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# Pb-210 in Ground-Level Air and Snow in the Environment of the Karlsruhe Nuclear Research Center

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## Abstract

Pb-210 concentrations in air and snow were measured near the Karlsruhe Nuclear Research Center (KfK). The results revealed that the Pb-210 concentrations in air near the KfK are lower by a factor of 1.5 to 2.0 compared to those in others parts of Germany. According to these results the Pb-210 concentration in snow is lower than the expected values for the middle latitudes of the northern hemisphere. These low values may be a consequence of the low content of natural radioactivity in the soil of the river Rhine valley. The deposition rate of Pb-210 aerosols on snow is extremely low. The low deposition can be explained by a combined effect of lower deposition velocity and an increased resuspension for Pb-210 aerosols on ice-covered snow layer.


## Pb-210 in der bodennahen Luft und in Schnee in der Umgebung des Kernforschungszentrums Karlsruhe


## Zusammenfassung

Nahe dem Kernforschungszentrum Karlsruhe (KfK) wurden Pb-210-Konzentrationen in Luft und Schnee gemessen. Die folgenden Ergebnisse zeigten, daß die Pb-210-Konzentrationen in Luft um den Faktor 1,5 bis 2 niedriger sind verglichen mit anderen Messungen in der Bundesrepublik Deutschland. Entsprechend diesem Ergebnis ist die Pb-210-Konzentration in Schnee kleiner als für mittlere geographische Breiten der Nordhalbkugel erwartet wird. Diese niedrigen Werte können eine Folge des niedrigen Gehalts der Erde im Rheintal an natürlicher Radioaktivität sein. Die Ablagerungsrate von Pb-210-Aerosolen auf Schnee ist extrem klein. Die niedrige Ablagerung kann erklärt werden als ein gemeinsamer Effekt einer niedrigeren Ablagerungsgeschwindigkeit und einer erhöhten Resuspension für Pb-210-Aerosole auf der eisbedeckten Schneeschicht.

## Content

## 1. Introduction

Pb-210 is the longlived daughter product of Rn-222 with a half-life of 22.3 y, and a maximum beta energies of 0.02 - 0.06 keV. Pb-210 decays and is converted into stable Pb-206 according to the following way:

$$Pb-210 \xrightarrow[22.3\,y]{\beta} Bi-210 \xrightarrow[5{,}0\,d]{\beta} Po-210 \xrightarrow[138\,d]{\alpha} Pb-206\ stable\ [1].$$

Since the half-lives of Bi-210 and Po-210 are short, the activity of these radionuclides are very often in equilibrium with Pb-210. Rn-222 released from the upper layer of soil decays to Pb-210 in the ground level air. The mean concentration in the middle latitudes of the northern hemisphere is about 0.5 mBq/m$^3$. The inhaled activity for non-smokers is about 3.8 Bq Pb-210/y. The inhaled activity causes an absorbed dose to the bone of about 0.1 mSv/y.

After a residence time of about 10 d in air, Pb-210 is deposited on plants and soil. The sum of dry and wet deposition equals $1.3 \cdot 10^8$ Bq/km$^2$·y. This is a small fraction of the Pb-210 activity always contained in the soil layer of agricultural interest. Pb-210 is strongly bound in soil, therefore the deposited fraction is mainly responsible for the absorbed dose caused by ingestion. As the uptake by ingestion is about 10 times higher than by inhalation, the annual dose exposure for bones is in the range of 1 mSv/y [2].

Following the daughter products of Radon, Pb-210 is the most important natural radionuclide. For that reason we measured its concentration in air near the Karlsruhe Nuclear Research Center. Since data about concentrations in snow are very rare, we performed some measurements in snow. Using the results we tried to determine the dry deposition on snow.

## 2. Literature Survey

### 2.1. Pb-210 in Air

The levels of Pb-210 in air are the result of a series of complex factors, such as the continentality of the air masses involved, the meteorology of the region and the content of radium in the upper layers of soil of the adjoining areas.

The northern hemispheric values are in general higher compared to the southern hemispheric values, possibly due to the land area being nearly three times that of the southern hemisphere. Maximum monthly values are reaching 3.3 mBq Pb-210/m$^3$ (Fig. 1, Tab. 1, Tab. 2) [2, 3].

