the last few hundred thousands years /CHOW et al., 1962/, and that the total discharge of rivers to the oceans is 3.7×10^{16} liters/year /POLDERVAART, 1955/. This estimated value of natural lead concentration is much lower than the values determined in existing surface waters. The average concentration of lead found recently in the dissolved solids of North American rivers, draining inhabited areas was 54 ppm, while in those draining uninhabited areas 43 ppm /DURUM et al., 1960/. So, accepting the average concentration of lead as about 50 ppm part of solids and an average salinity of 150 ppm /POLDERVAART, 1955/, the concentration of Pb in existing river waters can be inferred as 8.0 µg Pb/liter. In other studies the concentrations of Pb in surface waters were found to range from 0 to 55 µg per liter /TABLE 6/. The contemporary surface waters are contaminated by lead falling out from the air, and also by lead from insecticides and other chemicals used in agriculture.

TABLE 6. Existing lead content in surface waters.

Source of information	Pb concentration µg Pb/liter
/KEHOE et al., 1933/	4 - 20 /measured/
/DURUM et al., 1961/	0 - 55 /measured/
/PATTERSON, 1965/	8 /calculated/

It should be pointed out that surface waters are often lower in lead content than ordinary types of distilled water, which may become contaminated in the distilling equipment /DE TREVILLE, 1964/. In sea water, the lead content was found ranging from 0.002 to 0.20 µg per liter /TATSUMOTO et al., 1963/.

Drinking water

In drinking water tle lead concentration is usually higher than in surface one. As will be seen in TABLE 7 the lead content in drinking water from municipal supply systems may vary from 1 to 500 µg/liter.

TABLE 7. Lead content in drinking water

Source of information	Pb concentration µg Pb/liter
/KEHOE et al., 1944/	40 /av. 11/
/INGLESON 1938 quoted by MONIER - WILIAMS 1938/	5 - 500 /av. 118/.
/BACCHI et al., 1940/	2
/SCHROEDER et al., 1961/	0 - 112.5
/ANONYMOUS, 1961/	1 - 95

The data presented by SCHROEDER et al. /1961/ indicate that the sources of lead contamination of municipal drinking water are

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the materials used for piping and filtration. The standard for lead in U.S. drinking water is 50 µg/l /ANONYMOUS, 1962/. According to KEHOE /1944/ the mean lead concentration in drinking water, collected in 37 places in 16 states is 11 µg/l. Lead concentration may be higher in water from the first flow than in that collected after ten or more minutes of running.

Rainwater

The lead content in rainwater is obviously dependent on its concentration in the atmospheric air. In recent years the lead content in rainwater and snow ranged from 0 to 1858 µg per liter /TABLE 8/.

TABLE 8. The existing lead content in rainwater and snow

Source of information	Lead concentration / µg Pb/liter /
SCHROEDER et al.,/1961/	0 - 1858
TATSUMOTO et al.,/1963/	.2 /only soluble in 0.01 N HCL/
THIS WORK /Table 40 and 41 /	2 - 215 x/

x/ During the years 1950-1965

Soil

The average lead content of alcaline and acid rocks is about 8 µg Pb/g and 20 µg0B/g respectively. The average natural lead

content of soils should be close to this value. However some sedimentary and aluvial soils are reported to have lower concentrations of lead i.e. 0.04 to 0.06 µg Pb/g /BAGGHI et al.,1940/, while some agricultural soils may well surpass the level of 1000 µg Pb/g. /TABLE 9/.

TABLE 9. Lead content of soils / µg Pb/g /

Type of soil	Lead Concentration	Source of information
Alluvial soils	0.04 - 0.06	/BAGGHI et al. 1940/
Virgin soils	0.07 - 108	/KEHOE et al. 1933, WRIGHT et al. 1955, VINOGRADOV 1957/
Arable soils	0.5 - 1000	/PRINCE 1957, WARREN et al. 1960/
Soils in industrial areas	534 - 40.175	/PAKHOTINA 1958, MANLEY 1937/
Urban soils	16.4 - 360	/CHOLAK et al. 1961/
10 m apart the highway	9.0 - 13.6	THIS WORK

Food

Lead may enter food from soil or from the air and rainwater. Appreciable amounts of lead are often incorporated into food from the production tools, packing as well as during preparation prior

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to eating. The lead levels are generally lower in natural vegetable products than in prepared food /DE TREVILLE, 1964/. The concentrations of lead in more than a hundred kinds of food have been determined by SCHRÖDER et al. /1961/. The highest mean concentration was found in sea food. These concentrations are in substantial agreement with the levels found by other authors /KEHOE et al. 1940, BACCHI et al. 1940, WARREN et al. 1962., and LEROLD 1946/.

TABLE 10. Average lead content in food

Food	Lead concentration /µg Pb/g wet weight/	Source of information
Sea fish, shellfish and crustacea	0.49	SCHRÖDER et al. 1961
Nuts	0.21	IBID.
Grains and grain products	0.37	IBID.
Vegetables	0.21	IBID.
Milk evaporated and dried	0.22	IBID.
Milk fresh	0.0	IBID.
Wine	1.5	THIS WORK
Tea, hand picked	0.02/L	KEHOE, 1940
Tea, in corrored lead foil	43.2/L	IBID.
Tea, packed in tins	1.8/L	BACCHI, 1940
Tea, packed in lead foil	2.4	IBID.

The alimentary intake of lead can be inferred from the food and excretion data of CHOLAK et al. /1943/ and KEHOE /1947/ as about 0.30 mg per day.

MONIER-WILLIAMS /1950/ in his monograph on trace elements in food stated that "lead may pass into the milk of cows suffering from lead poisoning". According to WARREN /1962/ and MONIER-WILLIAMS /1950/ the mean lead concentration in food consumed in civilised areas is about 2 µg Pb/g wet weight. Comparing the natural abundances of lead and calcium, PATTERSON /1965/ estimated that the natural concentration of lead in food should average about 0.01 µg Pb/g wet weight, and he concluded that most of the lead now present in food originated from industrial sources. The amount of lead present in the total amount of food consumed yearly in the United States is only 1/50.000 fraction of the yearly lead production of that country. It seems that this small fraction may be considered as a possible major contributant to ingested lead, mainly by way of folial contamination of food crops, by the fallout of atmospheric lead, by etching of lead from solder in canned food, by use of lead arsenate insecticides, by use of lead in food processing equipment, and by chemicals used to prepare foods and food acids and salts.

It seems that since antiquity man has contaminated his food with lead by preparation, processing and storage, this being done in man's immediate environment i.e. at home, yard or workshop. However it is only recently that he has started polluting with lead the total biosphere of the Earth.

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5.2 THE ORIGIN AND LEVELS OF RADIOACTIVE LEAD IN THE ENVIRONMENT

Of the five natural radioactive leads, only lead-210 /Radium D/ contributes an important fraction to the natural radiation dose received by man, although its concentrations in ambient air are some three orders of magnitude lower than those of lead-214 /LOCKHART jr., 1964/. The concentration of lead-210 in living organisms however exceeds those of any other natural radiolead. Therefore in this work only lead-210 will be considered in detail.

Lead-210 is a member of the uranium family, and it occurs in nature in a partial radioactive equilibrium with its immediate daughters bismuth-210 /radium E/ and plonium-210 /Radium F/. The complete radioactive equilibrium of lead-210-bismuth-210-polonium--210 system in the biosphere is usually not reached due to the differences in the influence of biological, meteorological, chemical and other factors on each of these three nuclides /BURTON et al. 1960, HOLTZMAN 1960 A. JAWOROWSKI 1965/.

Practically all lead-210 originates from polonium-214 /Radium C'/ and only 0.04 per cent from thalium-210 /Radium C''/ /NESMEYANOV et al., 1956/. These two parents of lead-210 belong to a group of short-living radionuclides originating from radon-222, the half-lives of which ranged from 1.64×10^{-4} seconds to 26.8 minutes. As may be seen from TABLE 2. radon-222 originates from long living radium-226 the ultimate source of which is uranium--238 having a half-life of 4.5 miliards of years.

Atmosphere

The source of natural le
a noble gas emanated from the
atoms of radon can move from
reaching the atmospheric air.
on the behaviour of radon in
gration of radon in the soil
the radioactive formation and
its parent radium-226. 2. the
the interior of a mineral grai
of the neutral atom through p

When an atom of radium-22
particle and a radon atom of
atom of about 1×10^{5} electron
about 3×10^{-6} cm long, in mi
1943/. The radon-222 atom may
or escape from the mineral gra
atom in the air is some 9 x 1
cording to ZIMENS an isolated
uniform distribution of radium
tually all the recoiling ions
micron size. If such micron si
usually are in geologic situat
the grains will become buried

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the fraction terminating their recoil path in the pores is negligible.

However in a rock with the pores filled with water the situation will be different, as the recoil range in water is some two orders of magnitude less than in the air. In effect, in moist rock the recoil ions entering pores are likely to end their tracks in the pores filled with water, where diffusion may proceed at a rate many orders of magnitude greater than in solids. The moisture in the rock should therefore increase the emanation of radon as was often observed /KIRIKOV et al. 1932, HAHN 1936, STARIK et al. 1957, PENSKO et al. 1967/.

It seems that the recoil of the atoms of radon-222 is the only important factor determinating the rate of migration of radon from the mineral to the pores. The escape of radon from minerals by diffusion is negligible as may be inferred from the studies on the similar problem of leakage of argon from minerals /AMIRKHANOFF et al. 1961/ and lead from zirkon and monazite /NICOLAYSEN, 1957/, and from the studies of KOSOV et al. /1955/ on emanation powers of radon-222 and radon-219 in minerals. It may be supposed then that in intact crystals radon emanates from shallow regions, determined by the recoil range. On the other hand the development of an internal surface of the crystals such as may result from chemical corrosion, weathering or fracturing on a microscopic scale may largely increase the escaping capability of radon.

The diffusion and convective processes operating in the shal-

low layers of the soil influence the surface flux of radon. This flux depends on rainfall /WRIGHT et al., 1915/, freezing and snow cover /BENDER 1934, ZUPANCIC 1934/, atmospheric pressure /KOGOSLOVSKAYA et al. 1932, HATUDA 1953, SHEVCHENKO 1958, KRANER et al. 1964/, wind and termal atmospheric instability /KRAMER et al., 1964, MOSES et al., 1960/.

The diffusion depth of radon-222 in undisturbed rock and soil may be calculated as 160 cm, accepting the diffusion coefficient as 5×10^{-2} cm^2/sec., i.e. that for dry sand. With other values for diffusion coefficient and transport velocity, suitable for different types of soil, the mean migration distances of radon-222 are 1 to 200 cm /TANNER, 1964/.

The contribution from larger rock crevices or water sources to the global exhalation rate of radon-222 is negligible /JACOBI, 1962/.

It is generally assumed that the rate of radon-222 exhalation at the interface between ground and atmosphere is predominantly influenced by the diffusion mechanism, the transport mechanism being of minor importance. This assumption, however is not justified by observations which rather indicate that under given conditions transport phenomena may also play an important role /TANNER, 1964/ particularly all air motions may exert a "turbulent pumping" of the soil gas /KRANER et al., 1964/.

The exhalation rates of radon-222 were measured over land in different part of the world. The results of these measurements are compiled in TABLE 11.

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TABLE 11. Measured exhalation rates of radon-222 from the ground

Locality	Exhalation rate /10^{-17} Ci/cm^2.sek/	Source of information
Dublin /Ireland/	7.4	SMYTH /1912/
Manila /Philippine Is./	2.1	WRIGHT et al./1915/
Liebenau-Graz /Austria/	4.0	KOSMATH /1934/
Innsbruck /Austria/	2.3	ZUPANIC /1934/
Innsbruck /Austria/	5.0	ZEILINGER /1935/
Warsaw /Poland/	1.9	Perisko et al. /1967/
Kaluga /USSR/	0.6 - 2.9	SYSYGINA /1965/
Moscow /USSR/	1.9	ibid.
Kursk /USSR/	1.0 - 4.2	ibid.
Yaroslav /USSR/	0.2 - 2.3	ibid.
Tula /USSR/	2.7 - 4.0	ibid.
Orlov /USSR/	2.3	ibid.
Belgorodsk /USSR/	0.7 - 2.0	ibid.
Dnepropetrovsk /USSR/	0.5 - 2.4	ibid.
Zaporodhe /USSR/	1.6 - 2.3	ibid.
Azov Sea Coast /USSR/	0.4	ibid.
Muyun-Kum /Kazachstan SSR/	0.8 - 2.3	ibid.
Socorro /N.Mexico US/	9.0	WILKENING et al./1960/
Rainier Mesa /Nevada USA/	3.4 - 9.5	KRAMER et al. /1964/
Yucca Flat /Nevada USA/	3.2 - 6.5	ibid.
Lincoln /Mass. USA/	10 - 13	ibid.
Mean	4.0	

In TABLE 11 are compiled prob
halation rate published up to date
tative for the whole land surface
measurements of exhalation rate ov
of soil and rock types and also du
distribution of sampling stations.

This gives rise to some reser
mean global exhalation rate over t
may be inferred as about 4 x 10^{-17}

The radon-222 exhalation rate
over the land, due to the low conc
-water. JACOBI /1962/ calculated t
i.e. about 100 times lower than th
over land. The difference between
and sea is similar to the differen
air concentrations given by MAUCHL
suggests that in calculating the l
atmosphere, the contribution of th
surface of the seas can be neglect

Of the total land surface of
covered by glaciers or permafrost
are unlikely to exhale substantial
DER et al.,1961; KOVACH, 1944/, as
/NUTT, 1959 and 1961/ and cannot e
be accepted that the radon-222 pre

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duced by the remaining 112 x 10^6 km^2 of the land surface and its global production rate, calculated from the mean radon exhalation rate of 4 x 10^{-17} Ci/cm^2.sec. is about 3.5 MCi per day. This corresponds to the production rate of 0.62 MCi of lead-210 per annum.

If the residence time of lead-210 in the atmosphere was influenced only by the rate of its radioactive decay / T 1/2 - 20 years / then the total amount of lead-210 in the atmosphere would be about 20 MCi. The mean residence time of lead-210 in the atmosphere however is, due to meteorological factors, shorter than the radioactive half-life of this nuclide, and was calculated by BURTON et al. /1960/ from polonium-210 to lead-210 ratio, as 29 days. Using this last value the total amount of lead-210 in the atmosphere may be calculated as about 50 kCi.

According to BURTON et al. /1960/, the concentration of lead--210 in the atmosphere changes with altitude, from low values at ground level to the highest values over the tropopause . The vertical concentration gradient of lead-210 was measured by BURTON et al. above the United Kingdom during the years 1954 and 1957. These authors found that the lead-210 concentrations in ground level and stratospheric air differ by a factor of ten, suggesting the existence of a "stratospheric magazine" of this nuclide. This finding is in substantial agreement with the vertical distribution of lead--210 and polonium-210 in the atmosphere calculated teoretically by JACOBI /1962/. However the results of the extensive measurements of FEELY et al. /1965/ of the atmospheric radioactivity at altitudes between 5.6 and 20 km, are in disagreement with the above findings, indicating that both in continental and oceanic air, the concentrations of lead-210 and polonium-210 are more or less uniformly distributed at different altitudes, with no marked preference of high concentrations found in the stratospheric air.

TABLE 12. Natural production of radon-222 and lead-210 in the atmosphere

Mean exhalation rate of radon-222 over the land	4 x 10^{-17} Ci/cm^2.sec.
Mean exhalation rate of radon-222 over the sea /after JACOBI, 1962/	5 x 10^{-19} Ci/cm^2.sec.
Global production rate of radon-222	3.5 MCi per day
Global production rate of lead-210	0.62 MCi per year
Total content of lead-210 in the atmosphere calculated without considering meteorological factors / T1/2 = 22 years/	20 MCi
Total content of lead-210 in the atmosphere calculated for mean residence time τ of 29 days / T1/2 = 20 days /	50 kCi

The measurements of RAMA et al. /1961/ and PEIRSON et al. /1966/ also indicate that the vertical gradient of the air con-

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centration of lead-210 is rather small if any.

The air concentrations of lead-210 at ground level have been measured in different parts of the world since 1952 /CLIFFORD et al., 1952/. The mean concentration at ground level in the northern middle latitudes, as measured by several authors, was found to be 0.014 pCi/m^3, and was higher than those in other geographical regions /see par.7.2/.

Surface waters

Lead-210 is washed out into the surface waters from the atmosphere and from the soil. The soil on the other hand, can absorb lead-210 /SCHULZ, 1965/ from passing rain water. The concentrations of lead-210 in surface waters are lower by a factor of ten to a hundered than those usually found in rain water. This indicates that the amounts of lead-210 absorbed by the soil are higher than those which can be leached out.

HOLTZMAN /1964 A/ has determined the lead-210 content in surfacewater supplies in Illinois. Some of his samples were not treated by either filtration or settling and therefore may represent natural surface waters. The lead-210 concentrations found in these samples are compiled together with the data of RAMA et al. /1961/ in TABLE 13.

As may be seen from TABLE 13, the lead-210 concentrations in fresh surface water vary from 0.085 to 6.7 pCi/l, being about 2 - 100 times higher than those in ocean waters.

TABLE 13. Lead-210 content of

Type of water	Lead
Ocean surface	0.04
Colorado River, U.S.	0.1
Water supplies, Ill. US	
Pana	
Eureka	
Effingham	
Newton	

Drinking waters

The water consumed in dif
purified snow, rain and surfac
clean untreated surface water
ated surface water and well wa
found in different types of po

As may be seen from TABLE
ferent municipal water supplie
Some types of water /untreated
table may contain lead-210 in
in rainwater.

The lead-210 content in w
in that from shallow ones and

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The lead-210 concentration does not seem to be related to the mineral content of water as represented by sulfate, calcium and fluoride, neither does it seem to be related to the radium-226 content /HOLTZMAN, 1964/.

TABLE 14. Lead-210 content of potable waters

Type of water	Lead-210 / pCi/l /	Source of information
Tap water, La Jolla, Calif.,US	0.054	RAMA et al. 1961
Treated surface water, Illinois, US /average/	0.019	HOLTZMAN, 1964
Untreated surface water, Illinois, US /average/	0.127	IBID.
Untreated well water from 0-98.5 m depth /average/	0.083	IBID.
Untreated well water from 98.5-590 m depth /average/	0.028	IBID.
Treated surface water, Warsaw, Poland	0.021	THIS WORK
Untreated well water, Świerk, Poland	0.011	IBID.
Mineral water, Lądek, Poland	0.9-2.0	IBID.
Mineral water, Plombieres-les- Bains, France	1.0-7	KAYSER et al.1964

According to HOLTZMAN /1964/ 40 - 90 per cent of lead-210 may be removed from surface water by filtration or setling.

Rain water

The source of lead-210 in rain water is obviously lead-210 contained in the tropospheric air. The contributions from natural and artificial sources to rain water concentration and the time and geographical variations of this concentration will be discussed in detail in par. 7.2.

It will be seen that rain water concentrations of lead-210 are highly variable and in the same locality may differ by two orders of magnitude. These concentrations are not related to either season or amount of precipitation /BARANOV et al., 1965/.

Food

The data on the lead-210 content of food are rather scant. Some information may be derived from the studies on the organ distribution of this nuclide performed on domestic animals. Recently numerous determinations of lead-210 content have been performed on reindeer, cariboo and plant samples collected in the Arctic. Evaluations of the daily intake of lead-210 with food has been made by HOLTZMAN /1960 A/ on the basis of fecal and urine analysis in two persons. He assumed that the intake is of the order of 1.8 pc per day. From the measured content of lead-210 in a few components of the diet of United Kingdom residents, the alimentary intake of lead-210 was estimated as 1 - 10 pc per day /HILL, 1965/. In the other geographical regions where residents depend mainly on local food, this intake may differ considerably,

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reaching a mean value of 100 pCi per day in the Arctic /HILL,1966/. Time variation may also be anticipated on the basis of the study on the temporal distribution of lead-210 in the biosphere /JAWOROWSKI, 1966/. However, there is a lack of extensive studies on the lead--210 content of food, which could enable a realistic assessment of the intake of this nuclide by different population groups.

The lead-210 to polonium-210 ratio in human and animal food is an important factor in the study of the lead-210 food-chain. Information on the values of this ratio at particular levels of the food-chain may be useful for estimating the radiation dose received by humans and animals from the lead-210-bismuth-210-polonium-210 system, and also for elucidating some patterns of intake and metabolism of these nuclides.

In TABLE 15 are compiled the results of determinations of lead-210 and polonium-210 in some human and animal food.

TABLE 15. Lead-210 and polonium-210 in human and animal food

Food	Lead-210 /pCi/kg/	polonium-210 /pCi/kg/	Source of information
1	2	3	4
Sea food			
cockles, Europe		500- 1300	HILL, 1962 A
crab, Europe		1300; 1400	HILL, 1965
fish /meat/,Arctic	0.27-1.5		HOLTZMAN, 1966 A
seal /meat/,Arctic	2.0- 5.1		IBID.

TABLE 15. Cont.

1	2	3	4
plankton, Pacific		2000	HILL, 1965
Grains and grain products			
Australian wheat	30		HILL, 1962 A
breads and cereals, U.K.		1-7	
English white bread	1.5		IBID.
Vegetables			
green, U.K.		6-90 /1-3/	HILL, 1965
spinach, U.K.	90		HILL, 1962
carrots and potatoes, U.K.		1	IBID.
broccoli, U.K.	60		HILL, 1962
Dried milk, U.K.		2-6	HILL, 1965
Meat			
beef and lumb,U.K.		3	IBID.
reindeer, Lapland		15-50	IBID.
reindeer, Canada		200	IBID.
reindeer, Lapland	3	130	KAURANEN et al.1966
elk, S. Finland	1	20	IBID.
reindeer, Lapland	1.3-15	575	HOLTZMAN, 1966 A
caribou, Alaska	1.1-16.1	72-360	IBID.
caribou, Alaska		34-195	BLANCHARD, 1966
ptarmigan, Alaska	3.3-27.3		HOLTZMAN, 1966 A

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TABLE 15. Cont.

1	2	3	4
Liver			
beef and lamb,U.K.		4-100	HILL, 1965
ox ,U.K.	10		HILL, 1962
reindeer, Lapland		350-750	HILL, 1965
reindeer, Canada		2400-5600	IBID.
reindeer, Lapland	640	3300	KAURANEN et al.1966
elk S. Finland	26	230	IBID.
Kidney			
beef and lamb,U.K.		48-270	HILL, 1965
lamb, N.Wales		90-1800	IBID.
reindeer, Lapland		110-490	IBID.
reindeer, Canada		2300;4290	IBID.
reindeer, Lapland	360	1520	KAURANEN et al.1966
elk,S.Finland	41	120	IBID.
Bone			
cattle, U.S.		22-670	DI FERRANTE, 1961
cattle, U.K.	240;260		HILL et al. 1961
cattle, Poland	135-1500	140-280	JAWOROWSKI, 1967
lamb, Iceland	107-141		HOLTZMAN, 1966 B.
reindeer, Lapland		520-3500	HILL, 1965
reindeer, Lapland	2000	2530	HOLTZMAN, 1966 B.
reindeer, Lapland	3700	2110	KAURANEN et al.1966
elk, S.Finland	480	280	IBID.
deer, Scotland		1300	HILL, 1965

TABLE 15. Cont.

