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ENVIRONMENTAL RADIOACTIVITY ANNUAL REPORT 1993

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CONTENTS

SUMMARY

During 1993 artificial radioactivity in the environment in New Zealand and Rarotonga continued to be at trace levels only, typical of recent years. There has been no significant change in the radioactivity status of the environment since 1992.

Atmospheric and deposited radioactivity was monitored at Kaitaia, Hokitika and Rarotonga and milk radioactivity was monitored in Auckland, Taranaki and Westland. Average levels measured were: total beta activity in air, 0.15 mBq/m^3; ^{90}Sr deposition, 0.1 Bq/m^2; ^{137}Cs in milk, 0.15 Bq/g K; ^{90}Sr in milk, 0.02 Bq/g Ca. Total beta activity depositions at Hokitika and Rarotonga were 172 Bq/m^2 and 334 Bq/m^2 respectively. Annual total ^{210}Pb depositions at Kaitaia, Hokitika and Rarotonga were 48, 115 and 33 Bq/m^2 respectively.

No artificial radionuclides were detected by gamma spectroscopic analysis of high-volume air filters during 1993, with weekly sampling periods. Annual average atmospheric ^{137}Cs concentrations, assessed by analysis of yearly air filter aggregates, at Kaitaia, Hokitika and Rarotonga were 0.02, 0.05 and <0.02 μBq/m^3 respectively.

Average atmospheric concentrations of natural ^{7}Be and ^{210}Pb were 2.9 mBq/m^3 and 0.05 mBq/m^3 respectively. Atmospheric beta activity was found to be made up of contributions from cosmogenic radionuclides (60%), ^{210}Pb/^{210}Bi (25%) and natural radionuclides in suspended dust (15%).

A study of trends in atmospheric ^{137}Cs concentrations, derived from analyses of yearly aggregates of air filters for the period 1986 - 1993, indicated that fission products from the Chernobyl reactor did not reach detectable levels in New Zealand, confirming earlier projections and indications.

Fallout ^{210}Pb and ^{210}Po in diet contribute estimated annual average effective dose commitments of 4.5 and 2.3 μSv respectively, compared to the total dose commitment of ^{90}Sr and ^{137}Cs, of about 2 μSv/y. The consumption of rainwater may increase the ^{210}Pb contribution to about 22 μSv/y.

The report includes data on trends in atmospheric radioactivity levels since 1987.

1. INTRODUCTION

The National Radiation Laboratory (NRL) has monitored environmental radioactivity levels in the New Zealand and South Pacific regions since 1960, as described in earlier reports in this series[1,2]. Monitoring was initially conducted for radioactive fallout from nuclear weapons tests in the Northern Hemisphere, and then for fallout from the French testing programme in the Tuamotu Archipelago. When the French atmospheric testing programme was terminated in 1974 monitoring continued for residues from atmospheric tests and in order to detect any venting from the underground tests.

By 1986 levels of weapons-test debris in the atmosphere and rainwater had decreased to near the limits of detection for the monitoring techniques then in use. The extensive monitoring network was then scaled down[3] to a size more commensurate with the degree of hazard involved, contracting to only three monitoring sites with two in New Zealand and one in the Cook Islands, as described below. The sensitivity of atmospheric monitoring at the remaining sites was increased with the installation of high-volume air samplers[4].

The emphasis of the present monitoring programme is the need to provide warning of any influx of radioactivity into the New Zealand and South Pacific regions from any source, to monitor trends in levels, and to facilitate public protection planning in the unlikely event of a significant pollution incident. The Chernobyl nuclear reactor disaster of 1986 highlighted the need for sensitive environmental monitoring, particularly in major food producing countries. The New Zealand monitoring programme provides the basis for certification of the radioactivity content of foodstuffs, both exported and consumed locally. It also serves as the basis of consumer and commercial advisory services concerning radioactivity, and is the basis of comparisons of the environmental radioactivity status of the South Pacific region with that of other regions.

Through its environmental monitoring operations, NRL is involved in international networks which have been set up by the International Atomic Energy Agency (IAEA) and the World Health Organisation (WHO) to improve cooperation and information dissemination, particularly during nuclear emergencies (through the Emergency Notification Convention). The Laboratory is also a member of the WHO/UNEP (United Nations Environment Programme) Global Environmental Radiation Monitoring Programme (GERMON) and submits monitoring results quarterly to the GERMON headquarters in France; it is a WHO Collaborating Centre for environmental radiation measurement; and it has recently been invited to join the IAEA's international CREM (Compilation of Reported Environmental Monitoring data) network. The Laboratory thus acts as an international point of contact for the South Pacific region in all matters pertaining to environmental radioactivity.