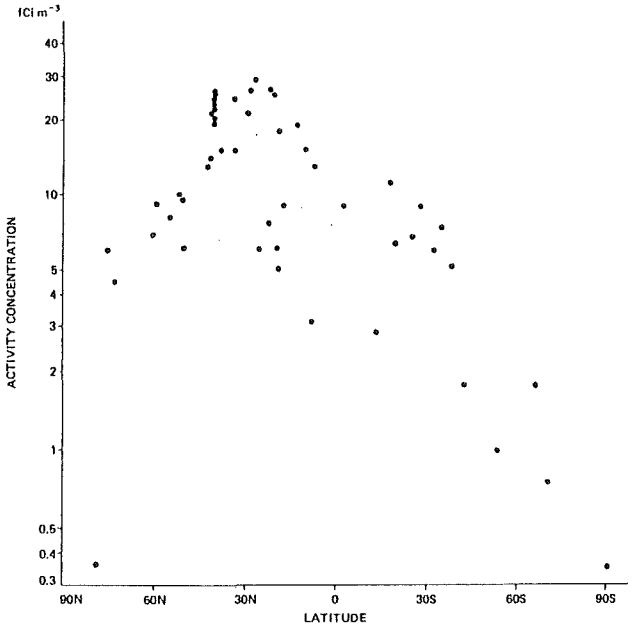


Fig. 1: Latitudinal Distribution of the Pb-210 Concentration in Surface Air [2].

The values in continental Europe seem to be less, being in the range of 0.1 - 1.4 mBq/m$^3$. Minimum values are observed in the polar regions. Snowfall preventing emanation of radon and the absence of large land masses are primarily responsible for this. A seasonal variation in Pb-210 has been observed with winter maxima and summer minima, which in some cases corresponds to

ambient radon levels. Other areas, however, do not show such variations or at least they do not have the same pattern each year. Some scientists attributed these observations to the increasing incidence of inversion conditions in winter [4 - 12].

In the troposphere the Pb-210 concentration in air decreases by height. The values in the stratosphere are higher than the values below the tropopause [13].

Table 1: Pb-210 Concentrations in Surface Air Measured in the Federal Republic of Germany.

| Sampling station | Concentration range in $mBq/m^3$ | Reference |
|---|---|---|
| Munich | 0.08 - 1.37 | [4] |
| Brunswick | 0.11 - 1.41 | [5, 6] |

Table 2: Pb-210 Concentration in Surface Air Measured in Different Continents.

| Sampling place | Concentration range in $mBq/m^3$ | Reference |
|---|---|---|
| Europe | 0.05 - 1.41 | [3 - 6] |
| North America | 0.00 - 2.81 | [3, 10, 14] |
| South America | 0.01 - 0.74 | [3, 14] |
| Asia | 0.00 - 2.37 | [3, 7, 9, 14] |
| Australia | 0.09 - 0.29 | [11] |
| Antarctic regions | 0.00 - 0.12 | [12] |

## 2.2. Pb-210 in Rain Water and Snow

The global distribution of Pb-210 in rain water follows the same pattern as that of surface air, being the highest in the middle latitudes (Fig. 2). The annual averages of Pb-210 concentration in rain at different locations have been found to vary between 0.04 - 0.26 Bq/l. A typical value for continental sampling stations of the middle northern latitudes is 0.1 Bq/l rain water. For the annual dry and wet

deposition, the values between 5 - 33 mBq/cm$^2$·y are measured. The deposition rates at continental sites exceed those of sea sites by a factor of 2. During one rainfall the first part of the collected water contains much higher concentrations than the rest [2, 3, 11, 14 -1 6].

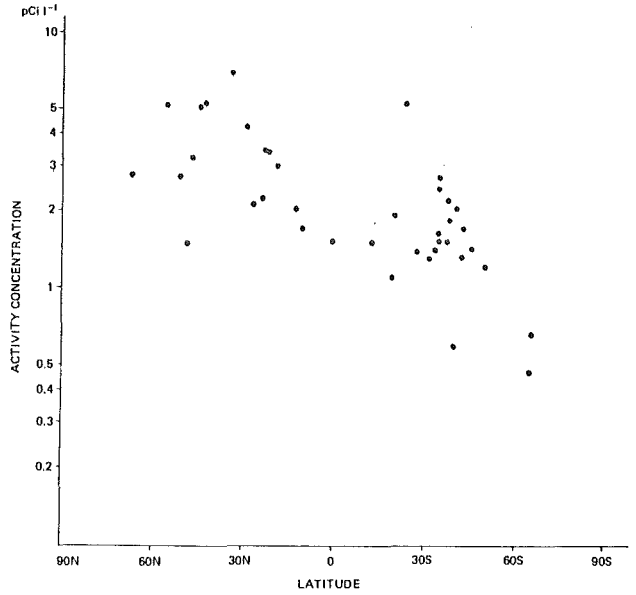


Fig. 2: Latitudinal Distribution of the Pb-210 Concentration in Rain Water [2].