1	2	3	4
caribou, Alaska	2440-4430	1100-6200	HOLTZMAN, 1966 B.
caribou, Alaska		1110-7380	BLANCHARD, 1966
camel, Jordan		1100;1500	HILL, 1965
Animal food			
grass /dried/.U.K.		400-16000	IBID.
grass, Illinois	128-717	53-939	HOLTZMAN, 1966 B.
grass, Ohio	180	620	BLANCHARD, 1966
horsetail, Finland		300	KAURANEN et al.1966
clover, S.Finland	830	230	IBID.
willow, leaves S.Finland	900	210	IBID.
birch, leaves S.Finland	1140	320	IBID.
lingonberry, S.Finland	310	230	IBID.
lichen, S.Finland	6950	5550	IBID.
lichen, Lapland	6400	5900	IBID.
lichen, U.K.		7800 ,10000	HILL, 1965
lichen, Canada		3500	IBID.
lichens, Alaska	11500--69600		HOLTZMAN, 1966 B.
lichens, New Hampshire	7610; 6950	7320;7500	IBID.

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Strinking features of the data in TABLE 15 are the geographical differences in lead-210 /or polonium-210/ content in food /see par.7.2/ as well as differences at different levels of the food--chain. It may be seen from this table that the lower the position in the food-chain the higher the concentration of lead-210.

Cigarette smoking may be a source of lead-210 and polonium--210 in the body. It was found that the concentration of polonium--210 in cigarettes ranged from 0.39 to 0.48 pCi per cigarette /RADFORT Jr. et al. 1964/ and concentrations of lead-210 from 210 to 1360 pCi per kilogram /HILL, 1965/. HOLTZMAN et al. /1966/ found in cigarette smoke 0.006 to 0.018 pCi of lead-210 per cigarette and 0,016 to 0.027 pCi of polonium-210 per cigarette. These last authors estimated that in a one-pack-per-day smoker the contribution of cigarettes to the total body burden of polonium-210 is probably less than 2 per cent. From the data of these authors it can be inferred that in case of lead-210 this contribution is more than 50 per cent.

6. STABLE AND RADIOACTIVE LEAD IN THE ORGANISM

6.1 STABLE LEAD IN THE BODY

For metabolic and toxicological studies on lead it is essentie to know the "existing" levels of lead in the body. These levels should be distinguished from the "natural" levels, e.i. those whicl would be observed in the body if it were not subject to man-made lead contamination. The natural levels are rather difficult to determine or estimate. The determination of these levels might be made in residents of primitive areas, remote from industrial centers and civilized regions, who depend predominantly on the local supply of food and utensils. Probably some Eskimo and Lapp populations would be suitable subjects for such a study. There is however only little information available as to the stable lead contents in these groups of people /KEHOE et al., 1933/.

An estimation of the natural levels of lead in the environment and human body has been made by PATTERSON /1965/ on the basis of a comparison of the relative abundances /in calcium/ of elements in the Earth's crust and in the components of biosphere. This author estimates that the natural daily intake of stable lead in food, water and air is about 20 µg per day, and the natural total body burden of lead about 2 mg per 70 kg body weight, if the biological half-life of lead in total body is 1460 days. Applying a more realistic value of this half-life of 2085 day /HOLTZMAN, 1960 A/,

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this burden may be estimated as about 5 mg.

The existing lead burdens in the total body can be assessed from the measured concentrations of lead in the skeleton and soft tissues. The values of these burdens as calculated by different authors are compiled in TABLE 16.

TABLE 16. The existing stable lead content in the total human body

Population group	Lead content /mg per 70 kg/	Source of information
United States	66	ICRP 1960
United States	111	HOLTZMAN 1960 A
United States	220	KRAUZE 1961
United States	100 - 400	KEHOE 1961 A
United States	125	SULTZER et al. 1953
United Kingdom	750	MONIER-WILLIAMS 1950
United Kingdom	190 X/	JAWOROWSKI 1963 A
Poland	86 X/	JAWOROWSKI /This work/
World mean	240 xx/	JAWOROWSKI /This work/

X/ Calculated from the mean lead concentration in vertebra and accepting the ratio of cortical/trabecular lead concentrations as 3.27 /TABLE 19/, and the fraction of total body lead in the skeleton as 0.7 /ICRP 1960/.

xx/ Calculated from the mean cortical concentrations in TABLE 19.

These measured levels o
mans living in the highly ci
higher than those theoretical
subject.

Tissue distribution of stable

The distribution of lead
been determined by several au
Report. An extract from this

As may be seen from TAB
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being in the soft tissues, wh
in the skeleton. Similar rela
and in the soft tissues were
of specimens collected in Pol
composed of poolled equal amo
thyroid, heart, duodenum, sto
and testis from two residents
was found to be 0.72 µg/g wet
centration of lead in the mus
poolled samples and that the
of these two subjects was the
subjects from the same region
body of residents in the Wars
62 per cent of it being in th
different from that tabulated

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TABLE 17. The existing average stable lead contents in various tissues of the human body /After ICRP 1960/.

Tissue	Lead concentration /µg/g wet/	Lead content /mg/	Fraction of total body lead content /per cent/
Bone	6.6	46	69.5
Muscle	0.17	5.1	7.7
Skin and subcutaneous tissue	0.55	3.4	5.1
Liver	2.0	3.4	5.1
Fat	/0.2/[x]	/2.0/	/3.0/
Blood	0.3	1.6	2.4
Lungs	0.68	0.7	1.1
G.I. tract	0.3	0.6	0.9
Bone marrow	/0.2/	/0.6/	/0.9/
Brain	/0.4/	/0.6/	/0.9/
Kidneys	1.24	0.4	0.6
Lymphoid tissue	/0.2/	/0.1/	/0.2/
Heart	0.2	0.1	0.2
Spleen	0.67	0.1	0.2
Other	0.7	1.4	2.1
Total body		66.1	100.0
Soft tissues	0.32	20.1	29.5
Skeleton	6.6	46.0	70.5

[x] Values in parentheses are maximum limits.

The data of HOFREUTER et al. /1961/ indicate that differences exist in the body lead content in relation to sex, smoking history and residential area of the subjects /TABLE 18/. It may be seen, that there are highly significant differences between the average blood lead concentrations in males and females from both urban and rural areas, and between smokers and nonsmokers. In both rural and urban populations, no notable tendency toward increase or decrease of the blood lead level with age was observed in 3 arbitrary age groups.

TABLE 18. Influence of sex, age and smoking history on blood lead concentration. /After Hofreuter et al. 1961/

	Urban areas		Rural areas	
group	No.of obs.	mean blood lead /mg/100 g/	No.of obs.	mean blood lead /mg/100 g/
Male	566	0.021	113	0.016
Female	169	0.016	49	0.010
15-30 yr.	297	0.019	50	0.015
30-45 yr.	288	0.020	73	0.014
45-60 yr.	150	0.021	39	0.014
Smokers	471	0.021	89	0.017
Nonsmokers	264	0.017	73	0.011

However, in the skeleton, lung, kidney, pancreas, liver and aorta a tendency towards increase of lead levels with age has been

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observed by SCHROEDER et al. /1961/. The maximum concentrations of lead in these tissues were reached between the fourth and sixth decade and the mean concentrations at birth or in childhood were four to ten times lower. A similar tendency in bone tissues was found by KRAUZE /1961/.

Lead is rather strongly bound with both the soft tissues and bone. The soft tissue lead is probably chelated to proteins, most likely to the sulfhydryl groups /SCHROEDER et al. 1961/. In these tissues extracellular lead may be rapidly removed by the action of EDTA but the strongly complexed lead inside the cells can be removed only very slowly /CASTELLINO et al. 1965/. In bone, neither a low nor high calcium diet, nor parathyroid hormones /CALHOUN et al. 1954/ nor EDTA /CASTELLINO et al. 1965/ can modify the fixation of lead.

In 1926, AUB et al. found experimentally that lead is deposited in the skeleton mainly in its mineral part. McDONALD et al. /1951/ found that lead exchanges with calcium atoms in the hydroxy apatite crystals replacing them in the crystal lattice. GITELMAN et al. /1959/ however, suppose that the lead deposition in hydroxy apatite crystals is not a simple ion exchange process as the structure of the hydroxy apatite molecule changes when lead atoms enter the crystal lattice. The presence of lead inside the hydroxy apatite crystals was confirmed by the studies of CHEN et al./1961/.

In growing hair, lead is probably bound with the sulfhydryl groups of the keratin precoursors in the hair matrix and then is

incorporated to the non metabolizing keratin of the hair stem /SCHWARTZ, 1960/ /see par.6.2/.

Stable lead in human bones

The skeleton concentrations are most representative for the long-time accumulation of lead in the body. GUSSEROW /1861/ was the first to observe that lead is stored in the skeleton. The "normal" concentrations of lead found in bones by different authors during the last three decades are given in TABLE 19. It is seen fro this TABLE that the distribution of lead in the skeleton is non-homogeneous. In the majority of studies it was found that lead concentrations were lower in trabecular bone, usually represented by the rib or vertebra, than in cortical bone represented by the tibia femur or humerus. In this measurements the lead concentrations increased along the chain: vertebra - rib - tibia - humerus - femur. In the study on lead distribution in the skeleton the same tendency was found by THOMPSETT /1936/. Different results however were presented by WEINIG et al. /1961/, who found in bone ash samples of 30 persons who had died in accidents or after a short illness, an invert distribution tendency of lead concentration and the ratio of cortical bone to trabecular bone concentrations of 0.77, which is in variance with the ratio of 3.27 calculated from the values in TABLE 19. A possible cause of this discrepancy is the different modes of expression of the results of the determinations. The data of WEINIG et al. /1961/ are given for ashed bones, whereas the

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data of THOMPSETT /1936/ and other values in TABLE 19 are expressed as lead contents in wet tissue. It will be seen from the discussion below, that the differences in the lead content of various bones represent methodical errors rather than any true difference in mineral composition.

The concentrations of elements in tissues are reported in literature with the use of different units, such as element/wet weight, element/dry weight, element/dry fat free weight, element/ /calcium weight and element/ash weight. This leads to difficulties when variously expressed results are to be compared. In bone studies, usually wet weight and ash weight units are used, and quite often the data are recalculated from one unit to the other. This is made with factors expressing ash to wet weight ratio in the skeleton or particular type of bone, usually derived from another set of samples by some other student. However, despite the wide use of such factors, largely differing in various publications, no systematic and comprehensive study on this ratio has yet been performed. It should be noted, that the values of this ratio vary depending on age, constitution, sex, disease, type of bone and analytical methods. Therefore the recalculation of values from one unit to another leads to doubtful results and when possible should rather be ommitted, which unfortunately, is not always possible.

For dosimetric reasons, it is necessary to express the concentrations of radionuclides in bone in wet weight units, corresponding as far as possible to in vivo conditions. It should be

emphacized, however, that the "wet bone weights" as used by different authors may largely differ from the actual wet bone weights in vivo, depending on the conditions in which the sample was preserved, the time of removal from the body and weighing. One may expect that large, rapid and irreversible losses in bone weight will occur, even with short exposure to air /HOLTZMAN, 1962/. However, as a rule, information regarding the time which elapsed between removal and weighing of the sample is not given in descriptions on preparing the samples. In effect, one never can be sure whether th "wet weight unit" used by a particular author is not a "partially dried unit".

In routine collecting of bone samples it is practically impossible to measure the wet weight of bone which would be actually that of in vivo weight, as losses in weight will occur even during the cleaning and scraping of the bone. There is also the problem of removal or non-removal of the bone marrow from the trabecular bones, the dry weight of which is small, whereas its wet weight is a considerable fraction of the bone weight. This makes the values of concentrations of elements in bone expressed in wet weight units, somewhat uncertain.

The use of ash weight units as proposed by HOLTZMAN /1962/ is also not an ideal solution to the problem as the ash content of different parts of the skeleton is highly variable. It seems that similar difficulties are encountered with the dry weight units as dry to wet weight ratios in different bones are not well known and

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probably are similarly varying and uncertain as the wet to ash weight ratios. There is a need therefore, of systematic studies on both wet to ash weight and wet to dry weight ratios in different components of the human skeleton.

TABLE 19. The existing mean concentrations of lead in bones of professionally unexposed humans /µg Pb/g wet bone/

Region	Type of bone				Source of information
	Femur	Tibia or humerus	Rib	Vertebra	
India	17.3	10	8.4	-	BAGCHI et al.1939
U.K. ?	59.9	44.3	-	-	LYNCH et al.1934
U.K.? U.S.?	50.0	45.2	9.9	8.3	THOMPSETT,1936
"	-	-	8.5	7.1	THOMPSETT et al. 1935
U.S.	20	-	6.9	-	KEHOE,1947
U.K.		4.5		6.6	JAWOROWSKI,1963
Poland	3.8[x]	-	-	2.3	JAWOROWSKI /this work/
Germany	11[xx]	11[xx]	6.7[xx]	7.8[xx]	WEINIG et al.1961
Mean	25.3	23.0	8.3	6.4	
Mean	cortical 24.2		trabecular 7.4		
Ratio: cortical/trabecular	3.27				

[x] One sample
[xx] Recalculated from ash-weight values /see text/.

WEINIG et al./1961/ compare spongiose and compact bone separ found in both parts of bones pr centrations /TABLE 20/. This is of TIMM /1936/ who found in "no distribution of lead in differe

TABLE 20. Distribution of lead /µg/g ash/. /After WE

Femur specimen	Compact
1.	2.14
2.	2.05
3.	1.09
4.	1.72

It may be supposed therefo lead concentration in different by variations in the soft tissu than by variations in the lead self. In preparation of cortica together with other soft tissue moved. This however is not the spongiosa like rib, vertebra of these samples most of the bone

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which substantially influences the results of analysis, as the lead concentration in the former is some two orders of magnitude lower than in the latter.

The turn-over rate of vertebra-like bone is higher than that of the femur-like one, the respective values being 8 and 1.5 per cent per annum /BRYANT et al. 1961/. As the majority of lead absorbed by bone is stored there for a longer time, one may expect that in the bone which has a higher turn-over rate the lead content will be greater than in the bone with a low turn-over. However, as may be seen from TABLE 19, just the opposite was found in respective types of bone, which supports the present authors opinion that a substantial methodical error is inherent in analyses of vertebra-like bone. It seems that for comparative studies on the skeleton content of elements, the best medium are the mid-shafts of long bones which can easily be cleaned from practically all interferring, non-osseous soft tissue. This suggestion is supported also by the fact, that the skeleton consist of seven-tenths femur-shaft-like bone and only three-tenths vertebrae-like bone /RIVERA, 1965/, the former being than, more representative for the whole skeleton than the latter.

It is interesting that the difference in lead content between the cortical and trabecular bones seen in TABLE 19 and expressed as cortical/trabecular ratio of 3.27, is similar to the difference in ash contents of both kinds of bone, which are respectively 21 and 60 per cent of wet weights /HOLTZMAN, 1960 B/. It is rather

improbable that this difference in ash content, reflects any real difference in mineral composition of the bone structure itself. It reflects rather the non-osseous components in the analysed samples. In Standard Man 7000 grams represent the mass of bone, free of non-osseous soft tissue. Therefore in calculating the skeleton content of lead in wet weight units, the concentrations in cortical bone should be used rather than those in trabecular bone. When trabecular bone data are used, recalculation for cortical bone values should be made using the factors representing the cortical/trabecular ratio of lead or ash content. Otherwise values, much too low, will be obtained.

Stable lead in the hair

High concentrations of lead in "naturally" contaminated people have been found in hair by the present author /JAWOROWSKI, 1964/. In this preliminary study on the relation between lead concentrations in hair and bone, it was found that the proportion between the mean concentrations in hair and bone was 1.63, the mean respective concentrations being 10.0 and 6.2 µg Pb/g tissue. In this study, however, the bone and hair samples were collected not from the same individuals. All of the bone samples were collected in the United Kingdom /London region/ whereas the majority of hair was sampled in Poland /Warsaw region/. In a further study on 21 subjects who had died in Warsaw /JAWOROWSKI, 1965 B/, the bones and hair were collected from the same individuals. As may be seen from TABLE 21. the

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mean lead concentration in vertebrae was 2.3 µg/g wet bone and in hair 8.9 µg/g. The hair to bone ratio was in this set of samples 3.8. It is interesting that the lead concentrations in vertebrae found in Poland are much lower than the concentrations found by other authors in Western countries and also lower than the concentrations found by the present writer in vertebrae from the United Kingdom. These discrepancies will be discussed in par.7.1. It will be seen in par.6.2 that lead is incorporated by growing hair and that its hair concentration depends on the level of the administered dose. The concentration of lead in hair reflects the blood levels of lead which existed during the period of hair growth, i.e. usually a period of several months. On the other hand the concentrations in bone in "naturally" exposed people reflect the blood levels of lead which existed over a period of many years. In adult persons living under strictly "normal" and steady environmental conditions one can expect that the amount of lead incorporated daily to the skeleton will be balanced by the amount leaving this organ in the same time. Actually there are brief periods /as compared with a lifetime/ during which an increase of lead content of the body may occur, /due to larger intake or impairment of excretion - LANGENBACH et al.1961/, and there are other periods in which there is a decrease of this content /due to lack of calcium in the diet or pathological conditions - LYNCH et al.1934/. These periods tend to alternate throughout life.

TABLE 21. Concentrations of lead in bone and hair of women who die in Warsaw in 1963

No. of person	Age /years/	Bone µg Pb/g wet	Hair µg Pb/g
1.	13	3.08 x/	20.71
1.	13	1.81	-
2.	23	3.10	12.73
3.	29	1.25	9.52
4.	33	1.27	6.30
5.	39	4.28	15.80
6.	43	3.72	12.40
7.	46	1.42	5.90
8.	50	2.79	4.85
9.	52	2.71	8.32
10.	53	1.70	7.09
11.	55	1.40	8.20
12.	59	3.33	13.02
13.	60	1.13	6.11
14.	61	3.39	15.10
15.	63	1.36	5.20
16.	65	4.70	18.31
17.	68	3.91	8.63
18.	68	1.51	7.10
19.	69	2.70	7.41
20.	80	0.57	5.11
21.	81	0.50	9.15
Average /in vertebrae/		2.32	8.90
Hair/bone ratio: 3.84			

x/ The sample tabulated in the first position i femur, all other are vertebrae.

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Any larger increases in lead intake will be reflected sooner in the hair than in bone tissue, due to both the smaller mass and shorter period of growth of hair. So one might expect that after a large short-time contamination with lead, the hair to bone ratio would be greater than the average ratio found in a "normal" population.

In TABLE 22 are presented the lead concentrations found in the hair of persons working in the lead industry, with a known history of small intoxications. It will be seen that these concentrations are nearly one order of magnitude higher than the levels found in an unexposed group.

TABLE 22. Concentration of lead in the hair of workers in the lead industry

No. of person	µg Pb/g of hair
1.	50.0
2.	69.0
3.	100.2
4.	85.3
5.	39.3
6.	47.7
7.	67.2
8.	107.1
9.	54.4
10.	87.2
Average	79.7

It seems that similarly as in the case of radioactive lead /see par. 6.2./ hair may be used successfully for the detection and evaluation of internal contamination with stable lead. It should however be emphasized that metabolic patterns of acute contamination differ from those of prolonged "natural" ones. This will effect both the hair to bone ratio /JAWOROWSKI, 1965 B/ and probably also the relative concentrations of lead in different parts of the skeleton. E.g. in acute absorption of abnormal quantities of lead its concentration in flat bone was found to be much higher than the concentration in long bones, being exactly the opposite of the relationship usually found under "normal" environmental conditions /KEHOE, 1961 B/.

Stable lead in some animal tissues

It will be interesting to compare the lead levels found in humans with those present in domestic and wild animals. For this reason in TABLE 23 are presented the concentrations of lead found by the present writer in some animal tissues.

As may be seen from TABLE 23 the lead content in the bones of wild animals in Poland is much lower than that of domestic cows.[x/] This may be an effect of the cows grazing along the sides of the roads /a custom popular in the region from which the cow samples originated/, on grass which may have two or more order of magnitude

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x/ For approximation wet weight units /cows/ to dry weight units /bison and red deer/ the latter should be divided by 1.7.

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TABLE 23. Stable lead in some animal tissues from Poland

Animal and tissue	Year of deth	µg Pb/g	
European bison x/	1958		
rib		3.78	
hair		3.5	
Red deer x/			
mandible	1961	4.20	
mandible	1961	7.46	
temporal	1961	9.18	
temporal	1962	5.41	
			6.56
antlers	1946	2.53	
antlers	1949	1.54	
antlers	1957	1.08	
antlers	1959	2.02	
antlers	1960	1.28	
antlers	1961	2.60	
antlers	1961	2.14	
antlers	1962	2.14	
			1.92
Cow xx/			
tibia	1963	4.27	
tibia	1963	21.20	
tibia	1963	2.41	
tibia	1963	35.00	
tibia	1963	36.12	
tibia	1963	11.50	
			18.41

x/ µg Pb/g dried sample xx/ µg Pb/g wet bone

higher concentrations of lead tha
away from the road /CANNON et al.

The lead concentrations foun
µg/g wet/ are higher than the con
from the same region /2.32 µg/g w
found in human vertebra recalcula
/using the factor of 3.27 from TA
in human cortical bone of about 7
times lower than the concentratio
other hand the concentrations of
and temporal bones of wild animal
concentrations in humans. This in
sources of lead in the food-chain
access than humans and cows.

Intake and elimination of stable

The approximate daily lead ba
been calculated by KEHOE /1964 A/

KOEHOE /1964 A/ has measured
verages in ten healthy adults ove
and found that it ranged from 0.1
intake from the air was calculate
centration is 2 µg per cubic mete
the lung is about 50 per cent. It
urban air the lead concentration i

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may be inferred from TABLE 24 as about 6.0 $\mu g/m^3$. So the contribution from air would amount to 0.09 mg daily which is not much different from the value calculated by MONIER-WILLIAMS /1950/ for residents in England. In MONIER-WILLIAMS calculations both the total intake and total output was 0.4 mg per day. In environmental conditions other than those studied by both authors, the values of the lead balance may differ considerably.