In order to maintain a high standard of capability in radiochemical analysis NRL takes part in quality assurance programmes run by the WHO, IAEA, the United States Environmental Protection Agency (EPA) and the South Pacific

Environmental Radioactivity Association (SPERA). Results of analytical intercomparisons undertaken during 1993 are included in this report.

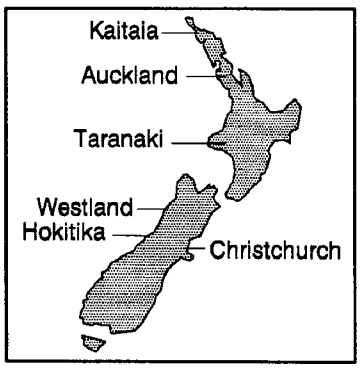
Although the environmental monitoring programme is designed primarily to detect and monitor levels of artificial radioactivity, measurements are extended to include naturally occurring radioactive materials wherever practicable, for comparative and scientific purposes. The atmospheric monitoring therefore includes measurements of concentrations of lead-210 (^{210}Pb), polonium-210 (^{210}Po), beryllium-7 (^{7}Be) and sodium-22 (^{22}Na) in the atmosphere. Beryllium-7 and ^{22}Na are cosmogenic nuclides (ie, products of spallation reactions of cosmic radiation within the upper atmosphere) while ^{210}Pb and ^{210}Po are decay products of gaseous radon-222 (^{222}Rn, which diffuses out of soil) and are of lower-atmosphere origin. The measurements provide information on atmospheric circulation patterns which affect the distribution of pollutants. Levels of uranium (^{238}U) and thorium (^{232}Th) decay products and potassium-40 (^{40}K) are also monitored as indicators of the suspended dust content of air filters, and to provide an indication of the natural component of any measured radioactivity. Lead-210 deposition is also monitored, together with that of ^{90}Sr.

2. MONITORING PROGRAMME

The environmental radioactivity monitoring programme comprises the following measurements.

Atmospheric radioactivity: atmospheric monitoring is regarded as the most important component of the monitoring programme because any radioactive pollution reaching the region will inevitably have been transported in the atmosphere and the high-sensitivity monitoring would provide early warning of any influx or changing trend in environmental radioactivity levels. Total beta activity (TBA), fission products, ^{7}Be, ^{210}Pb, ^{210}Po, ^{238}U and ^{232}Th decay products, and ^{40}K levels are monitored with weekly sample collections of surface air at Kaitaia, Hokitika and Rarotonga. In addition, air filters collected during the year are now bulked quarterly and annually for sensitive measurements of average levels of any other radionuclides which may have been present — for example, caesium-137 (^{137}Cs) and ^{22}Na.

Radioactive deposition: TBA deposition is monitored at Hokitika and Rarotonga, with weekly sample collection, while strontium-90 (^{90}Sr) deposition is monitored with monthly collections at Kaitaia, Hokitika and Rarotonga. Lead-210 deposition is also monitored on a monthly basis as an adjunct to the ^{90}Sr measurements.



Radioactivity in milk: ^{90}Sr and ^{137}Cs concentrations, together with calcium and potassium levels, are monitored in dairy milk powders with monthly sample collection from three New Zealand regions: Auckland, Taranaki and Westland.

Environmental radiation: outdoor environmental gamma radiation levels are monitored continuously in Christchurch as part of the GERMON programme, using both real-time and integrating monitoring methods.

This report summarises results of the above measurements during 1993. Earlier reports in this series give information on terms of reference, reference levels and potential health hazard[1,2], and technical information and the design of the programme[3,4]. The history of fallout monitoring in New Zealand has recently been reviewed[5].

3. 1993 MONITORING RESULTS

Monitoring results for 1993 are summarised in Tables 1, 4 and 5.

Radioactivity units used throughout this report are becquerels (Bq), millibecquerels (mBq) and microbecquerels (μBq): 1 Bq = 1 nuclear transformation per second.

3.1 Atmospheric radioactivity

Atmospheric monitoring has two components — weekly monitoring with continuous sampling for contiguous 7 day periods, and the analysis of quarterly and annual air filter aggregates. The weekly monitoring provides short-term information on trends in aerosol radioactivity and early warning of any significant changes, while the aggregate filter analyses provide information on long-term trends in average levels of trace atmospheric constituents.