## 3. Pb-210 Measurements and Results

### 3.1. Pb-210 Concentration in Snow Samples from KfK

**Sample Collection:** In January 1982 an area of 9 x 1 m$^2$ in the north-east of KfK was selected for the collection of snow samples. During the second week of January when Germany was totally covered with new snow a plastic rectangular container was used to collect the snow samples from the specified area thus yielding approximately the same volume of snow water. The selected site was divided into nine equal squares of 1 x 1 m$^2$. A daily sample was taken at 1.30 p.m. from each square with a sampling depth of 5 cm. The sampling period was nine days.

**Sample Preparation:** Excepting 12.2 l on the first day the collected snow water samples yielded 8.5 to 8.8 l of volume. After the addition of 70 ml $HNO_3$ each sample was evaporated to a volume of 200 ml. 20 mg of $Pb^{2+}$ carrier were added

and the sample was taken to dryness. To achieve an oxidation of organic compounds and a white residue a small amount of $H_2O_2$ was mixed to some samples.

**Analysis:** Pb-210 activity was determined by first separating its Bi-210 daughter from the sample and then measuring ß's from Bi-210 acitivity that had grown over a period of one week.

The radiochemical procedure used, was as follows: The white residue was covered with HBr, taken to dryness and then dissolved in 3 M HBr. Lead was extracted by trioctylamine acting as an ion-exchanger and thus separating it from Ra, Ba, Ca etc. The reextraction from trioctylamine was performed with 10 M HCl. Bi carrier was added at this stage and Pb was separated from Bi by precipitating Bi as Bismuthoxychloride. Finally lead was precipitated as $PbMoO_4$.

Radiochemical blanks were determined by analogous treatment of distilled water samples.

**Counting Procedure:** Lead molybdate precipitate was spread in form of a thin layer on a filter paper having a diameter of 4·8 cm and stored for one week to allow the ingrowth of Bi-210. The precipitates were covered with an aluminium foil weighing 6·3 mg/cm$^2$ in order to absorb the soft ß's of Pb-210 with an energy of 0.02 - 0.06 MeV and the α's of Po-210. Since the activity in snow-water samples was very low, a low background counting setup was required. A low background can be achieved by shielding the detector and operating it in anticoincidence mode. For this purpose a low background gas proportional counter Model BF 100-2L, Berthold, with automatic sample changer and an anticoincidence unit LB 2360 was used. The output from the anticoincidence unit was fed to a scaler and finally to a printer. With shielding and anticoincidence mode the background was reduced to about 2 counts/min. For counting the lead molybdate precipitate covered with aluminium foil was placed on a stainless steel planchet and installed in very close to the window of the counter.

**Calibration of the Counter:** The counter was calibrated with reference activities of lead-210. According to the analytical procedure ten lead-210 standard solutions were processed after adding the same amount of lead carrier, i. e., 20 mg. The final lead molybdate precipitate was weighed and chemical yield was calculated for each case varying from 80 - 90 %. After a period of one week the samples were counted and the counting efficiency was determined. An average value of 30.3 % was obtained.

**Results:** The results of Pb-210 analyses are presented in Table 3.

Table 3: Pb-210 Concentration in Snow Samples from the North-East Side of KfK.

| Sampling date | Pb-210 in snow | |
|---|---|---|
| | $Bq/m^2$ | mBq/l |
| 12.01.1982 | 0.23 | 21 |
| 13.01.1982 | 0.30 | 35 |
| 14.01.1982 | 0.77 | 91 |
| 15.01.1982 | 0.30 | 35 |
| 18.01.1982 | 0.25 | 29 |
| 19.01.1982 | 0.26 | 30 |
| 20.01.1982 | 0.42 | 48 |
| 21.01.1982 | 0.33 | 38 |
| 22.01.1982 | 0.42 | 47 |
| average | 0.36 | 42 |

### 3.2. Pb-210 Concentration in Air Samples from KfK

**Sample Collection:** The air particles were collected by drawing continuously about 60 $m^3$ of air/hour through a glass fibre filter of 20 cm diameter using a centrifugal pump. For these filters a 99.9 % efficiency for the retainment of submicron size particles is indicated. The filters were changed daily at 1.30 p.m., except for the weekend. During a sampling period of nine day nine samples were collected above each square of snow as mentioned before.