TABLE 24. Lead balance in average "normal" U.S. resident /After KEHOE 1964 A/

Daily intake /mg/		Daily excretion /mg/	
from food and drink	0.3	in feces	0.3
from air	0.03	in urine	0.03
totals	0.33		0.33

Somewhat less than 10 percent /nearer 5 per cent/ of ingested lead is absorbed to the circulatory system, some of this being excreted back to the alimentary canal /KEHOE 1964 A/. This is in agreement with the results of MIYASAKI /1930/ and EHRENS /1925/ who in animal experiments found that lead may be freely absorbed from the intestinal tract and excreted into it again. ALIAVDIN et al. /1936/ found that 10 - 30 times as much lead may be excreted

from the liver by way of the bile as in urine, which confirmed the earlier findings of WEYRAUCH /1930/.

In man, the output of lead from the body is mediated mainly by the gastroenteric tract and with urine. From the amounts excreted in these ways about 10 per cent is excreted in urine and the rest in feces /CHOLAK et al., 1943/. Some amounts of lead are excreted in sweat, the concentrations of lead therein being of the same order of magnitude as those in urine /KEHOE, 1964 A/. Some amounts are eliminated also in sebum /KLIGMAN, 1961/ and in keratinised epidermis, nails and hair. The amounts of stable lead which in an average Polish resident are excreted with the growing hair mass may be calculated from the known concentration of lead in the hair i.e. 8.9 µg Pb/g /TABLE 21/ and from the mean daily increase of the total body hair mass. This last value was determined as about 0.3 g per day /JAWOROWSKI, 1963 C/, thus the total excretion of lead in hair may be estimated as about 0.003 mg daily, i.e. about one tenth of the amount excreted in urine. In fury animals, however, this proportion is quite different, as up to 50 per cent of the body lead may be accumulated in hair /SCHWARTZ, 1960/.

The measure of the rate of elimination of an element from the body is its biological half-life. This elimination is a complicated process, rather difficult for exact mathematical interpretation /NORRIS et al. 1958/. Not being a clear-cut concept the biological half-life is however often used in the determination of the retention and residence time of nuclides in the body. For practical

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purposes a single exponential model is used in which

$$T_b = \frac{0.693\ m\ C}{I\ f} \qquad /EQ.2/$$

where T_b is the biological half-life in days of the element, m, is the mass in grams of the organ of reference /or total body/, C is the concentration of the element in grams per gram of tissue of the element, I is daily intake in grams of the element and f is the fraction of the daily intake retained by the organ or total body.

The values of biological half-lives of lead in different organs and in the total body tabulated in the ICRP Report /1960/, were calculated from the previously published data on "standart man" /m/, lead concentrations in soft tissues and bones /C/, lead intake /I/, and absorption rates in organs /f/. Basing on some more recent data HOLTZMAN /1960 A/ revised the ICRP values of lead half-lifes in total body, skeleton, liver and kidney, and JAWOROWSKI /1965/ in the skeleton. These values are tabulated in TABLE 25.

TABLE 25. Half-lives of stable lead /T_b/ in the human body /in days/

	Total Body	Skeleton	Liver	Kidney
ICRP /1960/	1460	3650	1947	531
HOLTZMAN /1960/	2085	6350	800	59.8
JAWOROWSKI /1965/	-	4620	-	-

It should be noted that the value of the stable lead half-life in the skeleton calculated by the present author is not deduced from the exponential model of T_b, but from the data on bone remodeling, which will be discussed in detail in par.6.2.

From /EQ.2/ the value of f_w i.e. the fraction of the ingested lead retained in the body may be deduced if the remaining factors in this EQUATION are known, as

$$f_w = \frac{C \cdot 0.693}{T_b \cdot I} \qquad /EQ.3/$$

The measured value of daily lead intake in the United States is 0.33 mg KEHOE 1964 A, the value of C may be inferred from TABLE 16 for United States residents as about 150 mg, and the value of T_b in the total body may be accepted as this calculated by HOLTZMAN /1960 A/ of 2085 days.

Inserting these values into /EQ.2/ the factor f_w, can be deduced as 0.15, a value almost twice as high as that found experimentally by KEHOE /1961 B/ in two healthy subjects. This discrepancy indicates that both the values of the factors used in /EQ.3/ and the values of fractional retention as found by KEHOE /1961 B/ are subject to uncertainty and need further study.

Essentiality and toxicity of lead

It is not known as yet whether lead is an essential trace element for mammals. According to SCHROEDER et al./1961/, a trace

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metal is essential for mammals if it has the following characteristics: 1/ is readily available in the Earth's crust, 2/ is present in plants and mammals, 3/ shows biological activity in vitro, 4/ its molecular weight is below elements in the lanthanum series, 5/ is relatively nontoxic to mammals orally. Only two of these criteria are fulfilled by lead. The known toxicity of lead argues against the essentiality of this element for mammals [x/]. In man symptoms of poisoning were observed after a daily ingestion of only 1 to 2 per cent of the "normal" body content of lead i.e. after about 0.4 to 2.5 mg per day in addition to the lead in food SCHROEDER et al. /1961/. From the toxicological point of view the body burden of lead may be divided into the lead deposited in the skeleton, not engaged directly in evolving the toxic symptoms and the circulating lead, responsible for the poisoning effects. This deposited lead, in some circumstances may be transferred back to the circulation, to produce intoxication /LANGENBACH et al. 1961/. The treshold levels in blood above which the toxic symptoms may be expected were recently suggested as 0.5 µg Pb/g /EGLI et al.1957/, whereas former publications suggested a value of 0.8 µg Pb/g /CANTAROW et al. 1944; HAMILTON et al. 1949/. These threshold values are surprisingly near the average existing lead concentration in the United States of 0.3 µg Pb per gram of blood. It seems improbable that nature defended the body so poorly as to leave

x/ Cadmium, however, which is toxic, has been considered as possibly essential /VALLEE, 1959/.

such a narrow margin of safety. Rather one should anticipate, that man-made contamination of the environment resulted in increasing the body levels of lead.

It should be emphasized that the present concept of the treshold limits for lead in the human body and environment is base on the existing levels in the body and also upon industrial standards which not necessarily are suitable as standards for the general population.

However, the widely accepted and officially sanctioned assumption that the existing lead levels in the body are natural and safe, is not supportedy by any acceptable evidence, with the exeption that during the last twenty years no lead increase has been found in the residents of large American cities /KEHOE,1964 B, tacidly accepting that the earlier lead contaminations of human environment were non existent. This point will be discussed later on in par.7.1. On the other hand the industrial standards are based on the "axiom that the worker must be either perfectly healthy or classically intoxicated with lead but cannot be either" /PATTERSON 1965/. Thus, the presently accepted standards for population should be regarded as "toxic limits" and not "safe limits". That the occupational standards and policy cannot be applied for general population is well recognized in radiological protection and also by classical toxicologists. According to MONIER-WILLIAMS /1950/ "it cannot be emphasized too strongly that in discussing the amount of lead which may be considered as negligible in food, consideration

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of toxic limits, so far as these are defined by the appearance of symptoms of poisoning, is beside the point, and tends to obscure the real question. What we want to know is not so much the toxic limit, as the safe limit, if indeed any limit, however small, for a cumulative poison can be regarded as safe. We cannot assume that there is a sharp dividing line between what is obviously toxic, giving rise to lead colic or other symptoms, and what is completely harmless. In all probability, there is a range of lead intake between these two extremes in which some effects, however slight, are produced upon metabolism, effects which, clinically, may be difficult or impossible to detect or to ascribe to their real cause ...".

The effects of the chronic lead insult caused by levels of lead comparable with the "normal" ones were recently noted in literature /MONCRIEFF et al. 1964, MARSH 1964, LANE 1964, HENDERSON et al. 1957/. Pathologic and histologic changes in the brain and spinal cord and functional disorders of the higher nervous activity were induced in animals by exposures to atmospheric lead concentrations at levels comparable to those in the air of large cities /GUSEV 1960/. In humans "the intellectual irritability and disfunction are associated with classical lead poisoning, and it is possible ... that similar impairments on a lesser but still significant scale might occur in persons subjected to sever chronic lead insult" /PATTERSON 1965/. BRADLEY et al. /1956/ found in children a "normal" lead level in blood of 0.5 to 0.6 µg/g, which is equal or somewhat higher than the accepted treshold levels. The

overlapp has been noted by these authors between the values of lead levels in the blood of children with and without the clinical symptoms of poisoning.

Altough it is not certain whether lead is essential for the human body it is certain that it is essential for wellfare and industrial development. One may expect, that the man-made pollution of the environment with lead will be more or less permanent and in all probability will increase. This is why the existing gap in our knowledge of the biological effects of small chronic lead intoxications needs to be filled by further studies.

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6.2 RADIOACTIVE LEAD IN THE BODY

For metabolic studies and for calculation of the radiation dose absorbed by man from natural sources it is essential to know, the total lead-210 content of the body. The body burden of lead-210 of an average member of a population may be estimated by applying the uptake or excretion data to a metabolic model of this nuclide or by measurement of its concentrations in different tissues. Both approaches have some limitations, resulting in uncertainty of the assessements. The uptake and excretion calculations are limited by the scarcity of intake and output data and also by the uncertainty of some metabolic parameters. The more direct approach, the measurement of lead-210 in tissues, is somewhat limited by the fact, that the great majority of studies concerned bone concentrations, much less informations being provided for soft tissues. In both methods the data on stable lead metabolism and distribution in the body may be applied. It seems however that the existing amount of information on the intake and excretion of lead-210 in man is too small to make a calculation of the world average body burden of lead-210 anything but highly uncertain. The excretion was studied only in one subject in United Kingdom /HILL 1965/ and in three subjects in the United States /HOLTZMAN 1963 A/. On the other hand, tissue concentration measurements were performed in more than a hundred subjects in various countries. Therefore, in the present study, the estimation of the average lead-210 body burden was based on these last measurements.

Tissue distribution of lead-210

The total body burden of lead-210 in the body may be inferred from the known body concentrations and from the data on the lead-210 distribution in the body, found by different authors, and given in TABLE 26.

TABLE 26. Tissue distribution of lead-210 in human body

Tissue	Lead-210 concentration /pCi/g wet tissue/	Fraction of total body burden /per cent/		Source of information
		Lead-210	Stable lead[x/]	
1	2	3	4	5
Bone	0.050	63		HOLTZMAN 1960 B
	0.026	58		JAWOROWSKI 1963 C
	0.028	80		OSBORNE 1963
	0.045	81		MUTH et al.1964
Average	0.037	70	70	
Liver	0.011	1.7		HOLTZMAN 1960 B
	0.004	2.8[xx/]		OSBORNE 1963
	0.0035	1.5[xx/]		MUTH et al.1964
Average	0.0088	2.0	5.1	
Muscle	0.006	17		HOLTZMAN 1960 B
	0.001	13[xx/]		OSBORNE 1963
	0.0012	9.2[xx/]		MUTH et al.1964

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1	2	3	4	5
Average	0.0027	13	7.7	
Spleen	0.003	0.18[xx/]		OSBORNE 1963
	0.0025	0.09[xx/]		MUTH et al.1964
Average	0.003	0.14	0.2	
Kidney	0.007	0.13[xx/]	0.6	OSBORNE 1963
	0.0037	-		
Average	0.005			
Blood	0.0014	2.0[xx/]	2.4	MUTH et al.1964
Soft tissues: pooled equal amounts of liver, muscle, spleen, kidney, thyroid, heart, duodenum, stomach, pancreas, brain, skin, prostate, testes; average for two subjects	0.003	42[xxx/]	38	JAWOROWSKI 1963 c
Overall average in soft tissues	-	30[xx/]	30	

x/ Average values from TABLE 17.
xx/ Calculated from bone concentrations and fractional skeleton content as estimated by respective authors.
xxx/ Calculated assuming that concentration in muscles is 1/3 of that in pooled samples.

The data compiled in this TABLE have two important drawbacks: one is the limited number of analyses from which the soft tissues data were derived, and the second is that the types of soft tissue analysed, contribute only a fraction to the total body mass of soft tissues. This leads to uncertainty in the estimation of the contribution of soft tissues to the total burden of lead-210. From that point of view, the stable lead contents in soft tissues, /TABLE 17/, are probably more certain, as they were determined in larger sets of samples. It is surprising however that the overall averages of fractional lead-210 contents in the skeleton and in soft tissues are the same as respective contents of stable lead. This coincidence suggests that the values of 70 per cent in the skeleton and 30 per cent in the soft tissues are probably close to the real distribution of both types of lead in the body.

Lead-210 in bones

Bone is a tissue in which the majority of the lead-210 in the body is concentrated and therefore was, up to now, the main subject of study.

The average concentration of lead-210 in bone found by different authors are compiled in TABLE 27. The data presented in this table are confined to the residents of the temperate Zone of the Northern Hemisphere, in highly developed countries. The data pertaining to the residents of other regions will be discussed in par. 7.2.

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TABLE 27. Average concentrations of lead-210 in human bones

Population group and year of collection or publication	Lead-210 concentration pCi per gram wet bone	Source of information
U.S., 1952	0.013 x/	HURSH 1960
U.S., 1960	0.035 x/	HOLTZMAN 1960 B
U.K., 1961	0.026	JAWOROWSKI 1964
U.K., 1963	0.018	OSBORNE 1963
Germany, 1963	0.032	RAJEWSKY et al.1963
U.S., 1963	0.037 x/	HOLTZMAN 1963 A
Germany, 1964	0.037	STAHLHOFEN 1964
Poland, 1964	0.041	JAWOROWSKI 1965 B
Germany, 1964	0.045	MUTH et al.1964
Average	0.032	

x/ Calculated using the value of 0.4 for ash /wet bone ratio/.

It should be noted that the majority of values compiled in this TABLE were determined as concentrations of polonium-210, excepting those measured by the present author /JAWOROWSKI, 1964 and JAWOROWSKI, 1965/, who in his sets of samples separated radioactive lead. The average value of 0.032 pCi per gram of wet bone, deduced from TABLE 27 is probably too low, as the existing mean for the population groups involved.

It was pointed out /JAWOROWSKI 1965 B, 1966/, that the increase in the lead-210 levels in human bones in the last few years /TABLE 27/ is probably caused by the introduction of man-made radiolead into the biosphere. Therefore it seems that the most recent values presented in this TABLE, rather than the results of all determinations should be used for estimation of the existing mean concentration. This recent mean concentration in continental Europe may be deduced from TABLE 27 as about 0.04 pCi per gram wet bone.

The existence of some correlation between the age of subjects and the lead-210 concentration in bone was suggested by HOLTZMAN /1960 B/, but was not confirmed by the later findings of the same author /HOLTZMAN 1963 A/ nor by the findings of STAHLHOFEN /1964/ and the present writer /JAWOROWSKI 1965 B/. BLANCHARD /1966/ suggests that a correlation with age may exist during the first three decades of life, however this author did not find in his set of samples any linear correlation.

It is interesting to compare the concentrations of lead-210 in the human bones with those in the bovine bones. In five samples of bovine bones, collected in 1963 in Warsaw region the present writer have determined lead-210 and polonium-210 x/. The results are given in TABLE 28.

As may be seen in this TABLE the mean concentration of lead-210 found in bovine bones was 0.82 pCi per gram wet bone, a value some 20 times higher than the mean concentration found in humans from the same region /TABLE 27/. In two bovine bone samples collected in Gr.Britain the present writer found mean lead-210 con-

x/ Polonium-210 has been determined according to the method of BLACK 1961.

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centration of 0.25 pCi per gram wet bone, i.e. ten times as high as concentration found at the same time in humans from London region /TABLE 27/. These concentrations are not much different than those found by DI FERRANTE /1961/ in United States cattle and by HOLTZMAN /1966 B/ in Iceland lamb, but are lower than found in Arctic animals /TABLE 15/.

TABLE 28. Concentrations of lead-210 and polonium-210 in bovine bones /In pCi per gram wet bone/

No.	Lead-210	Polonium-210	$^{210}Po/^{210}Pb$ ratio
1.	0.25	0.21	0.84
2.	1.35	0.12	0.10
3.	0.23	0.16	0.70
4.	1.50	0.14	0.09
5.	0.83	0.28	0.33
Average	0.82	0.18	0.41

Total body content of lead-210

Using the data on tissue distribution of lead-210 compiled in TABLE 26 and assuming that the mean concentration of lead-210 in the skeleton is 0.04 pCi per gram wet bone, the average body burden may be calculated as 400 pCi /TABLE 29/. It should be noted, however, that this estimation is in all probability valid only for

the residents of the Northern Temperate Zone, where majority of determinations of lead-210 content in the human body has been performed. In other geographical regions the body burden of lead-210 in humans may be different, being higher in the Arctic and probably lower in the south hemisphere /see par.7.2/.

TABLE 29. Lead-210 body burden in the residents of Northern Temperate Zone

	Lead-210 burden /pCi/	Fraction of total lead-210 activity /per cent/
Skeleton	280	70
Soft tissues	120	30
Total body	400	100

Lead-210 in hair

Besides bone, high concentrations of lead-210 have been found in hair /JAWOROWSKI 1963 C, 1965 B/ and the present writer found that hair may be used as a medium for monitoring the occupational poisonings with this nuclide and with radon and its daughters /JAWOROWSKI 1964, 1965/. It seems also that hair may be especially useful for studying the natural contaminations in residents of remote geographical regions due to the ease of collecting samples. Therefore the present author and his coworkers investigated in

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animal experiments some of the patterns of incorporation of lead--210 into the hair essential for the monitoring of the internal contamination of the body with this nuclide, and also with stable lead /JAWOROWSKI et al.1967 B/. The results of this investigation will be below briefly reviewed.

Monitoring of internal contaminations of the body with radionuclides is usually performed by whole body counting or analysis of excreta. Whole body counting, though an excellent and precise tool for determination of gamma emitting contaminants, is somewhat limited in application for other types of nuclides. For this and also for economic reasons the techniques based on radiochemical analysis of excreta are still widely used. These techniques, however, yield very unreliable information regarding body load and accidental over-exposure in the case of both long- and short--living radionuclides. Using radium-226 as a model HURSH /1958/ critically revised the value of urine and feces analyses for detection and evaluation of constant and accidental exposures. This author found that in the case of constant exposure, the excreta sample reflects predominantly recent daily exposures, 75 per cent of the sample originating from the four preceeding days. Thus the excreta measurements do not define the body burden but the most recent exposure history. For the same reasons, the excreta sampling techniques are able to detect large single accidental exposures only when the sampling is done at short intervals of the order of 1 to 2 weeks. In the case of exposure to small doses and

in evaluating a new program even more frequent sampling would be preferable.

In this respect, the "hair monitoring" technique offers some essential advantages. Radionuclides are accumulated by the hair during the period of its growth, which is of the order of many months. Owing to so long an accumulation, this excretory route enables effective monitoring of accidental and also permanent exposures to be made without the necessity of frequent sampling. Besides, the hair technique offers the advantage of extremely low detection limits in spotting the single exposures. The ease of sampling and simplicity of analytical procedures due to the minute mass of the samples and to the relatively high concentration of nuclides in them are unsurpassed.

Some 30 elements are known to be excreted to the hair from the body /SCHWARTZ 1960/, among them several important radionuclides. According to SCHWARTZ /1960/ hair is an important channel for excretion of metals. In some instances more than half of the administered element may be excreted to the hair /KRAUT et al. 1944/. External contamination is a most unlikely explanation for the high content of these substances in hair /SCHWARTZ 1960/.

The site of accumulation of radionuclides is predominantly the hair matrix of the hair follicle. In the fast-growing cells of the matrix, the radionuclides combine with the sulfhydryl groups of precursors of keratine. These sulfhydryl groups are known to be highly reactive with metals and this explains their special

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affinity for hair /WIDDOWSON et al. 1964/. Subsequently, radionuclides are incorporated into metabolically inactive keratin crystals, which constitute some 15 per cent of human hair /MARCHLEWSKI et al. 1947/. Once in the hair, the elements are never reabsorbed /WIDDOWSON et al.1964/ neither does any washing procedure remove them.

Using lead-210 as model "bone and hair seeker" the relationship between its incorporation into both these tissues was studied in an experiment with animals as well as the dependence of incorporation of the hair cycle and on the dose rate. The results of this study are summarized in TABLE 30.

TABLE 30. ^{210}Pb uptake by resting and growing hair of rabbits

Dosage		Mean incorporation of dose, per cent per gram of tissue / x 10^{-3} /			Anagen hair to bone ratio
Fractionation	Total dose /in uCi per animal/	Telogen hair	Anagen hair	Bone /wet, femur/	
1 dose	0.087	3.0[x/]	21[x/]	98	0.22
1 dose	0.612	1.3	17	43	0.41
2 doses	0.175	0.5[x/]	28	74	0.37
19 doses	0.332	3.6	46	71	0.65
19 doses	1.660	0.4	49	61	0.80

[x/] For one animal, all other values are averages for 5 animals.

In this experiment a new hair cycle was induced in all the animals on the left side of the trunk by plucking the hair from an

area of more than 100 cm^2. Thus each animal had old resting /telogen/ hair on the right side of the trunk, whereas on the left new growing /anagen/ hair developed. The animals were immobilized in a manner sufficient to prevent contamination of the hair by saliva or excreta. Subcutaneous injections of lead-210 acetate were started on the sixth day, when the new anagen hair emerged above the skin surface. Twenty one days later the animals were sacrifice and the tissues were analysed.

The striking feature of the data presented in TABLE 30 are the high concentrations of lead-210 found in growing hair comparable with those in bones. In hair, these concentrations ranged from 0.017 to 0.049 per cent of the administered dose per gram of hair. On the other hand, the concentrations in resting /telogen/ hair were found to be by one to two orders of magnitude lower. This difference seems to be related to the well-known morphological and physiological differences between both kinds of hair. The hair matrix which is characterized by a high metabolic turnover and rich in sulfhydryl groups, is present only in anagen hair. The telogen hair, on the contrary, is a dead structure with no metabolism, to which the material solved in body fluids has but poor access if any. The small amounts of lead-210 found in telogen hair samples might derive from the excretion of sebaceous glands /KLIGMAN 1961/ or from the small amounts of lead-210 excreted with urine or feces which may have contaminated the fur of even immobilized animals. However this is hardly probable since the activity

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found in hair washings did not exceed 0.05 per cent of the lead-210 content in the corresponding hair samples. A more plausible explanation is that lead-210 was introduced into the telogen samples with the small amounts of anagen hair present in the apparently synchronized telogen fur.

In anagen hair lead-210 concentrations were closely related to the varying amounts of radionuclide injected and also to the mode of dose fractionation. It is interesting that in the animals treated with a single dose, the percentage of the dose incorporated into anagen hair was 2.5 times lower than in those which had received 19 doses, the mean respective percentages being 0.019 and 0.047 of the total dose per gram of hair. An intermediate value of 0.027 per cent was found in animals treated with two doses.