The monitoring is conducted at Kaitaia, Hokitika and Rarotonga where centrifugal fan pumps draw air through 500 cm^2 filters (Carl Freudenberg, type FA2311) at a flow rate of approximately 3 m^3/min, with a weekly sample volume of approximately 30 000 m^3. The filters are analysed by high-resolution gamma spectroscopy for gamma-emitting artificial and natural radionuclides; by gas-flow proportional counting for TBA; and radiochemically for ^{210}Pb and ^{210}Po. Quarterly and annual filter aggregates, representing air volumes of approximately 4×10^5 m^3 and 1.6×10^6 m^3 respectively, are analysed using high-resolution gamma spectroscopy.

The limits of detection for a fission product such as ^{137}Cs in the weekly, quarterly and annual filter analyses are 0.5, 0.07 and 0.02 μBq/m^3 (average concentration) respectively.

Atmospheric radioactivity monitoring results for 1993 are summarised in Table 1. Fluctuations in atmospheric radioactivity during 1993 and preceding years at the 3 monitoring sites are illustrated in Figs 1 - 3 which show 5-week moving-average levels. Annual average ^{7}Be, ^{210}Pb and TBA levels are tabulated for the period 1987 - 1993 in Table 2.

3.1.1 Artificial radionuclides

No particulate artificial radionuclides were detected by gamma spectroscopic analysis of the weekly air filters from any monitoring site during 1993.

Caesium-137 was not detected on quarterly air filter aggregates but was found at trace levels on the annual filter aggregates from Kaitaia and Hokitika, at annual average levels of 0.02 ± 0.01 μBq/m^3 and 0.05 ± 0.01 μBq/m^3, respectively. At Rarotonga the concentration was less than 0.02 μBq/m^3.

During 1993 the filter aggregate analyses were extended to earlier years to include analysis of yearly and quarterly aggregates of all filters collected at the Kaitaia site since high-volume sampling commenced in 1986. The measured annual average ^{137}Cs levels are listed in Table 3 and their decreasing trend from 0.25 μBq/m^3 in 1986 to 0.02 μBq/m^3 in 1993, as illustrated in Fig. 4, is discussed in section 4.1.

3.1.2 Total beta activity

Weekly average atmospheric TBA levels during 1993 were very similar at all 3 monitoring sites and ranged from 0.04 to 0.27 mBq/m^3 (Table 1) with an overall average of 0.15 mBq/m^3, similar to the 1992[6] average of 0.14 mBq/m^3.

Radionuclides which contribute to the measured TBA are considered in detail in section 4.2.

3.1.3 Natural radionuclides

Cosmogenic nuclides

The cosmogenic radionuclides which contribute a quantifiable population dose commitment are carbon-14 (^{14}C), ^{22}Na, ^{7}Be and tritium (^{3}H)[7]. Because of its very low beta particle energy, ^{3}H is not detected in the NRL monitoring programme but ^{7}Be and ^{22}Na can be measured on weekly or yearly-aggregated air filters and ^{14}C contributes to the measured TBA.

Beryllium-7 continued to be the most significant radionuclide detected on the air filters during 1993, with weekly average concentrations in the range 0.7 - 6.4 mBq/m^3, with an overall average of 2.9 mBq/m^3. The annual trend in ^{7}Be levels at Kaitaia and Hokitika continued to demonstrate the normal cycle of summer maxima and winter minima (Figs 1 and 2), while at Rarotonga levels were, as usual, more variable but continued to demonstrate a spring maximum (Fig. 3).

Sodium-22 was detected on the yearly aggregates of air filters at annual average concentrations of 0.22 - 0.31 μBq/m^3 (Table 1). The significance of the ^{22}Na measurements is considered further in Section 4.2.

Radon decay products

Lead-210 and ^{210}Po were measured on the weekly air filters and levels were similar at all 3 monitoring sites, as indicated in Table 1. Lead-210 levels ranged from 3 to 200 μBq/m^3, with an overall average 50 μBq/m^3. Polonium-210 levels ranged from less than 1 to 17 μBq/m^3 with an overall average of 7 μBq/m^3. The ^{210}Po/^{210}Pb activity ratios are discussed further in section 4.3.

3.2 Radioactive deposition

Results of measurements of TBA, ^{90}Sr and ^{210}Pb deposition in New Zealand and Rarotonga are summarised in Table 4.