Additionally about 1 km north of the KfK air samples were collected in 1981 and 1982.

Three air samples of about 1300 $m^3$ of air were collected at the same time on February 18th, 1982, one west, one north and one north-east of the KfK.

**Sample Preparation:** The particle filters were cut into small pieces and soaked in aqua-regia after adding 20 mg of Pb carrier. The mixture was stirred well for one or two days, slightly heated at a low temperature and finally filtered. The treatment was repeated with the residue to ensure a complete leaching of Pb. The filtrate was evaporated yielding a white residue. Analyses using the same glass fibre filters were performed to determine the blanc value.

**Analysis, Counting Procedure, Calibration of the Counter:** Same as for air samples.

**Results:** The results of the analyses of air samples are presented in the Tables 4, 5 and 6.

Table 4: Pb-210 Concentration in Air Samples from North-East Side of KfK. Samples Collected above Squares of Snow.

| Sample taken | | Pb-210 concentration in air |
|---|---|---|
| from | to | mBq/$m^3$ |
| 11.01.1982 | 12.01.1982 | 0.54 |
| 12.01.1982 | 13.01.1982 | 0.58 |
| 13.01.1982 | 14.01.1982 | 0.53 |
| 14.01.1982 | 15.01.1982 | 1.10 |
| 15.01.1982 | 18.01.1982 | 1.54 |
| 18.01.1982 | 19.01.1982 | 2.22 |
| 19.01.1982 | 20.01.1982 | 2.20 |
| 20.01.1982 | 21.01.1982 | 2.08 |
| 21.01.1982 | 22.01.1982 | 1.68 |

Table 5: Pb-210 Concentration in Air Samples, Collected North of the KfK in 1981 and 1982.

| Sample taken from | to | Pb-210 concentration mBq/m³ |
|---|---|---|
| **1981:** | | |
| 11.02. | 24.02. | 0.20 |
| 24.02. | 10.03. | 0.27 |
| 24.03. | 07.04. | 0.20 |
| 07.04. | 21.04. | 0.27 |
| 21.04. | 05.05. | 0.13 |
| 05.05. | 19.05. | 0.15 |
| 19.05. | 29.05. | 0.17 |
| 09.06. | 22.06. | 0.10 |
| 22.06. | 06.07. | 0.07 |
| 06.07. | 21.07. | 0.17 |
| 21.07. | 03.08. | 0.14 |
| 03.08. | 17.08. | 0.30 |
| 17.08. | 31.08. | 0.14 |
| 31.08. | 07.09. | 0.30 |
| 11.11. | 23.11. | 0.43 |
| 14.12. | 28.12. | 0.71 |
| **1982:** | | |
| 28.12. | 11.01. | 0.16 |
| 11.01. | 18.01. | 0.92 |
| 22.01. | 01.02. | 0.04 |
| 01.02. | 15.02. | 0.39 |
| 15.02. | 01.03. | 0.42 |
| **average** | | 0.23 |

Table 6: Pb-210 Concentration in Air Samples Collected West, North and North-East of the KfK. Samples Taken in February 18th, 1982.

| Sampling station | Pb-210 concentration mBq/m³ |
|---|---|
| North | 0.71 |
| West | 0.51 |
| North-East | 1.06 |

## 4. Discussion of the Results

The annual mean value of the Pb-210 concentrations measured in samples collected north of the KfK is 0.23 mBq/m$^3$. The mean value was calculated from the single results weighing them according to the sampling time. In Brunswick between 1974 and 1979 annual mean values from 0.24 to 0.41 mBq Pb-210/m$^3$ were measured. In Munich between 1976 and 1979 values ranging from 0.50 to 0.57 mBq Pb-210/m$^3$ were determined. This means that the Pb-210 concentration in air near the KfK is lower than the other values measured in Germany. Due to this low concentration in air, the results for snow water are lower too, compared with the mean value expected for the middle latitudes of the northern hemisphere. The lower air concentrations and consequently the lower concentrations in snow water can be explained by the lower natural radioactivity in the soil of the river Rhine valley.