The differences in the percentage of incorporated dose might have resulted from the differences in mass of the fast-growing hair matrix cells available to the radiolead atoms, and from the differences in the duration of the high radiolead level in the circulating blood. In animals treated with the fractionated dose, this mass was greater and the contact of ^{210}Pb with the hair matrix longer than in those treated with a single dose.

The incorporation of lead-210 into the growing hair was also related to its accumulation in bone, the hair to bone concentration ratio ranging from 0.22 to 0.80. Higher values of this ratio were found in animals treated with the fractionated dose. This ratio

was similar to the values ranging from 0.1 to 0.66 observed by u in four naturally contaminated cattle specimens /unpublished dat but was lower than the mean value of 1.34 found in previous investigations carried out on naturally contaminated humans /JAWO-ROWSKI 1964/. The high value of the hair to bone ratio in humans probably caused by their having a far lower amount of hair in pr portion to body-weight as compared with rabbits or cattle. The p of the body lead which is excreted to the hair is, therefore, in fur-bearing animals more diluted in the total mass of hair than humans.

It was found in this experiment that the hair cycle is an e sential factor in the process of excretion of ^{210}Pb, and in all bability also of other nuclides, to the hair. This factor should considered in interpreting the results of hair analysis since th proportion of resting and growing hair is rather variable, the p centage of resting hair in humans ranging from 4 to 37 per cent /VAN SCOTT 1959/.

The sensitivity of "hair monitoring" may be estimated by co parison with whole-body counting and urine analysis in the case a single accidental exposure, which took place three months befo monitoring. The lowest single contamination of the body with ^{226} which after 90 days /5 per cent retention/ can be detected by wh -body counting, with counter detection limit 0.0001 µg ^{226}Ra equ valent, is 0.002 µg ^{226}Ra.

The lowest single contamination of the body with ^{226}Ra, whi

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after 90 days /excretion 0.0015 per cent/ can be detected by urine analysis /detection limit 0.26 pCi per 24 hour sample/ is as high as 0.024 μg ^{226}Ra.

The rate of hair growth is of the order of 1 cm per month. The age of scalp hair in men is about 1 to 15 months. It is relatively easy to collect, when necessary, samples older than 90 days. During a lapse of three months /or more/ between incorporation of the radionuclide and monitoring, no lose in its content can be observed in the hair so that its concentration will quantitatively reflect the old intake of nuclide. From a single dose which entered the circulation 0.02 per cent was incorporated per gram of hair /TABLE 30/. The detection limit for ^{210}Pb is 0.036 pCi per gram hair. Thus the lowest single exposure, which can be detected after a longer time period /say several months/ by the "hair method", is 0.00018 uCi.

TABLE 31. Lowest single internal exposures detectable after 90 days /No natural contamination taken into account/

Technique	Nuclide	Exposure μCi per total body
Whole-body counting	^{226}Ra	0.002
Urine	^{226}Ra	0.02
Hair	^{210}Pb	0.00018

As seen from TABLE 31 hair monitoring affords a two to three

orders of magnitude higher detectability of old single exposures than do other recognized health physics techniques.

The conclusion of the above reviewed investigation is that hair provides an excellent medium for detection and evaluation of radiopoisonings. The concentrations of radionuclides in the hair are related to those in critical organs and also to the dose administered. Two other features, i.e. the relatively high percentage of the incorporation and long-term storage of nuclides in hair ensure the high sensitivity of this method.

Elimination of lead-210 from the body

From the total amount of lead-210 present in the body a part is eliminated by different excretory routes. In humans these main excretory routes are feces and urine, the excretion through skin and skin appendages being of minor importance /JAWOROWSKI 1963 C/. However, in furry animals hair may play an important role in the excretion of radiolead. On the basis of the stable lead studies /see par. 6.1/ it is usually accepted that in "naturally" contaminated humans, some ten per cent of excretion of lead-210 is mediated by kidneys and the rest by the gastrointestinal tract /HOITZMAN 1963 A/.

In animal experiments, however, in which radiolead was administered by inhalation or injection these proportions were found different. HORIUCHI et al./1958/ found that, in rats, 20 days after inhalation of lead-210, 35 per cent of the dose had been eliminated

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with feces and 12 per cent with urine. This is in agreement with the results of CASTELLINO et al. /1964/ who found in rats injected with lead-210, that during 14 days of experiments, 35.7 per cent of the administered dose was excreted with feces and 15.9 per cent with urine. The rate of excretion by both these ways, was however strongly dependant on the time which had elapsed after injection of the radiolead. When the time after injection was short the rate was more than twice as high in feces as in urine. In both feces and urine the excretion reached maximum at 12 to 24 hours, was maintened for 72 to 96 hours and then decreased asymptotically, both excretion following the same course. At the later stage of the experiment i.e. on the 14th day, the same fractions of injected dose were excreted with urine as with feces. This is in agreement with the results of TOMPSETT et al. /1935/ who found in "normal" humans the excretion of stable lead to be equal in urine and feces.

The source of lead in the feces is bile, with which this element is readily excreted into the gastrointestinal tract /see par.6.1/ or lead ingested with food, which had not been absorbed into the circulation. This is why the experimental results indicating roughly equal urine and fecal excretion of radiolead from older single paraenteral exposures may considerably vary from the corresponding excretions found in naturally contaminated humans. In cases where the majority of lead-210 gain entrance into the body by daily ingestion, the non-absorbed lead-210 will dominate the concentration in feces, so that over-all elimination /i.e. excretion of absorbed and non-absorbed lead-210/ through the gastrointestinal tract will be greater than through the kidneys. There is however lack of data on the amount of lead-210 excreted, in uncontaminated humans, in urine and feces, which could vary this assumption. In contaminated humans, however, in whom excretion from old deposits is higher than from daily intake, the fecal and urine contributions will be probably roughly equal.

In "normal" humans the fecal excretion of lead-210 was found ranging from 1.7 to 6.4 pCi per day /HOLTZMAN 1963 A, HILL 1965/, and in urine the average excretion was found to be 3.8 pCi per day /MUTH et al.1964/. In urine lead is partially in ionic form, probably as lead phosphate, whereas a part of it is chelated or complexed with some organic substance /DINISCHIOTU et al.1960, NELSON et al.1958/.

After paraenteral administration, lead-210 disappears from the tissues and organs in a complex mode, which may be represented by a single exponential function, by the sum of two exponentials or by the sum of three exponentials /CASTELLINO et al. 1964/. According to these latter authors "the ionic lead injected can be supposed to enter the extracellular space very shrotly after injection. It is then lost from this partly to the blood and partly to the intracellular space. Inside the cells, the ionic lead may then be bound by absorption or complexing, or may return to the extracellular space and thence to the blood. The bound lead may either be released as ionic lead inside the cell, or it may be eliminated from

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the cells as complexed lead."

Half-lives and effective equilibrium of lead-210 and its daughters

The measure of the rate od disappearance of a nuclide from the organ or total body is the biological half-life, T_b, defined in par.6.1., and in the case of a radioactive substance the effective half-life, which is

$$T_{ef} = \frac{T_b \cdot T_r}{T_b + T_r} \qquad /EQ.4/$$

where T_r is radioactive half-life of the subetance.

In ICRP Report /1960/ are tabulated the values of T_b and T_{ef} of lead-210 in total body and various organs, which were calculated from a single exponential model of the biological half-life /EQ.2/ and from results of animal experiments with radioactive lead and from stable lead data from human subjects. HOLTZMAN /1960 A/, however, has shown that the ICRP values of T_b and T_{ef} are inconsistent with the EQ.2 and the values pertaining to the tissue distribution and ingestion of lead, tabulated in ICRP Report. It is not clear whether this discrepancy is the result of a misprint or other error. The present writer /JAWOROWSKI 1965 A/ calculated the lead-210 half-lives in the skeleton from the rate of bone remodelling. The values of these half-lives are listed in TABLE 32.

TABLE 32. Half-lives of lead-2

/T_b is biological hal

Tissue	Source of
Total body	ICRP /1960/ HOLTZMAN /1
Skeleton	ICRP /1960/ HOLTZMAN /1 JAWOROWSKI
Liver	ICRP /1960/ HOLTZMAN /1
Kidney	ICRP /1960/ HOLTZMAN /1

x/ Calculated accepting 7100 da lead-210.

xx/ Calculated from bone remodel

The striking differences w between the ICRP values and tho by misprints or miscalculations different data on intake and ti MAN's calculations. The uncerta

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however, impair the reliability of the results of the calculations, based on single exponential model. A more direct approach, such as using the bone remodelling data in the case of skeleton half-lives /JAWOROWSKI, 1965/ or kinetic distribution studies in animal experiments /HORIUCHI et al. 1958, CASTELLINO et al. 1964/ will probably supply more reliable data. It should be noted that the results of animal experiments /CASTELLINO, 1964/ indicate in general much shorter biological half-lives of lead than those tabulated in TABLE 31. No plausible explanation of this discrepancy, however, can be presented at this time and it seems that further experimental studies are needed to elucidate the problem.

The radiation dose absorbed by a tissue from the emmission of the lead-210 chain depends mainly on the high-energy alfa particles emitted by polonium-210 daughter of lead-210. Therefore it is essential to know the proportion between the amount of polonium-210 and the amount of parent lead-210 present in a tissue, i.e. $^{210}Po/^{210}Pb$ ratio.

The term "radioactive equilibrium" is sometimes used in literature to describe $^{210}Po/^{210}Pb$ ratio in human and animal tissues. This term however, which in the case of ^{210}Pb-^{210}Bi-^{210}Po chain can be described as

$$\frac{A_3}{A_1} = \frac{1}{\left(1 - \frac{\lambda_1}{\lambda_2}\right)\left(1 - \frac{\lambda_1}{\lambda_3}\right)} \qquad \text{/EQ.5/}$$

where A_1 and A_3 denotes the activities of lead-210 and polonium-210 respectively and λ_1, λ_2 and λ_3 are radioactive decay constants of lead-210, bismuth-210 and polonium-210 respectively, refers only to the physical decay and build-up of these radionuclides. In the living tissue both parent and daughter products behave according to their effective half-lives and therefore in such an instance rather the term "effective equilibrium" should be used, which in the case of the lead-210 chain can be expressed as

$$\frac{A_3}{A_1}_{ef} = \lambda_2 \lambda_3 \left[\frac{1}{(\lambda_{2ef} - \lambda_{1ef})(\lambda_{3ef} - \lambda_{1ef})} + \right.$$

$$\left. + \frac{e^{-(\lambda_{2ef} - \lambda_{1ef})\,t}}{(\lambda_{1ef} - \lambda_{2ef})(\lambda_{3ef} - \lambda_{2ef})} + \frac{e^{-(\lambda_{3ef} - \lambda_{1ef})\,t}}{(\lambda_{1ef} - \lambda_{3ef})(\lambda_{2ef} - \lambda_{3ef})} \right] \qquad \text{/EQ.6}$$

where λ_{1ef}, λ_{2ef} and λ_{3ef} are the effective decay constants of lead-210, bismuth-210 and polonium-210 respectively and t is time during which the effective equilibrium is established.

The ratio of $^{210}Po/^{210}Pb$ in radioactive equilibrium may be calculated as 1.02 from /EQ.5/ accepting that λ_1 is 1.0×10^{-9} sec.$^{-1}$, λ_2 is 1.6×10^{-6} sec.$^{-1}$ and λ_3 is 5.8×10^{-8} sec.$^{-1}$.

This ratio in effective equilibrium in a tissue might be calculated from the values of effective half-lives of lead-210, bismuth-210 and polonium-210 in this tissue and from /EQ.6/. However, often these values are unknown or uncertain. On the other hand the ratio $^{210}Po/^{210}Pb$ is comparatively easy to measure both in animal

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experiments and in naturally contaminated humans. Then the measured values of $^{210}Po/^{210}Pb$ ratios may be used for veryfying the values of effective half-lives of the lead-210 chain members in the tissues.

An example of such as application of $^{210}Po/^{210}Pb$ ratio was given by the present author in a study on the half-lives and effective equilibrium of lead-210 and polonium-210 in dogs /JAWOROWSKI, 1965 A/ which is briefed below. In the ICRP Report the respective half-lives of lead-210, bismuth-210 and polonium-210 in the bone are given as 2400, 3.6 and 20 days. When applied into the /EQ.6/ these values give the $^{210}Po/^{210}Pb$ ratio of 0.1, actually tabulated in the ICRP Report. This value however is in variance with the ratio measured in the bones of naturally contaminated humans, which ranged from 0.97 to 1.18 /HOLTZMAN 1963 A, RAJEWSKY et al. 1963, MUTH et al. 1964/. This discrepancy might be caused either by the unsupported polonium-210, which could daily enter the skeleton in excess of lead-210, or by erroneous values of half-lives of the lead-210 chain members accepted in ICRP Report.

To elucidate this question the present author determined the $^{210}Po/^{210}Pb$ ratio in a long-term experiment in dogs, in which the influence of unsupported polonium-210 or bismuth-210 was eliminated. Lead-210 has been injected intravenously into eight mongrel dogs in amounts of 0.0078 to 0.037 uCi per kilogram of body weight. In different periods ranging from 85 to 392 days the animals were sacrificed and lead-210 and polonium-210[x/] were determined in their

x/ Polonium-210 has been determined according to method of BLACK /1961/.

bones, livers and muscles. The average values of $^{210}Po/^{210}Pb$ ratios found in this experiment are given in TABLE 33.

TABLE 33. $^{210}Po/^{210}Pb$ ratios in dogs after intravenous injection of lead-210. /Average values after JAWOROWSKI 1965 A/

Tissue	Lead-210 pCi/g wet	Polonium-210 pCi/g wet	$^{210}Po/^{210}Pb$ ratio
Bone	21.6	20.6	0.98
Liver	2.37	1.11	0.49
Muscles	0.65	0.061	0.136

As may be seen in the TABLE 33 the $^{210}Po/^{210}Pb$ ratio in bone tissue was found to be 0.98 being in agreement with the values found in humans. This result suggest that it is not unsupported polonium-210 but incorrect half-life values that are responsible for the discrepancy between the ICRP value of this ratio and the values determined in humans.

It should be noted that there may be two different half-lives of lead-210 daughters in the skeleton: the half-lives of unsupported polonium-210 and bismuth-210, e.g. in a situation when these nuclides were injected into the body, and the half-lives of supported nuclides i.e. originated in the tissue from their parent lead-210.

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to those ions which like sodium are poor "bone-seekers". As the Haversian remodelling is a slow process /HAM, 1953/, it seems that the majority of atoms of polonium which originated in the apatite from lead-210, will not leave the bone before their radioactive decay. Therefore it may be expected that the effective half-life of polonium-210 in the mineral bone will be similar to the radioactive half-life of this nuclide, and that the $^{210}Po/^{210}Pb$ ratio in the state of effective equilibrium in that particular compartment of bone will be similar to the $^{210}Po/^{210}Pb$ ratio in radioactive equilibrium.

The behaviour of bismuth-210 daughter of lead-210 in apatite crystals will probably follow the same patterns as that of polonium-210, therefore not essentially influencing the $^{210}Po/^{210}Pb$ ratio.

On the basis of the above considerations one can deduce the biological half-lives of bismuth-210 and polonium-210 originating in bone from lead-210, and also of lead-210 itself. If no fraction of lead-210 daughters deposited in the apatite crystals leave these crystals, except when they are dissolved during Haversian remodelling and if both liberated nuclides are not back-incorporated into bone, then the biological half-lives of bismuth-210 and polonium-210 in the apatite crystals will be determined by the rate of bone removal. In the state of equilibrium, when the rate of bone formation and bone removal are the same, the rate of bone formation is a measure of bone removal. FROST /1961/ applying tetracycline labelling of growing bone, found that in human cortical bone the rate of bone

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formation is 0.01-0.02 /mean 0.015/ per cent per day, which corresponds to a cortical bone "half-life" of approximately 3300 days. This is then the biological half-life of bismuth-210 and polonium-210 trapped in the mineral part of bone. The corresponding effective half-lives are 4.99 and 133 days.

The biological half-life of lead-210 will be longer than the half-life of bone because a part of the lead liberated from removed osteons will be reabsorbed by bone. If the fraction of reabsorbed lead is accepted to be the same as the fraction reaching the bone from the blood, i.e. 0.28 /ICRP, 1960/, then the biological half-life of lead in bone can be calculated as 4620 days, and the effective half-life as 2800 days. The results of these calculations are summarised in TABLE 34 together with corresponding data from other sources.

The values of the bone half-lives calculated by the present author for members of the lead-210 chain, when applied in /EQ.6/ give the value of $^{210}Po/^{210}Pb$ ratio in the state of effective equilibrium of 1.01, which agrees well with the experimental data and with ratios found in "normal" humans. This accordance suggests that the majority of polonium-210 in the skeleton of "normal" humans is supported polonium, originating in bone from parent lead-210.

This is however, not necessarilly the case in other tissues. In the liver e.g., OSBORNE /1963/ found $^{210}Po/^{210}Pb$ ratio of 1.8 and 3.1, MUTH et al. /1964/ an average ratio of 2.7 and BLANCHARD /1966/ 0.88 to 5.04. The high values of this ratio are in variance

with the value of 0.18 in ICRP Report /1960/ and the value of 0.49 found in an animal experiment by the present author /JAWOROWSKI, 1965 A/.

TABLE 34. Half-lives of lead-210, bismuth-210 and polonium-210 in the mineral bone

Nuclide	Source of information	Half-lives /in days/	
		T_b	T_{ef}
Lead-210	JAWOROWSKI /1965 A/	4620	2800
	ICRP /1960/	3650	2400
	HOLTZMAN /1960 A/	6350	3350
Bismuth-210	JAWOROWSKI /1965 A/	3300	4.99
	ICRP /1960/	13.3	3.6
Polonium-210	JAWOROWSKI /1965/	3300	133
	ICRP /1960/	24	20

The excess polonium determined in human livers and also in other tissues might have resulted from external sources i.e. from food, water and air or from internal sources i.e. from lead-210 present in the liver and from polonium redistributed from other tissues. There is probably no external source of the excess polonium as the data of HILL /1965/ and BLANCHARD /1966/ indicate that in plants and human diet the $^{210}Po/^{210}Pb$ ratio is near unity or lower, this

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also being the case in drinking water /HOLTZMAN, 1964 A/ and in air /FEELY et al. 1965/. Neither does it seem that the internal sources are responsible for the high values of this ratio found in human livers. In the afore quoted long-term experiment in which the ratio of 0.49 have been found in dogs, the influence of unsupported polonium-210 was practically eliminated. In this experiment all polonium measured in the liver was supported, i.e. originated from the parent lead-210 absorbed in the body, This supported polonium was composed of one part originating from the parent lead-210 in the liver and an other part redistributed from other organs. These two parts steadily replenishing the polonium content of the dog liver did not however, produce any excess polonium in this organ, the concentration of this nuclide, one year after lead-210 administration, being only some half of that of parent lead-210.

The contribution to the polonium content in the human liver of the redistributed polonium from other soft tissues and from the skeleton can be calculated from the exponential retention model EQ.2/ and from known content of lead-210 in the body. For this calculation several simplyfying assumptionsare to be made. One is that the polonium content of the body is the same as that of lead-210 /about 400 pCi/, i.e. that polonium is in an equilibrium with lead-210. Then the release of polonium from organs will depend rather on the biological and not on the effective half-lives of this nuclide. The next assumption is that the amounts of polonium in the skeleton and in soft tissues are roughly equal, i.e. about 200 pCi

in each. If we accept the biological half-life of polonium in the skeleton as 3300 days /JAWOROWSKI, 1965 A/ and 30 days [x/] in the soft tissues, then the portion of polonium-210 which is daily released from the skeleton is 0.04 pCi and the portion from soft tissues 4.6 pCi. Fraction of released polonium reaching the liver is probably the same as that reaching the liver from the blood, i.e. 0.17 /ICRP, 1960/, then the over-all contribution from the skeleton and soft tissues to the polonium concentration in the liver will be about 0.054 pCi/g [xx/]. This contribution is of the order of several per cent as compared with the concentrations of polonium in the liver found by OSBORNE /1963/, MUTH et al. /1964/ and BLANCHARD /1966/, and can be neglected as the possible internal source of the excess polonium in this organ.

As there is no excess polonium in the external sources, and no internal source can produce in liver the high $^{210}Po/^{210}Pb$ ratio, it seems that the only plausible explanation of this phenomenon is an assumption that the rate of absorption of lead-210 into the liver by ingestion and inhalation is lower than that of polonium-210, in other words, that there exists a discrimination of lead-210 versus polonium-210. The scarcity of data on the $^{210}Po/^{210}Pb$ ratio in

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x/ This value is arbitrarily accepted after total body T_b in ICRP Report /1960/ and is probably too low. Therefore the calculated amount of polonium released from soft tissues is probably too high

xx/ Accepting the Standard Man weight of liver of 1700 g.

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human tissues and in food, water and air make, at present, rather doubtful any evaluation of the numerical value of the supposed discrimination factor.

It is interesting that the value of factors F_w representing the fractions reaching the liver by ingestion tabulated in ICRP Report /1960/ as 0.0064 for lead-210 and about twice as high for polonium-210 i.e. 0.01, when applied in the exponential model of retention give the $^{210}Po/^{210}Pb$ ratio five to ten times lower than the ratio determined in human livers. Assuming that the T_{ef} of lead--210 and polonium-210 in the liver are as stated in ICRP Report /1960/ this ratio will amount to 0.54, and will be 0.68[x/] if more realistic values of T_{ef} were accepted for lead-210 and polonium--210, which are 719 days /HOLTZMAN, 1960 A/ and 80 days /JAWOROWSKI, 1965 A/ respectively. This discrepancy indicate that the factors f_w as tabulated in ICRP Report /1960/ are probably erroneous when applied to natural ingestion in humans, what may be due to the difference in the chemical form of lead-210 and polonium-210 naturally ingested in food as compared with that in the experiments the results of which were used in ICRP Report /1960/.

The half-lives and other metabolic parameters of pure, unported polonium should be clearly distinguished from those of polonium-210 supported by the parent lead-210, in cases when the whole

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x/ In this calculation was taken into account the contribution of polonium-210 originated from the parent lead-210 in the liver, accepted as 0.49 fraction of the lead-210 content /TABLE 33/.

^{210}Pb-^{210}Bi-^{210}Po system is metabolized. The important biological differences between the supported and unsupported polonium-210 are examplified by their biological half-lives which in the case of supported polonium are 139 times longer in the skeleton and 6.6 times longer in the liver.[x/] This differences may be due in part to the various physico-chemical status of both types of polonium in the living tissue. The supported polonium and also bismuth-210 as daughters of lead-210 may be introduced into such compartments of a tissue as are inaccessible for unsupported bismuth and polonium.