3.2.1 Total beta activity

The total TBA deposition for 1993 at Hokitika was 172 ± 7 Bq/m^2, with 260 cm of rainfall, similar to that recorded in 1992[6]: 149 Bq/m^2 with the same rainfall. The mean weekly deposition was 3.2 Bq/m^2, with a range of 0.3 - 17.7 Bq/m^2. The mean weekly concentration of beta emitting nuclides in rainwater at Hokitika was 0.16 Bq/L, with a range of 0.02 - 2.71 Bq/L.

At Rarotonga the total deposition was 334 ± 7 Bq/m^2, with 184 cm of rain, similar (on a rainfall-normalised basis) to that recorded in 1992[6]: 214 Bq/m^2 with 139 cm of rain. The mean weekly deposition at Rarotonga was 6.3 Bq/m^2, with a range of 0.3 - 17.7 Bq/m^2. The mean weekly concentration of beta emitting nuclides in rainwater at Rarotonga was 1.7 Bq/L, with a range of 0.01 - 19.7 Bq/L.

3.2.2 Strontium-90

Strontium-90 deposition is measured in six-monthly aggregates of strontium recovered from the monthly rainwater collections.

At Kaitaia the total 1993 ^{90}Sr deposition continued to be at the limit of detection, 0.1 ± 0.2 Bq/m^2 (Table 4), as is typical of recent years. At Hokitika and Rarotonga the total deposit was less than 0.1 Bq/m^2.

These ^{90}Sr deposition results may be contrasted with TBA deposition which is of natural origin (as explained in Section 4.2), and with the peak ^{90}Sr deposition recorded in 1964 when the national average was 130 Bq/m^2.

3.2.3 Lead-210

Lead-210 and its decay product ^{210}Po are the most significant source of natural dietary radiation exposure due to terrestrial radionuclides (excluding ^{40}K), together

contributing over 80% of exposure from this source[7]. Lead-210 deposited from the atmosphere is easily extractable from soils[8] and is therefore likely to be a significant source of the ^{210}Pb in agricultural products and hence natural dietary radiation exposure, as discussed further in section 5.

Lead-210 deposition is measured together with that of ^{90}Sr. During 1993 monthly average ^{210}Pb depositions at Kaitaia, Hokitika and Rarotonga were 4.0, 9.6 and 2.8 Bq/m^2, with annual total depositions of 48, 115 and 33 Bq/m^2, and annual average rainwater concentrations of 0.05, 0.05 and 0.03 Bq/L respectively (the Rarotonga result was estimated on the basis of 6 months data only due to problems experienced in the analysis of samples from that site).

Lead-210, through its decay product bismuth-210 (^{210}Bi) also makes a significant contribution to deposited TBA, as described in Section 4.2.

3.3 Radioactivity in soil

Although annual deposition of artificial radioactivity is now at a very low level, measurable amounts of long-lived fission products remain in the soil due to the history of weapons testing. It is this accumulated radioactivity which is the source of the low levels of ^{90}Sr and ^{137}Cs still measurable in agricultural products.

Soil radioactivity is no longer routinely monitored in New Zealand but the cumulative amount of ^{90}Sr or ^{137}Cs in any area can be estimated on the basis of annual average rainfall[9]. During 1992 and 1993, however, many soil samples from various regions were analysed for ^{137}Cs under contracts related to soil erosion studies. Although this work was not part of the environmental monitoring programme it is of interest to record here a brief summary of the results.

Levels of ^{137}Cs in New Zealand soils were found to be in the range 300 - 1200 Bq per square metre of soil surface, at concentrations of up to 35 Bq/kg, with the ^{137}Cs levels being linearly dependent on local rainfall. The ^{137}Cs was found to still be retained in the upper 20 cm of the soil profile.

Strontium-90 levels would be expected to be about half of the ^{137}Cs levels (assuming a $^{137}Cs/^{90}Sr$ production ratio of 1.6 and a slightly greater degree of leaching of ^{90}Sr).

3.4 Radioactivity in milk

The presence of ^{137}Cs and ^{90}Sr in soil, as described above, leads to their presence in milk. Models describing their transfer through the soil-grass-milk foodchain in New Zealand have recently been published[10]. Results of milk monitoring during 1993 are summarised below and in Table 5.