As observed by others, the highest concentrations were reached in the winter months. Very high daily concentrations were observed some days after Central Europe had been covered by snow. This can be explained by the strong influence of inversion conditions sometimes existing in the winter. The high values measured after the snowfall can as well be attributed to inversion conditions and additionally to a very low deposition rate at this time.

Considering the determination of the original Pb-210 content on the first day after snowfall and the measurement of the daily increase of Pb-210 activity in Bq/m$^2$ a calculation of the deposition velocity of Pb-210 aerosols should be possible.

Using the 0.23 Bq/m$^2$ from the January 12th, 1982, the daily air concentrations and a deposition velocity of 0.1 cm/s for aerosols, 1.56 Bq/m$^2$ are calculated for January 22nd, 1982. This value is much higher than the measured value of 0.42 Bq/m$^2$ (the 0.77 Bq/m$^2$ for the January 14th, 1982 are unexplainable, however).

The standard deviation of the Pb-210 concentrations in snow water is too high to measure the real deposition velocity, but it should be about 0.015 cm/s. This value is only acceptable if the particle diameter of Pb-210 aerosols is very small.

The residence time of Pb-210 in surface air is about 10 d. Therefore it is to be expected that aerosols carrying Pb-210, should have normal, i.e. large, diameters. This means that a deposition velocity of 0.015 cm/s is too low.

An explanation for this very low deposition rate could be the fact, that already during the first day the snow was covered with an ice layer. The low deposition rate can be understood as a combined effect of a lower deposition rate and very effective resuspension from the ice layer. Further snowfall at January 23rd, 1982, followed by rain, stopped the experiment.

## 5. Literature

[1] Karlsruher Nuklidkarte, W. Seelmann-Eggebert et al., Kernforschungszentrum Karlsruhe GmbH, 7500 Karlsruhe, Postfach 36 40, Federal Republic of Germany

[2] Sources and Effects of Ionizing Radiation, UNSCEAR 1977, United Nations, New York, 1977.

[3] C. Rangarajan et al., "Global variation of Pb-210 in surface air and precipitation", HASL-298, 1976.

[4] R. Winkler et al., "Analysis of Pb-210 and Po-210 concentrations in surface air by an α spectrometric method", Health Physics, Vol. 41, No. 3, 1981, p. 495.

[5] W. Kolb, "Radionuclide concentration in Ground Level Air from 1974 to 1977 in North Germany and North Norway", PTB-Ra-9, November 1978.

[6] W. Kolb, "Radionuclide Concentration in Ground Level Air from 1978 to 1979 in North Germany and North Norway", PTB-Ra-11, February 1980.

[7] U.C. Mishra et al., "Natural Radioactivity of the Atmosphere over the Indian Land Mass, Inside Deep Mines and over Adjoining Oceans", CONF-780422, Vol. 1, Proceedings of a symposium on "Natural Radiation Environment III", Houston, Texas, April 23 - 28, 1978, p. 327.

[8] J. Sanak et al., "Lead-210 in the Atmosphere", ibida, Vol. 7, p. 445.

[9] L.U. Joshi et al., "Seasonal Variations of Radium-D (Lead-210) in Ground Level Air in India", Health Physics, Vol. 15, 1968, p. 67.

[10] S.S. Brar et al., "Lead-210 and stable lead concentrations in surface air at Argonne, Illinois", ANL-7278, December 1966.

[11] J. Bonnyman et al., "Lead-210 in the Australian environment, 1964-70", CONF-720805-P2, Volume II, Proceedings of "The Natural Radiation Environment II", Houston, Texas, USA, August 7 - 11, 1972, p. 819.

[12] W. Maenhaut et al., "Radionuclides in the Pole Atmosphere", Journal of Geophysical Research, Vol. 84, No.C6, June 20, 1979, p. 3131.

[13] Z. Jaworowski, Central Laboratory for Radiological Protection, 03-194 Warsaw, Konwaliowa 7, Poland, personal communication.

[14] A. Nevissi et al., "Use of Lead-210 and Polonium-210 as Tracers of Atmospheric Processes", CONF-780422, Vol. 1, as [7], p. 405.

[15] H.E. Moore et al., "Pb-210 Fluxes Determined from Pb-210 and Ra-226 Soil Profiles", Journal of Geophysical Research, Vol. 81, No. 6, 1976, p. 1056.

[16] G.L. Ter Haar et al., "Lead and Lead-210 in Rainwater", Nature, Vol. 216, 1967, p. 353.