The effective half-life of supported polonium-210 in the liver in which the effects of redistribution and the influence of the lead-210 parent are included, can be obtained from the measured $^{210}Po/^{210}Pb$ ratio in dogs of 0.49 and from the expression derived from /EQ.6/

$$T_{3ef} = \ln 2 \frac{k(\lambda_{2ef} - \lambda_{1ef})}{k\,\lambda_{1ef}(\lambda_{2ef} - \lambda_{1ef}) + \lambda_2 \lambda_3} \qquad /EQ.7/$$

where T_{3ef} denotes effective half-life of polonium-210, and k is the ratio of $^{210}Po/^{210}Pb$.

If the effective half-lives of bismuth-210 and lead-210 in the liver are respectively 3.8 /ICRP, 1960/ and 719 /HOLTZMAN, 1960 A/ days, then the effective half-life of supported polonium-210 is 80 days. The latter is 2.5 times longer than the 32 days half-life

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x/ See below.

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tabulated in ICRP Report /1960/.

In TABLE 35 are given the values of biological and effective half-lives in the liver, of the members of ^{210}Pb-^{210}Bi-^{210}Po system, calculated from the exponential model of retention and from ingestion and concentration data /ICRP 1960 and HOLTZMAN 1960 A/ and also from experimental data on unsupported polonium /ICRP 1960/ and from the measured value of $^{210}Po/^{210}Pb$ ratio /JAWOROWSKI 1965 A/.

TABLE 35. Half-lives of ^{210}Pb, ^{210}Bi and ^{210}Po in the liver /in days/

Nuclide	Supported nuclide		Unsupported nuclide		Source of information
	T_b	T_{ef}	T_b	T_{ef}	
^{210}Pb	-	-	1947	1500	ICRP 1960
	—	—	800	719	HOLTZMAN 1960 A
^{210}Bi	-	-	15	3.8	ICRP 1960
^{210}Po	188	80	-	-	JAWOROWSKI 1965 A
	-	-	41	32	ICRP 1960

The measured and calculated values of $^{210}Po/^{210}Pb$ ratios in the bone and liver are compiled in TABLE 36.

The conclusion of the above consideration is that the numerous inconsistencies between the ICRP Report /1960/ data and the values determined in humans or animals are caused by neglecting in that

Report the metabolic differences between the supported and unsupported daughters of lead-210. The hypothesis of the present write that there exists discrimination between the lead-210 and polonium -210 entering the body from the natural environment, needs further investigations on ingestion, excretion and tissue distribution of these nuclides.

TABLE 36. Values of $^{210}Po/^{210}Pb$ ratios in the bone and liver

	Bone	Liver	Source of information
Measured	0.98 x/	0.49 x/	JAWOROWSKI 1965 A
	1.0	-	HOLTZMAN 1960 A
	1.18; 0.97	-	RAJEWSKY et al. 1963
	0.82; 0.42	3.1; 1.8	OSBORNE 1963
	1.09	2.7	MUTH et al. 1964
	-	1.06	BLANCHARD 1966
Calculated	1.01	-	JAWOROWSKI 1965 A
	0.1	0.18	ICRP 1960
	-	0.54 xx/	THIS WORK, T_{ef} as in ICRP 1960
	-	0.68 xx/	THIS WORK, T_{ef} as in HOLTZMAN /1960 A/ and JAWOROWSKI /1965 A/

x/ Measured in dogs; otherwise in man.

xx/ Calculated from exponential model of retention and f_w values as tabulated in ICRP Report /1960/; see text.

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6.3 RADIATION DOSE TO PROFESSIONALLY UNEXPOSED PEOPLE

The concentration of lead-210 and its daughters in the skeleton is about one order of magnitude higher than in other organs /TABLE 26/ and one can expect that in bone the radiation dose from this source will be higher than in other tissues.

The radiation dosimetry in bone is a difficult problem due to the complex structure of this tissue. Mineral salts, which contain probably the majority of lead-210 and its daughters compose an extremely heterogenous system with soft tissue. The microcavities in which the living soft tissue is imbedded may vary in shape and size from spheres, fractions of micron in diameter, to plane slabs or tubes, tens of microns thick. The amount of energy absorbed by soft tissue in bone will depend on the geometry of this tissue, on the patterns of microdistribution of radionuclides and on other physical parameters such as energy loss in both soft tissue and mineral bone. Mathematical methods enabling calculation of the radiation dose in different bone structures have been developed by SPIERS /1953/, KONONENKO /1957/, MAYS et al. /1958/, HINDMARSH et al. /1958/ and more recently by CHARLTON et al. /1962/.

Here, for calculation of the radiation dose from ^{210}Pb-^{210}Bi-^{210}Po system the expression developed by SPIERS /1953/ will be used:

$$D = N\delta R_T F / 2\varrho \left[erg^{-3}/day\right] \qquad /EQ.8/$$

where N denotes number of alfa part
per day, δ is average energy loss
tissue in ergs/micron, R_T is alfa
microns, ϱ is ratio of alfa partic
neral bone, R_T/R_B, and F denotes d
sumed, averaged for varying size a

The radiation dose absorbed b
chain depends mainly on the highly
lonium-210. The contribution from
bismuth-210 is only 0.75 per cent
particles and therefore will be ne

Value of N. The $^{210}Po/^{210}Pb$ r
equilibrium in the living bone is
Then, per one disintegration of le
one alfa particle of polonium-210
of interstitial bone lamellae is 2
-210 concentration in bone in 0.04
as a mean for Northern Hemisphere/
particle emmitted per cubic micron
x 10^{-10}.

Value of δ and R_T. If the av
lonium-210 is 5.3 Mev, then its av
range in soft tissue, R_T, extrapol
will be $\delta = 0.148 \times 10^{-6}$ ergs/mic

Value of F. The approximate v

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range, R_T, of 44 microns and different bone cavities varying in size from 0 to 50 microns have been calculated by CHARLTON et al. /1962/. The data and methods given by these authors were used here for calculation of F for polonium-210 alfa particle range of 39 microns. Results of this calculation are given in TABLE 37.

TABLE 37. Average values of geometric factor F for polonium-210 alfa particles of the soft tissue range of 39 microns

Tissue	Diameter of cavities or thickness of bone marrow layer /microns/	F
Soft tissue in:		
Canaliculi	0	1.0
Lacunae	5	0.95
Haversian	20	0.78
canals	50	0.48
Bone marrow	0 - 40	0.16

Value of ϱ. The range of alfa particle in a medium, R, can be calculated by the following expression /PRICE 1958/:

$$R \ /mg/cm^2/ = 0.56 \ R_A/cm/A^{1/3} \qquad /EQ.9/$$

where R_A is alfa particle range in the air /in 15°C and 760 mm Hg/ and A atomic mass of the medium. The range in air of 5.3 Mev alfa

particles can be deduced from equation given by PRICE /1958/:

$$R_A = 0.309 \ E^{3/2} \qquad /EQ.10/$$

as 3.77 cm. If the effective atomic number of bone is 13.8 /SPIERS 1946/, then the atomic mass of bone can be accepted as 27.6. Inserting the values for A and R_A in /EQ.9/ it can be calculated that 5.3 Mev alfa particle range in mineral bone, R_B, is 26.6 microns, if the density of bone is accepted as 2.4. Then R_T/R_B = = 1.45.

Inserting the values for N, δ, R_T, F and ϱ in /EQ.8/ and assuming the uniform distribution of lead-210 in the skeleton and the value of RBE for alfa particles as 10, the dose rate to the soft tissue enclosed in the bone cavities and to the bone marrow has been calculated, and the particular values obtained are given in TABLE 38.

TABLE 38. Radiation dose rates to soft tissue in the bone cavities and to the bone marrow from ^{210}Pb

Tissue	Dose rate in mrems per year
Soft tissue in:	
Canaliculi $\emptyset$ = 0 u	21.9
Lacunae $\emptyset$ 5 u	20.8
Haversian canals $\emptyset$ 20 u	17.0
$\emptyset$ 50 u	10.5
Bone marrow 0-40 u	3.5

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As compared with the dose rates from other natural radionuclides deposited in the skeleton i.e. radium-226 and thorium-228, the dose rates from the lead-210 chain are about four times higher /STAHLHOFEN 1964/. In soft tissues /other than contained in bone structures/ the dose rates from lead-210 chain are about 10 times lower than in the skeleton, whereas in the case of radium-226 the soft tissue rates are about 50 times lower /STAHLHOFEN 1964/. It appears then, that lead-210 with its daughters is the major contributor to the radiation burden of the body of all natural "bone - -seeking" radionuclides.

As compared with strontium-90 + itrium-90 dose rates the contribution from lead-210 chain is about 15 times higher /STAHLHOFEN 1964/.

7. TEMPORAL AND GEOGRAPHICAL DISTRIBUTION OF STABLE AND RADIOACTIVE LEAD IN ENVIRONMENT

7.1 TEMPORAL AND GEOGRAPHICAL DISTRIBUTION OF STABLE LEAD

Time distribution of stable lead

In chapters 5. and 6. it was pointed out that the existing levels of stable lead in the environment and in the human body are higher than the calculated natural levels. Some of the data on the existing and natural levels of stable lead, discussed in these chapters are compiled in TABLE 39.

TABLE 39. Existing and natural levels of stable lead.

Material	Existing /measured/	Natural /calculated/	Approximate difference
Air	0.1-6.0 $\mu g/m^3$	$5.43 \times 10^{-4} \mu g/m^3$	10^3-10^4 times
Surface water	0-55 $\mu g/l$	0.5 $\mu g/l$	0-100 times
Rainwater	0-1858 $\mu g/l$	-	-
Human food	2 $\mu g/g$ wet	0.01 $\mu g/g$ wet	200 times
Human body	66-750 mg	5.0 mg	10-100 times
Human bones	3.9-100.0 $\mu g/g$ dry x/	0.85 $\mu g/g$ dry x/	5-100 times

x/ Calculated from total body content assuming 70 per cent of it being in the skeleton, the mass of which is 7000 g, and from wet/dry bone ratio of 1.7.

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The increase of stable lead in the environment has been probably caused in recent decades by industrial revolution and the combustion of lead alkyls in gasoline /PATTERSON 1965/.

According to CHOW et al. /1962/ during the last century about $1x10^{14}$ g of lead have been dispersed by man over the land surfaces, of which about $3x10^{13}$ was in a form highly accessible to the wethering cycle. The latter is an amount equal to 30 per cent of all the lead in the oceans. Presently, about $2.3x10^{12}$ g of lead is dispersed yearly over land as a result of industrial activities.

The main sources of artificial lead in the air are the industrial production of lead,and coal and gasoline combustion. The two latter sources were discussed in detail in par.5.2. Between 1860 and 1947, the input of coal to the energy system of the World increased about ten fold /PUTNAM 1953/ and the introduction of lead alkyls into automotive gasoline was started about 1928. During the past twenty years about $2.6x10^{12}$ g of stable lead has been introduced into the air as combusted lead alkyls /CHOW et al. 1965/, and probably similar amounts were contributed from the burning of coal /par.5.2./.

Any increase of lead content in the air from these sources should be reflected in the precipitations, which wash-out the lead aerosols from the atmosphere. The fossil precipitations, preserved in glaciers as the annual ice layers, may be therefore a suitable medium for studying the temporal changes of lead content in the environment. For studying these changes in the human body, bones

may be a proper material, providing they were preserved in special conditions, in which the chemical alterations due to the action of percolating ground water were avoided.

The present writer collected glacier ice samples in two localities in Poland, down to a depth of about 1861 and also human bone samples covering the period from the III to XX Century. The results of his study are presented below.

Glacier ice

Ice samples were collected from a miniature corrie glacier in the Great Mięguszowiecki Cirque /GMC/ in the High Tatra Mountains, Poland, and from a glacier in the cave Grota Lodowa /GL/ in Western Tatra. In both localities determination of the age of the ice was based on the annual stratification of glaciers, with a possible error ranging from about $\pm$ 1 year in the upper-ice strata to about $\pm$ 2 years in the layers about 1925 and to $\pm$ 10 years in the layers below 1900. The thickness of ice layers in the GMC glacier from which the samples were collected ranged from 80 to 320 cm and in the GL glacier from 3 to 12 cm. The ice was collected uniformly from the total thickness of a particular strata with the exception of the GMC sample from 1965, which was collected only from the upper part of the stratum. Some of the GL samples covered two or three strata. From some interjacent GMC strata, samples were not collected. Several samples were also lost during analysis. From each sample, 4 liters of water were evaporated in "Pyrex" glass beakers and analysed according to the method described in APPENDIX I.

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The results of lead determinations in the ice samples are given in TABLES 40 and 41 and in FIGURE 4.

TABLE 40. Concentration of stable lead in the ice from Grota Lodowa /GL/ glacier.

Year	Lead concentration /µg per liter/	
	In ice samples	Mean for group of 5 samples
1	2	3
1965	78.0	
1964	77.7	
1963	40.6	80.0
1962	31.3	
1961	170.0	
1959	100.0	
1958	141.7	
1957	130.0	148.0
1956	215.0	
1955/54	162.5	
1953/52	76.5	
1951/50	122.5	
1949/47	13.2	83.7
1946/45	70.0	
1944/43	136.5	

Cont.tab.40

1	2	3
1942/41	87.0	
1940/39	56.2	
1938/37	13.2	69.3
1936/35	152.5	
1934/32	37.5	
1931/30	40.0	
1929/28	12.0	
1927/26	28.1	38.3
1925/24	54.4	
1923	57.3	

TABLE 41. Concentrations of stable lead in the ice from Great Mięguszowiecki Cirque /GMC/ glacier

Year	Lead concentration /µg per liter/	
	In ice samples	Mean for group of samples
1	2	3
1965	27.0	
1964	37.8	
1963	126.6	78.6
1962	168.4	
1960	33.3	

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Cont.tab.41.

1	2	3
1956	133.0	
1953	22.6	
1952	13.8	46.3
1951	2.0	
1950	60.3	
1949	38.3	
1948	38.8	
1947	62.5	47.0
1945	28.3	
1944	54.6	
1943	14.6	
1899	25.8	
1898	32.7	
1897	14.2	25.0
1896	36.3	
1895	15.6	
1894	15.1	
1893	11.4	
1892	1.0	10.2
1891	17.9	
1890	5.6	

Cont.tab.41.

1	2	3
188	7.3	
188	2.8	6.3
1871	8.7	
1866	3.1	
1865	4.3	5.0
1861	7.6	

As may be seen from TABLE 40 in the GL ice the mean lead concentrations, increased steadily during the last five decades being about twice as high in the seventh decade as in the third one. A similar increase may be seen in the GMC ice /TABLE 41/. In this set of samples the oldest samples were collected from the layers corresponding to about the seventh decade of nineteenth century, i.e. the period when, in this part of the European continent, the industrial revolution started. It may be seen that the mean lead concentration in the samples from this decade was about 15 times lower than in contemporary samples. The low lead concentrations in the samples from the years 1861/1866 are probably not the natural concentrations, as it may be anticipated that they could have been polluted by industrial activities in the not too distant Silesian centers.

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FIGURE 4. Concentrations of stable lead in the ice from Grota Lodowa /GL/ and Great Mięgusowiecki Cirque /GMC/ glaciers. Lines represent mean concentrations for a period covered.

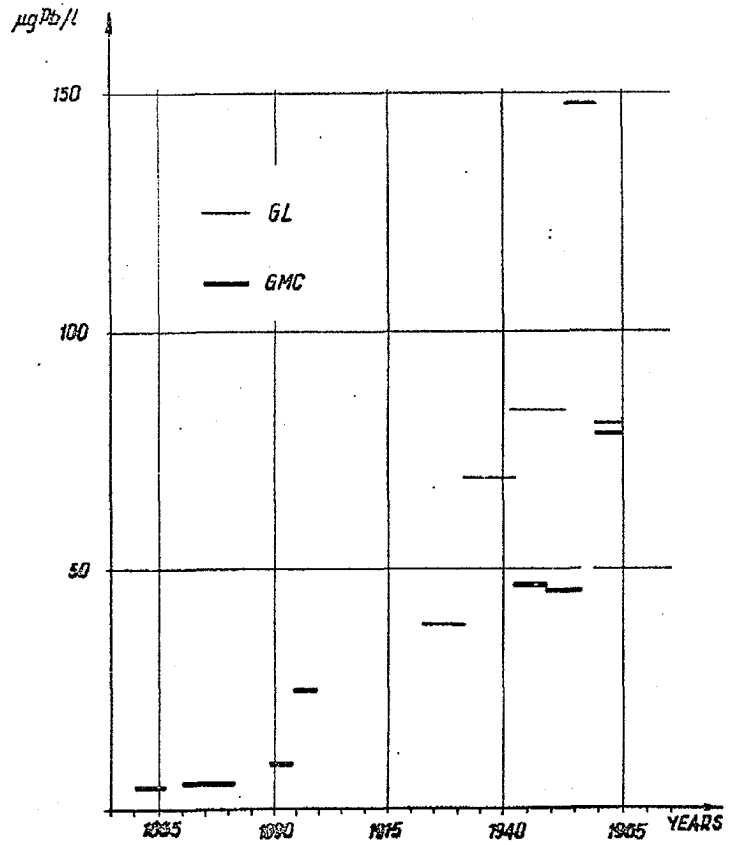
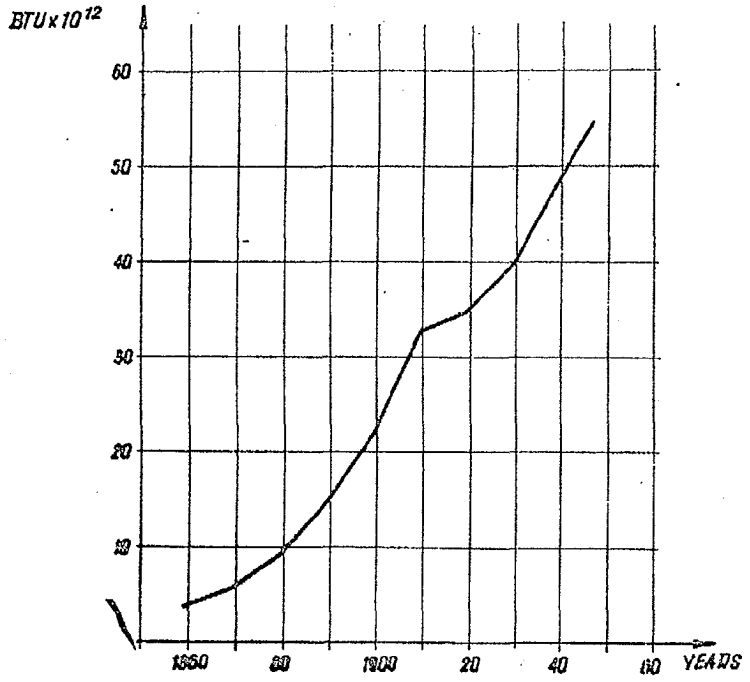


FIGURE 5. Total annual input to the World energy system from coal burning between 1860 and 1947 /after PUTNAM 1955/.



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These results may be compared with the data of MUROZUMI et al. /1965/ who found in Greenland snow sample from 1905 some 16 times lower stable lead content than in the sample from 1964. It is surprising that the rate of increase of lead concentration in the ice is similar to the rate of increase of the total annual input of coal burning to the energy system of the World /FIGURE 5/. This correlation may not necesserilly be directly connected with volatilising the lead by fuel combustion, but probably is connected with the increase in general industrial activity reflected by the rate of coal burning.

Human bones

The increase of lead concentration in the air, caused by human activities, is reflected, in the fossil precipitations, but not necessorily must be reflected in the human bones, because of the influence of other sources of lead, which may be more important in the contamination the human body than the atmospheric lead. For studying the possible temporal variation in the lead contamination of humans, the present author collected bone samples of the persons who had died in Poland in different times between about III Century to the present day. A majority of bone samples, exept the III Century and contemporary ones, were collected from wooden or stone coffins storred in dry underground crypts, usually on wooden shelves in four churches in Southern Poland. In two churches /Kościół Mariacki and Kościół O.Reformatów/ the bones were collected

from the naturally mumified bod
other soft tissues. The remains
ków/ from which the bones were
with few poorly preserved soft
in the Tyniec Monastery /No.6/
buried in the loess below the p
wever, had no direct contact wi
from this locality /No. 7, 8, 9
in the loess. The influence of
composition of the latter sampl
fore be excluded, although grav
which stands on the top of a hi
niec locality differs strictly
ristics of other localities whe
ground water can be excluded. T
in a cave near Kroczyce, provin
cave was accidentally opened by
group of skeletons was found, l
the hearth with preserved, parti
is located in limestone rock on
The age of the bones was determi
utensils found in the cave /KOW
The samples No. 1 and 2 were co
opening the entry to the cave,
Museum in Kraków. The other samp

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in 1967 and they could be contaminated by visitors.

Before analysis the bone samples were dried to the constant weight, then after wet ashing a stable lead was determined in them according to the method described in APPENDIX I. The results of lead determinations in these samples are given in TABLE 42.

TABLE 42. Concentration of stable lead in the human fossil bones

Number of skeleton and sample	Age	Locality	Lead concentration /µg/g dried bone/	Type of bone
1	2	3	4	5
1.	III century	Kroczyce	1.81	pubic
2.	III century	Kroczyce	1.84	scapula
3.	III century	Kroczyce	2.13	ulna
4.	III century	Kroczyce	2.55	humerus
5.	III century	Kroczyce	3.40	tibia
6.	XI century	Tyniec	92.5	temporal+frontal
7.a.	XIII century	Tyniec	16.0	vertebra
7.b.			26.07	temporal+frontal
7.c.			35.6	tibia
8.a.	XIII century	Tyniec	87.5	phalanx
8.b.			185.9	talus
8.c.			318.6	vertebra
9.a.	XIII century	Tyniec	19.6	mandibula
9.b.			24.8	temporal+frontal

Cont.tab.42.

1	2	3	4	5
10.	XIII century	Tyniec	8.52	tibia
11.a.	XIV century	Tyniec	5.6	temporal+frontal
11.b.			7.8	ulna
12.	XVII century	Tyniec	31.1	ulna
13.	1650	Bielany	199.3	femur
14.a.	1710	k.Reformatów	33.8	femur - cortical
14.b.			58.6	femur - trabecular
15.	1750	Bielany	22.7	femur
16.	1792	Bielany	15.4	temporal+frontal
17.	1797	Bielany	58.7	femur
18.	XVIII century	K.Mariacki	80.8	ulna
19.	XVIII century	K.Mariacki	46.1	phalanx
20.	XVIII century	K.Mariacki	64.4	ulna
21.	about 1800	Bielany	38.8	tibia
22.	early XIX c.	K.Mariacki	116.5	femur
23.a.	early XIX c.	K.Mariacki	25.9	vertebra
23.b.			31.6	femur
24.	1835	K.Reformatów	55.1	phalanx
25.a.	1838	K.Reformatów	59.2	talus
25.b.			91.9	phalanx
	Recent /mean in TABLE 21/	Poland	3.9 x/	vertebrae

x/ Calculated from wet weight data in TABLE 21.