3.4.1 Caesium-137

Caesium-137 levels in cows' milk were assessed by gamma spectroscopic analysis of 3-month aggregates of monthly samples. Results are shown in Table 5, with units of Bq per gram of potassium (Bq/g K) and Bq per kilogram of milk powder (Bq/kg).

The 1993 average ^{137}Cs levels were: Auckland 0.10 Bq/g K or 1.2 Bq/kg powder; Taranaki 0.27 Bq/g K or 2.7 Bq/kg powder; Westland 0.07 Bq/g K or 0.7 Bq/kg powder. The three-region mean was 0.15 Bq/g K or 1.5 Bq/kg powder, which is lower than the 1992 mean[6] of 0.21 Bq/g K or 2.7 Bq/kg powder.

3.4.2 Strontium-90

Strontium-90 levels in cows' milk were measured by radiochemical analysis of 3-month aggregated samples. Results are shown in Table 5, with units of Bq per gram of calcium (Bq/g Ca) and Bq/kg powder.

The 1993 average ^{90}Sr levels were: Auckland 0.02 Bq/g Ca or 0.24 Bq/kg powder; Taranaki 0.02 Bq/g Ca or 0.29 Bq/kg powder; Westland 0.03 Bq/g Ca or 0.34 Bq/kg powder. The three-region mean was 0.02 Bq/g Ca or 0.3 Bq/kg powder, which is similar to the 1992[6] mean of 0.03 Bq/g Ca or 0.4 Bq/kg powder.

3.5 Environmental radiation

Environmental gamma radiation levels, due to cosmic, atmospheric and terrestrial gamma radiation, were monitored continuously through 1993 using a monitor installed on the Laboratory roof. This monitoring is conducted primarily as part of the WHO/UNEP GERMON programme.

The radiation level was virtually constant during 1993 at 100 nanogray per hour, or 0.9 milligray per year, which is the same as the level reported in 1992[6].

An integrating measurement package comprising thermoluminescent dosemeters and films for 3-month exposure periods, provided by the WHO as part of the GERMON programme, is also used to assess environmental radiation levels. These packages are analysed quarterly by the WHO in France. Results of the integrating measurement and the continuous monitoring were similar during 1993.

4. DISCUSSION OF RESULTS

4.1 Trends in atmospheric ^{137}Cs levels

The trends in annual average atmospheric ^{137}Cs levels at Kaitaia since 1986 are illustrated in Fig. 4. The rapid decline in levels since 1986 raised speculation as to whether or not the detected ^{137}Cs was of Chernobyl origin (the Chernobyl reactor explosion occurred in April 1986), in apparent conflict with earlier indications[11] that Chernobyl debris did not reach New Zealand.

An analysis of the ^{137}Cs trend at Kaitaia, involving a comparison with trends observed in Australia (in the UK Atomic Energy Authority monitoring programme) revealed, however, that the detected ^{137}Cs originated in the Chinese nuclear weapons tests, particularly the final test of late 1980. Details of this study have been submitted for publication elsewhere and so are not reported here (the publication will be referenced in the next report). The study indicated that there was no ^{137}Cs input from Chernobyl, with the trend since 1986 being a continuation of the trend which would have been observed prior to 1986 if high-volume monitoring had been conducted then.

Projections regarding the atmospheric distribution of pollutants following the Chernobyl incident, and their potential impact on New Zealand, were based largely on information gained from the monitoring of nuclear weapons test debris. As a source of pollution the Chernobyl reactor explosion was, however, quite different to a nuclear weapon — the explosion caused a significant pollutant release, with a ^{137}Cs yield approximately equivalent to a 6 megatonne nuclear (fission) explosion, but it was a low altitude release occurring over a time interval of several days, rather than a high-altitude, rapid release as in the case of a weapon detonation.

Measurements of atmospheric TBA during 1986[11] and following years appeared to confirm the projection that no significant Chernobyl radioactivity would be detectable in New Zealand. However, the retrospective study of the trend in atmospheric ^{137}Cs levels provided the first concrete evidence that Chernobyl debris did not reach New Zealand in measurable quantities, as either tropospheric or stratospheric fallout. Inter-hemispheric atmospheric distribution models developed from weapons test data thus seem to have been validated in their application to pollution from other sources.