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As may be seen in TABLE 40 the III century samples contained lead in concentrations much lower than the XI - XIX century bones. Especially low concentrations were found in the samples No. 1 and 2, collected immediately after discovering the cave in Kroczyce. These concentrations of 1.81 and 1.84 µg Pb per gram of dried bone respectively, are surprisingly near the value of 0.85 µg Pb per gram of dry bone calculated for natural concentration /TABLE 39/. The data presented in TABLE 42 indicate that in Poland the contamination of humans with lead started early in our history, probably coincidently with the introduction of metals into common usage. The unusually high concentration in the bones of an individual from XIII century /skeleton No.8/ could be due to post mortem contamination from the loess, but rather low concentrations found in other skeletons in the same locality, only 1 to 2 meters from the skeleton No. 8, indicate, however, that the high lead content in this skeleton was probably aquired during life. The other samples high in lead /No.13, 22, and 25.a. and b./ were preserved in the wooden coffins, in conditions in which contamination with ground water or by the coffins' content was excluded. The concentrations found in the bones from XI to XIX century are similar to the concentrations found in contemporary bones in Western Countries /TABLE 19/.

On the other hand the lead contents in the bones of contemporary Polish residents are lower than they were in the past, although the contamination of environment with lead, as reflected by the rainwater concentration of lead, increased during the last hundred

years. This indicates that the present contribution from the air is smaller than was the contribution from other sources of contamination in the past, the latter contribution being presently non existent in this country. These presently non existent sources were probably tin kitchen utensils, widely used in the past, which contained large proportions of lead /FRIEND 1951, HENSEL 1965/.

The prevalence of sources other than air contributing to the body burden of lead may be inferred from the results of the lead balance studies /TABLE 24/. So the high lead contents in the contemporary residents of the Western countries are probably caused not by the lead pollution of the biosphere, but more directly by contamination of food during its processing and preserving. Processed and preserved foods are consumed on a rather limited scale in Poland, which probably results in the low levels of lead in the body as observed by the present writer.

Geographical distribution of stable lead

There is a scarcity of data on the geographical distribution of lead in the environment besides these already related in par. 5.2. It can be seen from TABLE 5. that the mean lead concentrations in non-urban air are about 60 times lower than in urban air. Also the soils in the industrial areas are usually highly contaminated with lead as compared with virgin and other types of soils /TABLE 9/.

A study on the geographical distribution of lead in lichens

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has been carried out by the present writer in a set of samples collected along the 20th Meridian /East/, in different parts of Europe and in South Spitzbergen. Differences in the lead concentration factor in different families of lichens can be anticipated as a result of variations in their mass to surface ratios and also because of their physiological differences. In the present author's investigation, therefore, the members of only one family, Cladoniaceae, have been used. The results of this study are given in TABLE 43.

Table 43. Geographical distribution of stable lead concentrations in lichens, along the 20th Meridian /East/, during the year 1965.

Locality		Lead concentration /µg/g dried lichens/	Species
1		2	3
77°18'N	Spitsbergen	1.45	Cladonia mitis
	"	4.34	"
	"	2.10	"
59°50'N	Stockholm region	31.39	Cladonia silvatica
54°26'N	Mierzeja Wiślana	16.72	"
	"	23.07	"
53°35'N	Olsztynek	9.39	Cladonia mitis
53°11'N	Nidzica	15.07	"
53°06'N	Mława	7.50	"
52°37'N	Płońsk	16.80	"

Cont.tab.43

1		2	3
52°20'N	Kampinos	19.4	Cladonia silvatica
52°00'N	Pamiętna	16.89	Cladonia mitis
49°32'N	Rzeki	21.40	Cladonia rangiformis
49°12'N	Opalone	23.12	Cladonia silvatica
	"	26.50	"
49°00'N	Kralova Hola	22.37	"
48°33'N	Zemplin Mtns.	12.61	Cladonia rangiferina
	"	14.40	Cladonia furcata
48°00'N	Matra Mtns.	34.12	"
48°00'N	Bükk Mtns.	13.54	Cladonia rangiformis
46°50'N	Szakonyfalu	11.20	Cladonia furcata
46°50'N	"	13.21	Cladonia rangiformis
46°08'N	Mecsek Mtns.	24.51	Cladonia rangiferina
	"	10.76	Cladonia rangiformis
	"	15.98	Cladonia furcata
	"	12.31	Cladonia mitis
41°20'N	South Bulgaria	16.39	Cladonia rangiformis + rangiferina

As may be seen in the TABLE 43 the concentrations found in samples collected in Spitsbergen localities, remote from industrial centers, are about 10 times lower than the concentrations found in samples from Scandinavia and Middle Europe. This low concentration

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of lead found in Arctic vegetation corresponds to the low lead content the livers of Eskimos from Alaska as compared with those of residents of other regions /TABLE 44/. As may be seen from TABLE 44. the highest lead concentrations in liver have been found in great cities and the lowest in Arctic and developing African regions.

These findings supports the supposition, discussed in par. 5.2., that the civilisational factors increase the levels of stable lead both is the environment and in human body.

TABLE 44. Average concentrations of stable lead in the liver of residents of different geographical regions. /After SCHROEDER et al. 1961 and TIPTON et al. 1965/

Locality	Lead concentration /p.p.m./
Hongkong	213
Cairo	200
U.S. /7 cities/	150
Honolulu	150
Bejrut	125
Taipei /Taiwan/	125
Kyoto	105
Lambarene /Gabon/	104
Bern	100
Tokyo	94
/Nigeria/ Lagos	94
Vellore /India/	92
Manila	77
Bangkok	74
Bombay	74
Welkom /Orange Free State/	60
Delhi	55
Anchorage /Alaska	37
Kampala /Uganda/	25
Usumbura /Burundi/	20

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7.2. TEMPORAL AND GEOGRAPHICAL DISTRIBUTION OF RADIOACTIVE LEAD

In most investigations of lead-210 in natural concentrations, the data derived from the samples collected in various years have been interpreted as if the time variations of lead-210 concentrations in the biosphere were either non-existent or negligible. In a preliminary investigation on the time distribution of lead-210 in glacier ice, however, the present author found that in the years 1952-59 the concentration of lead-210 in ice had increased by about a factor of ten /JAWOROWSKI 1963 D/. These concentrations were in substantial agreement with the results of other authors for the concentration of lead-210 in rainwater over the same period /BURTON et al. 1960, Baranov et al. 1962, BLIFFORD et al. 1952/. Fluctuations in the concentration of lead-210 over a period of years have also been found in Antarctic snow /LAMBERT et al. 1965/.

Here will be briefed the previously published results of the present author's investigation of the temporal and geographical distribution of lead-210 in glacier ice, red deer antlers and lichens /JAWOROWSKI 1966/. It will be shown that the concentration of lead-210 in the biosphere does fluctuate with time and that this may be due to both natural and artificial causes. Such fluctuations could well be of importance when interpreting radioactivity in human and other biological systems.

Time distribution of radioactive lead.

The water samples and lichen material used in this study hav already been described in par.7.2. Twelve red-deer antlers were collected from three localities: one in north-western and two in north-eastern Poland. Lead-210 in antlers is deposited during the period of growth of this tissue, that is between the months of April and August. Each year the antlers are shed and new ones develop, therefore, the concentrations of lead-210 found in such ti sue correspond to the contamination of the pastures in the spring and summer of a particular year.

The concentration of lead-210 in water from the glacier in Grota Lodowa in the Tatra Mountains /GL/ was 0.32-6.6 pCi/l and that from the glacier in Great Mięguszowiecki Cirque /GMC/ 0.19-5. pCi/l. As can be seen from FIGURE 6 the temporal fluctuations in the concentration of lead-210 in water are similar in samples collected from the two localities, which were about 15 km apart. The only three exceptions occurred in 1952/53, 1962/63 and 1965. The first might have been caused by an error in the age determination of the samples. The other two were probably caused by a difference in the periods of ice accumulations, which in the GL glacier is between December and August and in the GMC glacier between Novembe and April. In the GL samples the concentrations of lead-210 were greatest in the years 1924/25, 1930/31, 1935/36, 1944, 1948, 1956, 1958/59 and 1962. In a smaller GMC group the concentrations were greatest in the years 1944, 1948, 1953, 1956, 1959, 1953 and 1965.

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These results can be compared with the maxima found by LAMBERT et al. /1965/ in Antarctic snow in the years 1925/27, 1937/40, 1943/45, 1949/51, 1955/56 and 1956/61. In GL and GMC samples the maxima in the concentration of lead-210 fluctuated with a periodicity of 3-5 years. A similar frequency of maxima can be observed in the set of samples from the Antarctic. The suggestion by LAMBERT et al. /1965/, that an 11 year cycle occurs in the concentration of lead-210 in the Antarctic snow seems to have been made by neglecting the smaller maxima lying in between the greater ones. In the European sets of samples, however, the small Antarctic maxima of 1943/45 and 1955/56 can be traced as the distinct 1944 and 1956 maxima found in both the GL and GMC groups. No correlation was found between the concentration of lead-210 in ice and the annual precipitation in the Tatra Mountains /for 1940-65/. This is illustrated by the lack of correlation between the concentration of lead-210 and the thickness of ice layers /FIGURE 6/.

The striking feature of the data presented in FIGURE 6 is that the two last maxima, for the years 1958/59 and 1962/63 are higher than the earlier ones. This agrees with the results of BARANOV et al. 1962, 1965/ who found concentrations of lead-210 in rainwater collected in the vicinity of Moscow similar in value to those in the GL and GMC samples for the corresponding years 1959-1963. The increase in lead-210 concentrations between the years 1952 and 1965 has been found also in red-deer antlers /FIGURE 7/ and lichens /TABLE 45/. It is interesting that the mean concentrations of lead-

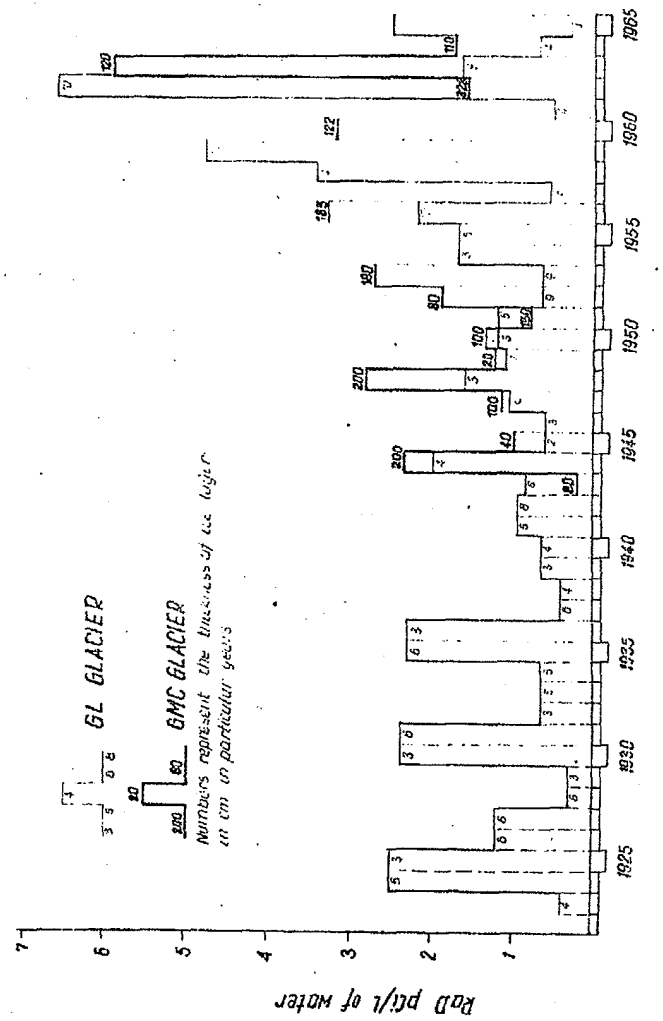


FIGURE 6. Concentration of lead-210 in glacier ice

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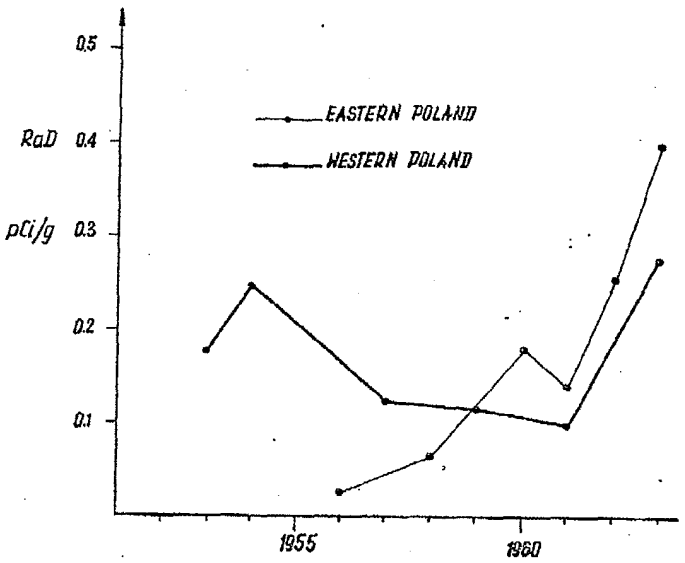


FIGURE 7. Lead-210 in red-deer antlers.

-210 in human bones also increased between the years 1952 and 1965 /TABLE 27/. An increase of lead-210 concentrations has also been observed in the stratospheric air during the year 1962 as compared with 1961. It can be seen from the Project Star Dust data /FEELY et al. 1965/ that the mean concentration of lead-210 in the stratospheric air, collected in corresponding time and geographical points, was higher and its age younger in 1962 than in 1961. For example the respective mean values for the periods November - December are 1.83 and 0.40 d.p.m. /1000 SCF, and 195 and 453 days. It is of interest that in air at ground level, the increase in the concentration of lead-210 was not observed in the samples, collected during 1961 and 1962 by PATTERSON et al. /1964/ but were observed by PEIRSON et al. /1966/. This discrepancy in the data for ground level air might have result from the short duration of the increased concentrations of lead-210 in the northern troposphere, according to the mechanism proposed by KOPCEWICZ /1964/ for middle latitudes, consisting of the transfer of radioactive dust from the stratosphere to ground level - first through the discontinuities of the tropopause in the cold front areas and then down in the troposphere by means of the intensive vertical turbulance in the cold fronts /II. type after Bergeron/. Because of the high velocity of these fronts the increases of radioactive dust concentrations in the ground level air are considered to be of very short duration. This supposition is in agreement with the results of BARANOV et al. /1965/ who found short-term variations by two orders of magnitude in the lead-210 concentration in rainwater.

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TABLE 45. Variations in time of lead-210 concentration in lichens

Year	Radium D pCi/g of dried lichen	Species	Locality
1957	1.4	Cladonia mitis	
1957	2.3	Cladonia rangiferina	
1958	8.0	Cladonia mitis	South
1965	6.3	Cladonia mitis	Spitzbergen
1965	10.4	Cladonia mitis	
1965	13.4	Cladonia mitis	
1957	1.7	Cladonia silvatica	
1958	2.6	Cladonia silvatica	
1958	4.7	Cladonia alpestris	Poland,
1959	4.5	Cladonia silvatica	Forestry
1960	1.9	Cladonia silvatica	Kampinos
1961	1.5	Cladonia silvatica	
1963	6.7	Cladonia alpestris	
1965	1.7	Cladonia silvatica	
1955	1.0	Cladonia mitis	
1958	1.4	Cladonia mitis	Poland,
1959	3.1	Cladonia mitis	Cracow-Wieluń
1963	2.6	Cladonia mitis	Highland
1964	3.2	Cladonia silvatica	

Geographical distribution of lead-

The results of measurements o
atmospheric air, published in rece
geographical distribution of this
level and at high altitudes. From
it may be seen, that in the sample
over an area ranging from Franz-Jo
concentration of lead-210 was rela
the highest values being found in
the lowest in the Antarctic and th
highest lead-210 concentrations in
the samples collected over the lan
latitudes, much lower being found
This suggeste that the ground-leve
the air are influenced by radon-22
however, not the case in the highe
ranges of 3 - 4.6 and 18 - 21 km R
uniform concentrations of lead-210
over the both hemispheres from 74°
of FEELY et al. /1965/ indicate th
regions from 90° N to 61° S these
uniformly.

According to PEIRSON et al. /
lead-210 concentration in rain is
air.

DUP050313892

TABLE 46. The concentrations of lead-210 in the ground level air

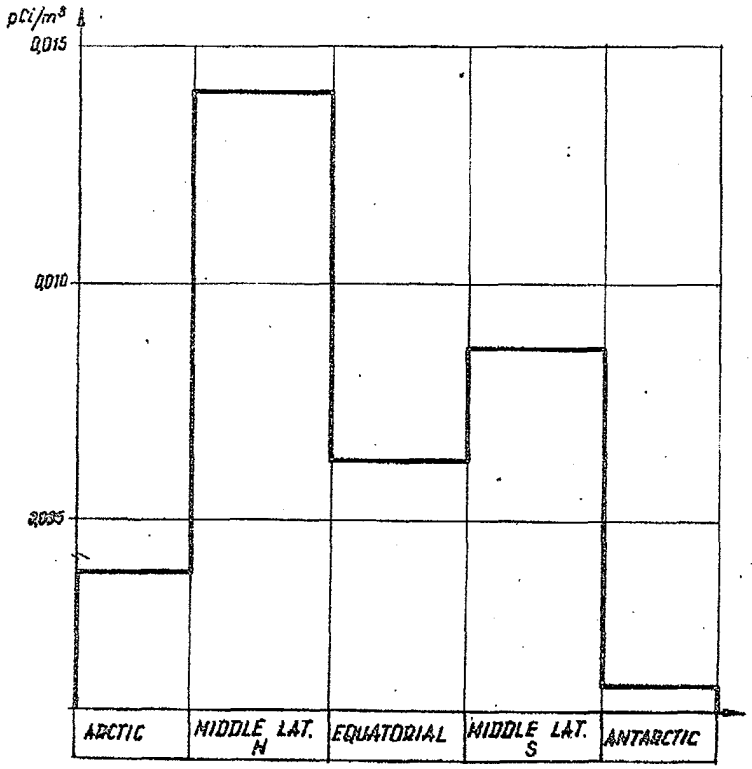
Lead-210 /pCi/m^3/			Source of information
Locality		Mean for the region	
1		2	3
Franz-Joseph Land, USSR	0.00006-0.002	0.0039 Arctica	VILENSKI et al.1965
Thule, Greenland	0.0011-0.012		PATTERSON Jr. et al. 1964
Moscow, USSR	0.0039-0.02	0.0140 Middle latitudes N	VILENSKI et al.1965
/USSR/	5.0		FEDOROV et al.1956
Chilton, UK	0.0027-0.007		PEIRSON et al.1966
Harwell, UK	0.0032		BURTON et al.1960
London, UK	0.0026		OSBORNE 1963 A
Heidelberg, Germany	0.01		HAXEL et al.1955
Freiburg, Germany	0.01		LEHMAN et al.1959
Warsaw, Poland	0.004		JAWOROWSKI 1963 C
Chicago, Ill., US	0.03		HOLTZMAN 1963 A
Winchester, Mass., US	0.01-0.041		SHLEIEN et al.1966
Washington, D.C., US	0.03		BLIFFORD et al.1952
Washington, D.C., US	0.005-0.027		LOCKHART et al.1958
Washington, D.C., US	0.0021-0.028		PATTERSON et al.1964
Yokosuka, Japan	0.013		LOCKHART et al.1958
Concinnati, Ohio, US	0.015-0.03		PATTERSON et al.1964
Miami, Flor. US	0.0022-0.0127		IBID.

1	
Pacific Ocean	0.0001-0
Mauna Loa, Havaii	0.0026-0
Indian Ocean	0.0005-0
Panama	0.0004-0
Lima, Peru	0.0008-0
Chacaltaya, Boliwia	0.0075-0
Antofagasta, Chile	0.0014-0
Santiago, Chile	0.0037-0
Little America V.Antarctica	0.00021-
South Pole	0.00014-

In TABLE 47 are compiled the r
concentration in the rainwater
ranging from North Norway to A
data in this TABLE is the diff
found in samples collected in
the low concentrations in samp
lities. Unusually high concent
tions were confined to the per
be discussed later on.

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FIGURE 8. Concentrations of lead-210 in ground-level air /mean values from TABLE 46/.



The geographical distribution of lead-210 was also studied by the present author /JAWOROWSKI 1966/ in lichen samples collected along the 20th Meridian /East/ ranging from 77°18´N to 41°20´N. The results of lead-210 determination in these samples are presented in TABLE 48. It is surprising that the concentrations of lead-210 found in the Arctic lichens /6.3 - 13.4 pCi/g/ were higher than those found in the temperate zone /0.6 - 3.5 pCi/g/. Recently, in Cladoniaceae lichens collected in Lapland HILL /1965/ has found high concentrations of lead-210 of 6.6 - 8.1 pCi/g, and HOLTZMANN /1963 B/ has found concentrations of 10.5 and 8.29 pCi/g in lichens from Finland and Alaska respectively. In other families of lichens from Alaska, HOLTZMANN has found an unusually high concentration of lead-210 of 61,2 pCi/g. It is also interesting that in Eskimo bones from the Canadian Arctic, collected about 1958, lead-210 concentrations were found to be more than twenty times higher than in bones from the Temperate Zone /HILL 1965/. In those Eskimo subjects who had been living in southern Canada for some time prior to the removal of the bone, i.e. prior to about 1958, the lead-210 concentrations were similar to the Temperate Zone levels /HILL 1965/. Due to the long effective half life of lead-210 in the bones some 20 to 30 years would be necessary to bring the high Eskimo bone concentrations to the temperate zone levels. Therefore it can be supposed that in the high concentration Eskimo subjects lead-210 had been accumulated in the time the low concentration subjects were not dwelling in the Arctic, i.e. about the year 1958.

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TABLE 47. Lead-210 concentration in rain water.

Location and year of collection	Lead-210 /pCi/l/	Source of information
1	2	3
Tromsø 70°N 1962	0.9-1.5	PEIRSON et al. 1966
Bodø 67°N 1961-1964	1.4-4.6	IBID.
Kodiak 58°N 1950	0.77	KING et al.1956
Moscow 56°N ? 1959-1963	1.2-300	BARANOV et al.1962 BARANOV et al.1965
Świerk 52°N 1963-1965	0.41-16.2	JAWOROWSKI - THIS WORK
Harwell 52°N 1956	0.63-4.2	BURTON et al.1960
Milford Haven 51°N 1961-1964	1.7-6.1	PEIRSON et al.1966
Tatra Mountains 49°N 1950-1965	0.32-23.1	JAWOROWSKI 1963 D JAWOROWSKI 1966
Linz 48°N 1961-1963	2.5-3.8	PEIRSON et al.1966
Cosausee 48°N 1961	2.8-2.9	IBID.
Schauinsland 48°N 1957	3.1	LEHMAN et al.1959
Ottawa 45°N 1961-1964	3.4-9.7	PEIRSON et al.1966
Glenview Ill. 42°N 1950	2.5	KING et al.1956
Washington D.C. 39°N 1950	0.24-8.2	IBID.

Cont.tab.47

1	2	3
La Jolla 33°N 1960	3.2	Rama et al.1961
San Diego 32°N 1960	0.85	IBID.
Nassau 25°N 1963-1964	1.4-3.1	PEIRSON et al.1966
Havai 21°N 1950	1.1	KING et al.1956
Philippine Is. 14°N 1950	1.1	IBID.
Panama 9°N 1950-1951	0.31	IBID.
Lagos 6°N 1961	4.5-4.8	PEIRSON et al.1966
Singapore 01°N 1961-1964	1.0-2.1	IBID.
Samoa 14°S 1950	0.23	KING et al.1956
Melbourne 38°S 1963-1964	2.0-4.0	PEIRSON et al.1966
Ohakea 40°S 1961-1962	0.5-0.8	IBID.
Argentine Is. 65°S 1964	0.2	IBID.
Antarctic 66°S /?/ 1950-1963	0.61-0.75	LAMBERT et al.1965
Base Roi Baudovin 70°S 1958-1963	0.75	CROZAZ et al.1964

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TABLE 48. Geographical distribution of lead-210 concentration in lichens, during the year 1965 along the 20th Meridian /East/

Locality	Radium D pCi/g of dried lichens	Species
1	2	3
77°18´N Spitzbergen	6.3	Cladonia mitis
"	10.4	Cladonia mitis
"	13.4	Cladonia mitis
54°26´N Mierzeja Wiślana	2.3	Cladonia silvatica
53°35´N Olszwynek	0.9	Cladonia mitis
53°11´N Nidzica	1.8	Cladonia mitis
53°06´N Mława	0.6	Cladonia mitis
52°37´N Płońsk	1.5	Cladonia mitis
52°20´N Kampinos	1.7	Cladonia silvatica
52°00´N Pamiętna	1.6	Cladonia mitis
49°32´N Rzeki	0.8	Cladonia rangiformis
49°12´N Tatra Mts.	2.5	Cladonia silvatica
49°00´N Kralova Hola	2.7	Cladonia silvatica
48°33´N Zemplin Mts.	2.9	Cladonia rangiferina
48°33´N " "	1.83	Cladonia furcata
48°00´N Matra Mts.	0.9	Cladonia furcata
48°00´N Bükk Mts.	1.2	Cladonia rangiformis
46°50´N Szakonyfalu	0.6	Cladonia furcata
46°50´N "	1.4	Cladonia rangiformis

Cont.tab.48

1	2	3
46°08´N Mecsek Mts.	3.2	Cladonia rangiferina
" "	1.4	Cladonia rangiformis
" "	2.2	Cladonia furcata
" "	2.7	Cladonia mitis
41°20´N South Bulgaria	3.5	Cladonia rangiformis + " rangiferina

This is supported by the low-level lead-210 concentrations found in Arctic lichens collected in 1957, which did not differ substantially from those found in European lichens from the corresponding period /TABLE 45/. HILL /1965/ and HOLTZMANN /1963 B/ explained the high concentrations of lead-210 in the Eskimo bones by the dietary habits of Eskimos, who consume the meat of lichen-eating animals. This explanation was given with the obvious assumption that the lead-210 concentrations in the lichens were the same in both the Arctic and temperate zone and also that the lead-210 concentrations in lichens were higher than in other cattle fodder. The former, however, is not the case as can be seen from TABLE 48. and also the levels of lead-210 in U.K. grass of 0.4-16. pCi/g /HILL 1965/ are similar to, if not higher than, the levels found by the present writer in European lichens. This suggests that the high lead-210 concentrations in Eskimo bones resulted not solely from dietary habits, but also from the high concentration of this nuclide in the Arctic biosphere.

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Sources of lead-210.

As has been pointed out in par.5.2. the natural source of lead-210 in the atmosphere is radon-222, a gas exhaled by land, the mean exhalation rate of which over land is about 4×10^{-17} Ci/cm^{2}sec., which corresponds to the global production rate of 0.62 MCi of lead-210 per annum. If this amount of lead-210 produced during one year were rained and washed out from the atmosphere with the total volume of water which is precipitated yearly on the surface of the Earth, that is, with 4.0×10^5 km^3 KOSTIN et al.1957 then the mean global concentration of lead-210 in rainwater should be 1.5 pCi. It is interesting that the mean lead-210 level found in the GL samples from before 1958 of 1.55 pCi/l is closer to the theoretical global concentration than to the value of 2.0 pCi/l calculated for the northern hemisphere only.

As has been demonstrated, the levels of lead-210 in the biosphere increased in the years 1958/59 and 1962/63. The combustion lead-210 introduced into the atmosphere with tetraethyl lead may be considered as a cause of these increases. The total amount of lead-210 which together with 2.6×10^{12} grams of stable lead was introduced into the air during the last 20 years /CHOW et al.1965/, is some 31 Ci, assuming that one gram of stable lead contains 12 pCi of lead-210 /WELLER 1965/, and can be ignored as compared with 0.62 MCi of lead-210, introduced yearly into the atmosphere by radon-222. The local effects of industrial centers in Europe should be

excluded as the study on the latitudinal distribution of the lead-210 concentration in lichen samples revealed that in the Arctic areas remote from industrial centers the lead-210 concentrations were higher than in Europe /TABLE 48/.

It is possible that the 1958/59 and 1962/63 lead-210 maximums are partially caused by the nuclear explosions, probably those performed in Arctic regions. It was already suggested by the present writer /JAWOROWSKI 1963 D/ that the 1958/59 maximum could have been caused by the attachment of the lead-210, which is naturally present in the atmosphere, to the particulate material injected into the stratosphere by nuclear explosions, which subsequently falls to the surface of the Earth. However, the higher lead-210 concentrations and younger age of lead-210 found in the air in 1962 as compared with 1961 /FEELY et al.1965/ indicate that artificial lead-210 has been introduced into the atmosphere. This is in agreement with the suggestion of STEBBINS /1961/ and PEIRSON et al. /1966/ that lead-210 in the atmosphere may have originated in part from nuclear test explosions. PEIRSON et al. suggest that the artificial lead-210 was formed by the nuclear reaction ^{208}Pb /2n, γ/ ^{210}Pb. According to these authors, some 30-50 % of 1962 lead-210 may be of that origin. The difference found in the GL group of samples between the 1962 maximum and that from before 1958 indicates that about 60 % of 1962 lead-210 was probably produced by nuclear explosions. In 1959, the artificial contribution was about 45 %. If the natural production of lead-210 is about 0.6 MCi per year, then

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it can be deduced that the artificial contribution in 1962 was about 0.9 MCi and in 1959 about 0.5 MCi. It seems that before 1958 the nuclear test contribution to lead-210 fallout was minute if any. It seems also that the artificial increase in lead-210 concentration was more pronounced in the Arctic regions than in the temperate or the southern ones.

APPENDIX I.

METHOD OF DETERMINATION OF STABLE LEAD

Reagents

All reagents must be lead-free, either as purchased or by special preparation. Double distilled water is used throughout the analysis.

Hydrochloric acid: /a/ conc.sp.gr. 1.18, /b/ 5 M.

Nitric acid: /a/ conc.sp.gr. 1.5, /b/ 1 per cent v.v. in water

Perchloric acid: sp.gr. 1.54.

Sulphuric acid: diluted 1:1 .

Ammonium hydroxide: sp.gr. 0.880.

Ammonium citrate solution: a 25 per cent w.v. solution in water.

Potassium cyanide solution: a 10 per cent w.v. solution in water. This solution should be at least 2 days old, so that traces of sulfide may become oxidized. Store in a refrigerator in black coloured bottle.

Hydroxylamine hydrochloride solution: a 20 per cent w.v. solution in water.

Chloroform: shake 250 ml of chloroform with 25 ml of water containing 1 ml of 10 per cent w.v. potassium cyanide solution and about 20 drops of 5 M ammonium hydroxide, separate and reject the aqueous layer, wash the chloroform with water and filter.

Dithizone: Stock solution - a 0.1 per cent w.v. solution of dithizone in chloroform. Filter and store in a refrigerator in

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black coloured bottle.

Working solution - shake 6 ml of dithizone stock solution with 9 ml of water and 1 ml of 5 M ammonium hydroxide. Separate and reject the lower layer and spin the aqueous layer in a centrifuge until clear. Prepare this solution freshly on the day of use.

Ammoniacal sulfite-cyanide solution: mix 340 ml of ammonium hydroxide, sp.gr. 0.880, 75 ml of 2 per cent w.v. sodium sulphite, Na_2SO_3, solution, 30 ml of 10 per cent w.v. potassium cyanide solution and 605 ml of water /the concentrations of these reagents are critical/.

Sodium hexametaphosphate, /$Na\ PO_3/_6$, : a 10 per cent w.v. solution in water.

Standard lead solution: /a/ dissolve 1.60 g of lead nitrate in water, add 10 ml of concentrated nitric acid, and dilute to 1 litre. /b/ Dilute 1 volume of /a/ to 100 volumes with water. Prepare solution /b/ sreshly as required. 1 ml = 10 µg of lead.

Thymol blue indicator solution: 0.04 per cent w.v. - warm 0.1 g of thymol blue with 4.3 ml of 0.05N sodium hydroxide and 5 ml of 90 per cent ethanol: when solution is complete, add sufficient 20 per cent ethanol to the final volume of 250 ml.

The following additional reagents are required when diethylammonium diethyldithiocarbamate extraction is used:

Sodium iodide solution: a 20 per cent w.v. solution in water.

Sodium metabisulphite solution a 1.25 per cent w.v. solution in water. Prepare this solution freshly as required, and filter

before use.

Diethylammonium diethyldi
/carbamate reagent/: dissolve
100 ml of purified chloroform,
tle. This solution is not stab
week.

Methyl red indicator solu
of methyl red with 0.95 ml of
90 per cent ethanol; when solu
per cent ethanol to produce 25

Pyrex glass must be used

Wet ashing of samples containi

1. To about a 20 g sample in
centrated nitric acid, cover w
till dissolution is complete.
acid. /The amount of acids use
sidue should be measured for c
Evaporate on a hot plate avoid
add 5 ml of concentrated nitri
repeat with a mixture of 5 ml
concentrated perchloric acid u
white.

2. Dissolve the ash in 5 ml
acid and boil gently for 5 min

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just alkaline, then just acidify with concentrated hydrochloric acid, boil and filter if any insoluble matter is present. Transfer solution to a 100 ml flask.

Separation of lead - Method A: in samples with low content of calcium, magnesium and phosphate.

3. Cool the solution, and add 5 ml of ammonium citrate solution and 10 ml of sodium hexametaphosphate solution. /For certain samples the high calcium and phosphate content may be held up by the addition of 10 ml of ammonium citrate solution, Otherwise method B must be used/.

4. Add a few drops of thymol blue indicator solution and sufficient ammonium hydroxide to give a blue-green colour indicating pH 9.0 to 9.5.

5. Cool, add 1 ml of potassium cyanide solution and, if much iron is present, add 1 ml of hydroxylamine hydrochloride solution.

6. Transfer the solution to a 100 ml separating funnel containing 10 ml of chloroform, and rinse with a few milliliters of water. The volume of the aqueous layer at this stage should be approximately 50 ml.

7. Add 0.5 ml of dithizone stock solution, shake vigorously for 1 minute and allow to separate. If the lower layer is red, add dithizone working solution until, after shaking, a purple, blue colour is obtained.

8. Run the chloroform layer into a second separating funnel and wash through with 1 or 2 ml of

9. Add to the liquid in the f
chloroform and 0.2 ml of dithiz
rously for 30 seconds, allow th
add it to the main chloroform e
tract should be green. If not,
and dithizone must be made unti
tract indicates that all the le

10. Reject the aqueous layer.

11. Add 10 ml of dilute nitric
tracts and shake vigorously for
reject the chloroform layer as

12. Wash the aqueous layer 6 t
form /to eliminate interference
and then adjust to pH 1.

Separation of lead - Method B:
calcium, magnesium and phosphat

3. Adjust pH to approximately
hydroxide, then add concentrate
tion is again clear and boil fo

4. Cool the solution to 50°-
lution and reduce any liberated
sufite solution.

5. Cool the solution, transfe
just the volume in order to bri

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respect to hydrochloric acid.

6. Add 10 ml of carbamate reagent by pipette, shake the funnel vigorously for 30 seconds. Allow the layers to separate and transfer the chloroform layer to a 100 ml flask.

7. Wash the aqueous layer twice with small amounts of chloroform without mixing and add these washings to the flask.

8. Repeat extraction with 10 ml of carbamate reagent twice and add the second and third extracts to the main extract.

9. Reject the aqueous layer.

10. To the combined extracts add 2.0 ml of diluted sulphuric acid and evaporate the chloroform.

11. Add 0.5 ml of perchloric acid to the residual solution and heat until fumes are evolved and the fuming solution is clear and colourless.

12. Cool the solution and proceed to step 3 under SEPARATION OF LEAD - Method A.

Removal of stable bismuth

13. Add 1 ml of 1 per cent cupferron solution in water, mix and add 5 ml of chloroform; shake well for 1 minute.

14. Transfer the chloroform layer to another funnel and repeat extraction 3 times. Finally wash the aqueous phase twice with chloroform. The chloroform phase now contains any bismuth present while the lead remains in the water phase.

Determination of stable lead

15. To the water layer left in the separating funnel add 30 ml o ammoniacal sulfite-cyanide solution, exactly 10 ml of chloroform and 0.5 ml of dithizone working solution, shake vigorously for 1 minute and allow to settle.

16. Run off a little of the chloroform layer into a second funn Insert a plug of cotton-wool into the dry stem of the funnel and after transferring the first runnings to the second funnel, fill spectrophotometer cell with the chloroform solution.

17. Measure the optical density of the test /and a blank/ soluti against chloroform at 5200 A with a spectrophotometer.

18. Read the number of micrograms of lead equivalent to the observed optical densities of the test and blank solutions from a previously prepared calibration graph. Prepare the calibration graph as follows: Measure 0, 1.0, 2.0, 3.0 and 4.0 ml of standard lead solution /b/ into separating funnels and dilute each to 10 m with dilute nitric acid. Proceed as described under "Determinatio of stable lead" step 15. through 17. Construct a graph relating the optical densities to the number of micrograms of lead.

The average recovery of stable lead by this method as applie in biological samples was found to be 96.4 ± 4 per cent.

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APPENDIX II.

METHOD OF DETERMINATION OF RADIOACTIVE LEAD

A. Separation of Bismuth-210 by cupferron extraction

The following additional /see APPENDIX I./ reagents are required for separation of bismuth with cupferron:

Cupferron 1 per cent solution in water freshly made up.

Bismuth nitrate carrier solution /A/: 6.0 mg of $Bi/NO_3/_3 \cdot H_2O$ dissolved in 10 ml of concentrated nitric acid and diluted with 500 ml of water /1 ml = 10 µg Bi/.

Sodium di-hydrogen phosphate: 5 M solution in water.

Polyvinyl acetate /PVA/: 6 per cent solution in methyl alcohol.

1. To the collected chloroform solutions of lead dithizonates from step 16. of the "Determination of stable lead" APPENDIX I. add 10 ml of dilute nitric acid and shake for 1 minute. Reject the chloroform layer.
2. To the aqueous phase add 2 ml of bismuth nitrate carrier solution /i.e. 20 µg of Bi/.
3. Store the solution for approximately 15 days to allow a build up of bismuth-210.
4. Extract the bismuth as in step 13 and 14 of the "Removal of stable bismuth" APPENDIX I.
5. To the chloroform layers from step 4. add 0.2 ml of 50 per cent sulphuric acid and evaporate the chloroform.
6. Add 0.5 ml of concentrated perchloric acid and boil on a hot

plate in a 50 ml beaker covered with a watch glass until the perchloric acid vapours cease to evolve; the solution should be colourless.

7. Cool and rinse watch glass with a jet of water, collect washings in a beaker and add 10 ml of water.
8. Add 2 ml of bismuth nitrate solution /B/ then an excess of 5 M sodium di-hydrogen phosphate solution and adjust the pH to approximately 7.
9. Centrifuge the bismuth phosphate onto a counting planchette x/ held in a demountable centrifuge tube. Decant and wash the precipitate with methyl alcohol, centrifuge and decant.
10. Add 0.05 ml of PVA solution to the planchette and allow to dry slowly. This helps to reduce cracking.
11. Measure the beta activity of the sample in a low-background counter at intervals of a few days and estimate the half-life. Calculate the bismuth-210 equilibrium activity using the following formula

$$N_0 = \frac{N}{\left(1 - e^{-\frac{0.693\ t}{T}}\right)} \qquad /EQ.11/$$

where N_0 = pCi of lead-210; N = pCi of bismuth-210; t = time of bismuth-210 build up in days; T = bismuth-210 half-life in days.

x/ Planchette consist of a perspex ring with a 1.3 mg/cm^2 aluminium foil bottom.

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The recovery of bismuth-210 in this method was found to be 98.9 ± 1.0 per cent.

B. Separation of bismuth-210 by internal electrolysis.

The following additional reagents are required when electrolytic separation of bismuth is used:

Stock electrolyte solution: 40 ml of concentrated nitric acid and 6 g of tartaric acid per 1000 ml of water.

Bismuth nitrate solution /B/: 12.1 g of $Bi/NO_3/_3 \cdot H_2O$ dissolved in 10 ml of concentrated nitric acid and 500 ml of water /1 ml = 20 mg Bi/.

"Aged" glass should be used for electrolysis. Before using the beaker, sintered glass pipe and the stirring rod should be stored for three days in a saturated solution of bismuth and lead nitrates. Glass, so prepared, can be used repeatedly if concentrated acids are not used for cleaning.

Preceed according to APPENDIX I, SEPARATION OF LEAD - Method A, step 10 inclusively and then

1. To the chloroform extracts containing dithizonates of lead and bismuth add 25 ml of stock electrolyte solution. Shake vigorously for 1 minute, allow to separate and reject the chloroform layer as completely as possible.
2. Wash the aqueous layer three times with 5 ml portions of chloroform.
3. Add one drop of carrier bismuth solution /B/, 10 ml of concentrated nitric acid and 500 ml of distilled water /this step should be ommitted when a carrierfree bismuth-210 is to be prepare

4. Store the electrolyte solution for approximately 15 days to allow build up of bismuth-210.
5. Transfer the electrolyte solution from step 4. to a 50 ml beaker and fill the glass pipe with stock electrolyte solution so that the levels in the pipe and in the beaker were equal.
6. Mount the platinum and lead elelctrodes as in FIGURE 8 and mi strongly for 60 minutes in the water bath at 70°C.
7. Remove from the solution both electrodes without disconnectin them, wash gently with distilled water and dry at room temperature
8. Disconnect the platinum electrode and cover it with 1.3 mg/cm aluminium foil and count bismuth-210 beta activity of both sides o the electrode in a low-background counting unit. Calculate the lead-210 activity as in Separation of bismuth-210 by cupferron extraction, step 11.

Cleaning of the platinum elektrode

9. Connect the platinum electrode with the positive pole of a 6 V current source and the negative one with another electrode /Cu o Ni/ and immerse both electrodes in 1:2 diluted nitric acid for 1 minute.
10. Wash in distilled water and clean with steel wool.
11. Heat to 400°C for 30 minutes.
12. Cool and boil in distilled water for 1 hour.

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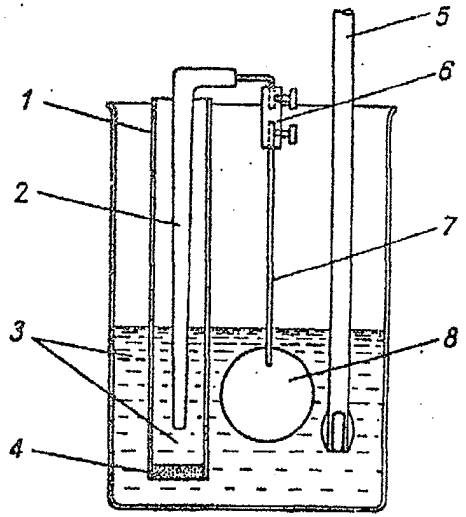


FIGURA 9. The assembly for deposition of bismuth-210 on platinum foil: 1 - glass pipe; 2 - Pb anode; 3 - electrolyte solution; 4 - sintered glass; 5 - glass stirring rod; 6 - screw link; 7 - Pt wire; 8 - Pt cathode.

LITERATURE

ALBERIGI-QUARANTA A., B.RIGHINI, V.PROD and O.RIMONDI: Nucl. Instr. Meth. 14:13, 1961.

ALIAVDIN R. and J.PEREGOODE: J. Ind. Hyg. 18: 139, 1936.

AMIRKHANOFF K.I., S.B.BRANDT and E.N.BARNITSKY: Ann. N.York Acad. Sci. 91: 235, 1961.

AMPRINO R.: Experimentia 8: 380, 1952.

ANDERSON E.C., P.N.DEAN and U.W.Rowe: LA-3132-MS p.145, 1964.

ANONYMOUS: Advisory Committee on Tetraethyl Lead to Suregon General of the Public Health Service. Public Health Aspects of Increasing Tetraethyl Lead content in Motor Fuel. Public Health Service. Publ. No. 712. Washington 1959.

ANONYMOUS: Bericht der Eidg. Bleinbenzin Kommissionen und den Bundesrat ueber ihre Taetigkeit in Zeitraum 1947-1960. Mitt. Lebensmitt. Hyg. 52: 135-243, 1961.

ANONYMOUS: US Public Health Service. Drinking Water Standards. Fed. Register 27: 2152, 1962.

ANONYMOUS: Bureau of the Census: Statistical Abstracts of the United States. US Dept. of Commerce, Bureau of the Census. US Government Printing Office, 1963 A.

ANONYMOUS: "Mały Rocznik Statystyczny" 1963. Główny Urząd Statystyczny. Warszawa, 1963 B.

AUB J.C., L.T.FAIRHALL, A.S.MINOT and P.REZNIKOFF: "Lead Poisoning". Williams and Wilkins Company. Baltimore 1926.

DUP050313904

BAGHI K.N., H.D.GANGULY and J.N.SUDOR: Indian J.Med. Res. 28: 441, 1940.

BARANOV V.I. and V.D. VILENSKII: Radiochimia 4: 493, 1962.

- Atomnaya Energiya 18: 503, 1965.

BEHRENS B.: Arch. Exp. Path. Pharm. 109: 332, 1925.

BEHRENS B. and H.TAEGER: Z. Ges. Exp. Med. 96: 282, 1935.

BENDER H.: Gerlands. Beitr. Geophys. 41: 401, 1934.

BERG J.M. and M.ZAPELLA: J. Mental Deficiency Res. 8: Pt.1.44.1964.

BLACK R.: Bull. Geol. Soc. Amer. 65: 839, 1954.

BLACK S.C.: H.Phys. 7: 87, 1961.

BLANCHARD R.L.: Relationship between ^{210}Po and ^{210}Pb an man and his environment. Symposium on Radioecological Concentration Process. Stockholm, 25 - 29 April, 1966.

BLIFFORD I.H., L.B.LOCKHART Jr. and H.B.ROSENSTOCK: J. Geophys. Res. 57: 499, 1952.

BODE H. and G.HENRICH: Z. anal. Chem. 135: 98, 1952.

BOSLAVSKAYA T.N., A.G.GRAMMAKOV, A.P.KIRIKOV and P.N.TVERSKOY: Vess. Geol.-Razved. Ob'yedineniye Izv. 51: 1283, 1932.

BRADLEY J.E., A.E.POWELL, W.NIERMANN, K.R. MC GRADY and E.KAPLAN: J. Pediatr. 49: 1. 1956.

BRYANT F.J. and J.F.LOUTIT: AERE - R - 3718, 1961.

BURTON W.M. and N.G.STEWART: Nature, 186: 584, 1960.

CALHOUN J.A., R. MC LEAN, J.C.HUDSON and J.C.AUB: Arch. Industr. Hyg. 9: 9, 1954.

CANNON H.L. and J.M.BOWLES: Science 137: 765, 1962.

CANTAROV A. and M.TRUMPER: "Lead Poisoning". Williams and Wilkins Company. Baltimore. 1944.

CASTELLINO N. S.ALOJ: Brit. J. Industr. Med. 21: 308, 1964.

- IBID. 22: 172, 1965.

CHARLTON D.E. and D.M.CORMACK: Rad. Res. 17: 34, 1962.

CHEN P., A.R.TEREPKA and H.H.HODGE: Ann. Rev. Pharm. 1: 369, 1961.

CHOLAK J.: Arch. Environ. Health. 8: 222, 1964 A.

- IBID. 8: 314, 1964 B.

CHOLAK J. and K.HAMBACH: J. Ind. Hyg. Toxicol. 25: 47, 1943.

CHOLAK J., L.J.SCHAFER and T.D.STERLING: J. Air. Pollut. Contr. Ass. 11: 281, 1961.

CHOW T.J. and M.S.JOHNSTON: Science 147: 502, 1965.

CHOW T.J. and C.C.PATTERSON: Geochim. Cosmochim. Acta 26: 262, 1962.

CROZAZ G. and E.PICCIOTO and W. DE BRENCK: J. Geophys. Res. 69: 2597, 1964.

DELLA ROSA R.J., R.G.THOMAS and J.N.STANNARD: UR - 392, 1953.

DI FERRANTE E.R.: ANL - 6398, p. 71, 1961.

DINISCHIOTU G.T., B.NESTORESCU, I.C.RADULESCU, C.IONESCU, N.PREDA and G.ILUTZA: Brit. J. Industr. Med. 17: 141, 1960.

DURUM W.H. and J.HAPTY: Implications of the minor element content of some major streems of the world. Symposium on Geochemical Evolution - The First Five Billion Years. Am. Ass. for the Advancement of Science. Denver, Dec, 29. 1961.

DURUM W.H. and S.G.HEIDEL: IASH Commission of Surface Waters. Publication No.51, p.518, 1960.

DUP050313905

EGLI R. et al.: Schweiz. Med. Wschr. 87: 1171, 1957. - fide PATTERSON 1965.

FEDOROV E.K. and B.J.BARANOV: Sodezhoniye prirodnikh radioactivnikh veshestv v atmosfere i v vodakh v predelakh territorii SSSR. Moskva 1956. Document OOH, A/AC.82/G/R.39, - fide UN Document A/5216, 1962.

FEELY H.W., J.P.FRIEND, P.W.KREY and B.A.RUSSEL: HASL-153, 1965.

FINK R.M.: "Biological Studies with Polonium, Radium and Plutonium". McGrow-Hill Book Comp. Inc., N.York, 1950.

FISCHER E.: Ann. 190: 118, 1878.

FISCHER H.: Wiss. Veröffentl. Siemens-Konzern 4: 158, 1925.

FRIEND J.N.: "Man and the Chemical Elements". Charles Griffin and Co.Ltd. London, 1951.

FROST H.M.: Henry Ford Hosp. Bull. 8: 87, 1961.

GAŁKIEWICZ T.: Przegl. Geolog. 5: 7, 1957.

- Rudy Met. Nieżel. 1: 1, 1956.

- IBID. 4: 2, 1959.

GILFILLAN S.C.: J. Occup. Med. 1965.

GITELMAN H.J. and W.F.NEULAN: "Lead - Hydroxy Apatite Interaction". The University of Rochester Atomic Energy Project. Rochester, N.York 22.6. 1959. - fide WEINIG et al. 1967.

GORSUCH T.T.: Analyst 84: 135, 1959.

GUSEV M.I.: Limits of Allowable Lead Concentration in the Air of Inhabited Localities. Ed. V.A.Ryazanov, 1960. Translation distributed by US Dept. of Commerce, Office of Technical Service. - fide PATTERSON 1965.

GUSSEROV A.: Virchows Arch. p

HAHN O.: "Applied Radiochemis Ithaca N.Y. 1936.

HAMILTON A., and H.L.HARDY: " Inc., N.York 1949.

HAM A.R.: "Histology", Lippin

HARLEY J.H., N.A. HALDEN and

HATUDA Z.: Kyoto Univ. Coll.

HAXEL O. and G.Schumann: Z.f.

HENDERSON D.A. and J.A.INGLIS

HENSEL W.: Słowiańszczyzna Wc 1965.

HILL C.R.: H. Phys. 8: 17, 19

- Nature 208: 423, 1

- Routes of Uptake o national Symposium Processes. Stockhol

HILL C.R. and Z.JAWOROWSKI: Ne

HINDMARSH M.O., J.VAUGHAN, L. Radiol. 24: 365, 1

HIRSCHLER D.A. and L.F.GILBERT

HOFMANN K.B.: "Das Blei bei de chow-Holzendorff

HOFREUTER D.H., E.J.CATCOTT, R viron. Health. 3: 5

DUP050313906

HOLLARD M.A.: Bull. Soc. Chim. 29: 116, 1903.

HOLTZMAN R.B.: ANL - 6297, 1960 A.

ANL - 6199, 1960 B.

H. Phys. 8: 315, 1962.

IBID. 9: 385, 1963 A.

ANL - 6769, 1963 B.

Lead-210 /RaD/ and Polonium-210 /RaF/ in Potable Waters in Illinois. in "The Natural Radiation Environment". eds. J.A.S. Adams and W.M.Lowder. The University of Chicago Press, Chicago, 1964.

^{226}Ra and the Natural Airborne Nuclides ^{210}Pb and ^{210}Po in Arctic Biota. XI International.Congress of Radiology. Rome, 22-28 Sept. 1965.

Nature: 210: 1094, 1966 B.

HOLTZMAN R.B. and F.H.ILCEWICZ: Science 153: 1259, 1966.

HORIUCHI I. and Z.HORIUCHI: AEC - tr - 4482, p.751, 1958.

HURSH J.B.: UR 522, 1958.

- Science 132: 1666, 1960.

INGLESON H.: Analyst 63: 546, 1938.

ISRAEL H.: Radioactivity in the Atmosphere. in "Comp, of Meteorology" p.155. American Meteor. Soc., Boston 1951.

JACOBI W.: HMI-B - 21, 1962.

JAWOROWSKI Z.: Nukleonika 8: 333, 1963 A.

IBID. 8: 681, 1963 B.

Thesis, Institute of Nuclear Research, Warsaw, 1963 C.

JAWOROWSKI Z.: Nukleonika 8: 853, 1963 D.

- IBID. 9: 57, 1964.

- Bull. Acad. Polon. Sc. Cl.VI. 8: 439, 1965 A.

- Nukleonika 10: 297, 1965 B.

- Atompraxis 11: 271, 1965 C.

- Nature 212: 886, 1966.

- Unpublished data: Lead was determined in four averaged samples of coals, used in a great power station in Warsaw and in 4 averaged samples of fly ash from the same station.

JAWOROWSKI Z.,J.BILKIEWICZ and W.KOSTANECKI: Int. J. Rad. Biol.1967 B.

JAWOROWSKI Z. and J.BILKIEWICZ: Nukleonika 12: 225, 1967 A .

JAWOROWSKI Z. and J.PENSKO: Nature 214: 161, 1967 C.

JOHNOSON E.J. and R.D.A.POLHILL: Analyst 80: 364, 1955.

KAURANEN P. and J.K.MIETTINEN: Po^{210} and Pb^{210} in environment samples in Finland. Symposium on Radioecological Concentration Processes. Stockholm 25 - 29 April, 1966.

KAWINSKI S. and J.PERLINSKI: Hutnictwo Metali Nieżelaznych. in "Encyklopedia Przyroda i Technika". Ed. J. Hurwic. Wiedza Powszechna, Warszawa 1963.

KEHOE R.A.: Occupat. Med. 3 156, 1947.

- J.Roy, Inst. Publ. Health, Hyg. 24: 81; 177; 203, 1961 A.

- Arch. Environ. Health. 2: 418, 1961 B.

- IBID. 8: 232; 235, 1964 A.

- IBID. 8: 348, 1964 B.

DUP050313907

KEHOE R.A., F.J.THALMANN and J.CHOLAK: J. Industr. Hyg. 15: 257, 1933.

KEHOE R.A., J.CHOLAK and R.V.STORY: J. Nutr. 19: 579, 1940.

KEHOE R.A., J.CHOLAK and E.J.LORGENT: J. Amer. Water Works Ass. 36: 637, 1944.

KING P., L.B.LOCKHART Jr., R.A.BAUS, R.L.PATTERSON Jr., H.FRIEDMAN and J.H.HLIFFORD Jr.: Nucleonics 14: 78, 1956.

KIRIKOV A.P., T.N.BOGOSLOVSKAYA and G.V.GORSAKOV: Vses. Geol.-Razved. Ob'yedineniye Izv. 51: 1293, 1932.

KLIGMAN A.M.: Arch. Derm. 83: 175, 1961.

KONONENKO A.M.: Biofizika 2: 98, 1957.

KOPCEWICZ T.: Przegl. Geofiz. 8: 3, 1964.

KOSMATH W.: Gerlands Beitr. z. Geophys. 43: 258, 1934.

KOSOV N.D. and V.V.CHERDYNTSEV: Akad. Nauk SSSR Komiss. opredel. absolyut. vozrasta geol. formatsiy Byull., vypusk 1, pp 22-28, 1955. - fide TANNER 1964.

KOSTIN S. and T.POKROWSKA: "Klimatologia". PWN, Warszawa 1957.

KOSTRZEWSKI J., W.CHMIELEWSKI and K.JAŻDŻEWSKI: "Pradzieje Polski". Zakład Narodowy im. Ossolińskich, Wrocław, 1965.

KOWALSKI K.: "Jaskinie Polski". Vol. 1. p. 402. Archeological Museum, Warszawa 1951.

KOVACH M.: Trans. Amer. Geophys. Union. 28: 563, 1944.

KRAMER H.W., G.L.SCHROEDER and R.D.EVANS: Measurements of the Effect of Atmospheric Variables on ^{222}Rn flux soil gas concentrations. In "The Natural Radiation Environment", eds. J.A.S. Adams and W.M.Lowder, The University of Chicago Press, Chicago, 1964.

KRAKUZE D.P.: ANL - 6398, 1961.

KRAUT H. and M.WEBER: Biochem. Z. 317: 133, 1944.

KAYSER P. and A.MEUMIER: Ann. Phyrm. Franc. 22: 365, 1964.

LAMBERT G., M.NEZAMI and J.LAHEYRIE: Comp. Rend. 260: 619, 1965.

LANGENBACH J. and D.W.UNSELD: Klin. Wschr. 39: 386, 1961.

LANE R.E.: Arch. Environ. Health 8: 243, 1964.

LEA D.E.: "Actions of Radiation on Living Cells". Cambridge University Press. Cambridge, 1946.

LEHMAN L. and A.SITTKUS: Naturwiss. 46: 9, 1959.

LOCKHART Jr. L.B.: Radioactivity of the Radon-222 and Radon-220 Series in the Air at Ground Level. In "The Natural Radiation Environment". eds. J.A.S. Adams and W.M.Lowder. The University of Chicago Press, Chicago 1964.

LOCKHART Jr. L.B., R.A.BAUS, R.L.PATTERSON Jr. and I.H.HLIFFORD Jr.: NRL - 5208, 1958.

LYNCH G.R., R.S.SLATER and T.G.OSLER: Analyst 59: 787, 1934.

MANLEY C.H.: Analyst 62: 544, 1937.

MARCHLEWSKI L. and B.SKARŻYŃSKI: "Chemia Fizjologiczna". PZWL, Kraków, 1947.

MARSH F.: Lancet 1: 941, 1964.

MAUCHLY S.J.: Terr. Magn. Atmos. Electr. 29: 187, 1924.

MAYNEORD W.V. and G.R.HILL: Nature 184: 667, 1959.

MAYS C.W., R.HALDIN and M.A. VAN DILLA: Rad. Res. 9: 438, 1958.

MC DONALD W.S., F.EZMIRLIAN, P.SPAIN and C. MC ARTHUR: J. Biol. Chem. 189: 387, 1951.

DUP05031390B

MC LENNAN J.C.: Phil. Mag. 14: 760, 1907.

MIYASAKI S.: Arch. Exp. Pathol. Pharm. 150: 39, 1930.

MONCRIEFF A.A., O.P.KOSMIDES, B.E.CLAYTON, A.D.PATRICK, A.G.C. RENWICK and G.E.ROBERT: Arch. Dis. Childr. 47: 1. 1964.

MONIER-WILLIAMS G.W.: "Lead in Food". Reports, Public Health Medical Subjects. Ministry of Health. London, No. 88, 1938.

- Trace Elements in Food. John Wiley and Sons Inc., N.York, 1950.

MONKHOUSE A.C.: "Coke and Gas." 1950. - fide W.Francis: "Coal". F.Arnold /Publishers/ LTD/, London, 1954.

MOSES H., A.F.STEHNEY and H.P.LUCAS Jr.: J. Geophys. Res. 65: 1223, 1960.

MUROZUMI M., T.J.CHOW and C.C.PATTERSON: NYO - 3450 - 1, p. 213, 1965.

MUTH H. and E.OBERHAUSEN: Die Verteilung der natürlichen radionuclide RaD /^{210}Pb/ und ^{210}Po in Menschlichen Körper und die dadurch bedingte Strahlenwirkung. Tagungsbericht, Gemeinsame Tagung der Deutschen Gesellschaft für Biophysik E.V., der Oesterreichischen Gesellschaft für Reine und Angewandte Biophysic und der Schweizerischen Gesellschaft für Strahlenbiologie. Vienna, 1964, p. 295.

NELSON K.W. and R.E.HAMM: Arch. Industr. Health. 17: 38, 1958.

NEUMAN W.F. and B.J.MULRYAN: J. Biol. Chem. 195: 843, 1952.

NEUMAN W.P. and W.M.NEUMAN: "The Chemical Dynamics of Bone Mineral". The University of Chicago Press. Chicago 1958.

NESMEYANOV A., A.ŁAŃICKI and N.RUDENKO: "Otrzymywanie izotopów promieniotwórczych". /Translation from Russian/. PWN, Warszawa, 1956.

NICOLAYSEN L.O.: Geochim. Cosmochim. Acta 11: 41, 1957.

NORRIS W.P., A.A.TYLER and A.M.RUES: Science 128: 456, 1958.

NUTT D.C.: Polar Notes. Dartmouth College Library. Hanover, New Hampshire, No. 1. Nov. 1959.

NUTT D.C.: Polar Forschung 31: 82, 1961.

OSBORNE R.V.: Nature 199: 143, 1963 A.

- IBID. 199: 295, 1963 B.

PAKHOTINA N.S.: Gig. Sanit. 23: 3, 1958.

PATTERSON C.C.: Geochim. Cosmochim. Acta 7: 151, 1955.

- Arch. Envir. Health 11: 344, 1965.

PATTERSON Jr. K.L. and L.B.LOCKHART Jr. : Geographical Distribution of Lead-210 /RaD/ in the Ground Level Air. In "The Natural Radiation Environment". Eds. J.A.S. Adams and W.M. Lowder. The University of Chicago Press. Chicago, 1964.

PEIRSON D.H., R.S.CAMERAY and G.S.SPICER: Tellus 18: 427, 1966.

PENSKO J., T.WARDASZKO, K.MAMONT and M.WOCHNA: Dependence of Natural Atmospheric Radioactivity on Some Geophysical Factors. - in preparation, 1967.

POLDERVAART A.: Special Paper, Geol. Soc. Amer. 62: 119, 1955.

PRINCE A.L.: Soil Sci. 84: 413, 1957.

PRICE W.J.: "Nuclear Radiation Detection". McGrow-Hill Book Co. N.York, 1958.

DUP050313909

PUTNAM P.C.: "Energy in the Future". D.Van Nostrand Co., Inc. N.York, 1953.

RADFORD Jr. E.P. and V.R.HUNT: Science 143: 247, 1964.

RAJEWSKY B. and W.STAHLHOFEN: Nature 198: 960, 1963.

RAMA M.K. and E.D.GOLDBERG: Science 143: 98, 1961.

RAMA M.K. and M.HONDA: J. Geophys. Res. 66: 3227, 1961.

RANKAMA K.: "Isotope Geology". Pergamon Press Ltd., London 1954.

REED G.W., K.KIGOSHI and A.TURKEVICH: Geochim. Cosmochim Acta 20: 122, 1960.

REYNOLDS P.M.: J. Soc. Chem. Ind. 67: 341, 1948.

RIVERA J.: Nature 207: 1330, 1965.

RATHERFORD Sir E., J.CHADWICK and C.D.ELLIS: "Radiations from Radioactive Substances". Cambridge 1951. - fide ANDERSON et al. 1964.

SAND H.J.S.: Analyst 55: 309, 1930.

SANDELL E.B.: "Colorimetric Determination of Trace Metals". Interscience Publishers Inc. N.York, 1959.

SCHOLLANDER F.F., E.A.HEMMUNGSEN, L.K.COACHMAN and D.C.NUTT. J. Glaciol. 3: 813, 1961.

SCHROEDER H.A. and I.J.BALASSA: J. Chron. Dis. 14: 408, 1961.

SCHULTZ G.K.: H. Phys. 11: 1317, 1965.

SCHWARTZ I.C.: Extrarenal Regulation with Special Reference to the sweat glands. In "Mineral Metabolism - An Advanced Treatise". Eds. C.L. Comar and F. Bronner. Academic Press, N.York, 1960. Vol. 1. Part A.

SCHEVCHENKO Yu. V.: Priroda 48: 13, 1959.

SHLEIEN B. and A.G.FRIEND: Nature 210: 579, 1966.

SISIGHINA T.I.: Ekshalacya radona s poverkhnosti neskolkikh pochv evropeiskoi chasti SSSR i Kazakhstana. In "Radioactivniye isotopy v atmosfere i ich ispolsovaniye v orologii". Ed. I.L. Karol, L.V. Kirichenko, Yu.B. K pevcev, V.M. Kurganskaya, C.G. Malakhov, G.A. Sered I.V. Yagodovskii. Atomisdat, Moskva 1965.

SMYTH L.B.: Phil. Mag. 24: 632, 1912.

SNYDER H., W.F.KISIELESKI, P.S.ANTHONY and R.D.FINKLE: MDDC 1946.

SNYDER L.J.: Anal. Chem. 19: 684, 1947.

SPIERS F.W.: Brit. J. Radiol. 19: 52, 1946.

" IBID. 26: 296, 1953.

STAHLHOFEN W.: THESIS J.W. Goethe Universität, Frankfurt a 1964.

STARIK I.Ye. and O.S.MELIKOVA: Radiyevyi Institut imeni V. pina, Trudy 5: 184, 1957.

STEBBENINS A.K.: DASA - 536 B, 1961.

SULTZER M. and J.B.HURSH: UR - 266, 1953.

TANNER A.B.: Radon Migration in the Ground - A Review. In tural Radiation Environment". Eds. J.A.S. Adams a Lowder. The University of Chicago Press, Chicago,

TATSUMOTO M. and PATTERSON C.C.: Nature 199: 350, 1963.

THOMPSETT S.L.: BIOCHEM. J. 30: 345, 1936.

DUP050313910

REVIEW REPORTS

of the

NUCLEAR ENERGY INFORMATION CENTER

17. Oncological Institute, PROGRESS REPORT FOR THE YEAR 1964 (In Polish, English and Russian), 1965, 194 p.
18. B. Kuchowicz, NEW PROPOSALS OF NEUTRINO EXPERIMENTS (In English), 1965, 40 p.
19. M. Taube, HYDROGEN–THE CARRIER OF LIFE An approach to the atomic biology (In Polish), 1965, 138 p.
20. and 21. B. Kuchowicz, THE BIBLIOGRAPHY OF THE NEUTRINO, Part I and II, 1966 (In English), 212 p. – 228 p.
22. POLISH PAPERS ON NUCLEAR ENERGY PUBLISHED IN 1964, A bibliography, 1966, 244 p.
23. A BIBLIOGRAPHY OF ONCOLOGY, Vol I, Part I, 1966, 222 p.
24. INSTYTUT ONKOLOGII – RAPORT Z PRAC BADAWCZYCH PROWADZONYCH W 1965 r., 1966, 87 s.
25. M. Taube, HYDROGEN–THE CARRIER OF LIFE An approach to the atomic biology (In English), 1967, 103 p.
26. INSTYTUT ONKOLOGII – RAPORT Z PRAC BADAWCZYCH PROWADZONYCH W 1966 r., 1967, 154 s.
27. A BIBLIOGRAPHY OF ONCOLOGY, Vol I. Part II and III, 1967, 326 p.
28. B. Kuchowicz, RELATIVISTIC SPHERES AS MODELS OF NEUTRON STARS, 1967, 28 p.
29. Z. Jaworski, STABLE AND RADIOACTIVE LEAD IN ENVIRONMENT AND HUMAN BODY, 1967, 181 p.

DUP050313911