4.2 Atmospheric beta activity

Recent annual reports have indicated an apparent discrepancy between atmospheric TBA and measured concentrations of contributing radionuclides. Since 1990 the annual average TBA at all 3 monitoring sites has been in the range 0.08 - 0.15 mBq/m^3 while the concentration of ^{210}Pb, which is obviously a beta activity contributor (due to its ^{210}Bi decay product, as described below), has been constant at about 0.06 mBq/m^3. The residual beta activity has been partly attributed[6] to natural ^{238}U and ^{232}Th decay products and ^{40}K but there was still an

unexplained residue. Now the analysis of annual aggregates of air filters and the high sensitivity of such measurements, as reported above, has provided further information on the contributions made by cosmogenic radionuclides in particular. The various sources of beta activity are described below.

Cosmogenic radionuclides

A range of radionuclides is produced in the atmosphere by cosmic radiation and those with half-lives greater than one day are listed in Table 6. Some of these radionuclides would make a negligible contribution to TBA because their half-lives are very long and activities therefore very low (^{10}Be, ^{36}Cl, also produced in very low yield[12] compared to the other nuclides listed), and the beta energy of ^{3}H is too low for detection by the NRL equipment. The remaining nuclides, ^{14}C, ^{35}S, ^{33}P and ^{32}P, would be expected to contribute to the measured TBA.

Sodium-22 was measured in the aggregate filter analyses described above with a 1993 annual average concentration at Kaitaia and Hokitika of 0.27 μBq/m^3. Average tropospheric activity ratios relative to ^{22}Na, ie, (nuclide activity)/ (^{22}Na activity), for ^{35}S, ^{33}P, ^{32}P have been calculated from published data[12] as follows: ^{35}S, 116; ^{33}P, 113; ^{32}P, 209. On the basis of the measured ^{22}Na concentration, average annual atmospheric concentrations of ^{35}S, ^{33}P and ^{32}P during 1993 would therefore have been approximately: ^{35}S, 35 μBq/m^3; ^{33}P, 33 μBq/m^3; ^{32}P 63 μBq/m^3, with a total activity of 131 μBq/m^3. The effect of the relatively low beta energies of ^{35}S and ^{33}P (Table 6) and the 4 - 5 day delay between sampling and analysis (to allow short-lived radon progeny to decay) would reduce the total measured beta activity of the 3 nuclides to an equivalent atmospheric concentration of about 80 μBq/m^3. In addition there would be a small contribution due to the ^{14}C content of organic and carbonate aerosols.

^{210}Pb and ^{210}Bi

Lead-210, with a half-life of 22.3 y, is a low-energy beta emitter (47 keV) and so makes little direct contribution to the measured TBA of air filters. Its decay product ^{210}Bi (half-life 5 d), on the other hand, has a beta energy of 1160 keV and is therefore easily detected. A limited number of measurements of atmospheric ^{210}Bi concentrations have been performed at the Hokitika monitoring site and the results indicate an average ^{210}Bi/^{210}Pb activity ratio of 1.0, with the two radionuclides being in radioactive equilibrium, as would be expected from the estimated aerosol age (Section 4.3). The average atmospheric concentration of ^{210}Bi during 1993 would therefore have been similar to that of ^{210}Pb or 52 μBq/m^3. The delay between sampling and analysis would reduce the contribution to the measured TBA to an equivalent atmospheric concentration of about 30 μBq/m^3.

Suspended dust

During 1993 and earlier years the weekly air filters were analysed for the natural radionuclides ^{226}Ra (decay product of ^{238}U), ^{232}Th and ^{40}K derived from suspended dust and sea spray. Their contribution to the measured beta activity was estimated

assuming a condition of radioactive equilibrium in the ^{238}U and ^{232}Th decay series. The average total contribution of the U and Th decay products and ^{40}K to the measured TBA during 1993 was estimated to be equivalent to an atmospheric concentration of approximately 20 $\mu Bq/m^3$.

Fission products

The annual average ^{137}Cs concentration at Kaitaia and Hokitika during 1993 was approximately 0.04 $\mu Bq/m^3$ and the ^{90}Sr concentration would be expected to be about 60% of this, or 0.02 $\mu Bq/m^3$ (assuming a ^{137}Cs:^{90}Sr production ratio of 1.6). The total contribution of the two fission products to the TBA would therefore have been roughly 0.06 $\mu Bq/m^3$, which is insignificant in relation to the contributions of naturally occurring radionuclides described above.

To summarise, the total contributions to measured atmospheric TBA during 1993 are estimated to be as follows:

Cosmogenic radionuclides (excluding ^{14}C)	0.08	mBq/m^3
^{210}Bi	0.03	mBq/m^3
Suspended dust	0.02	mBq/m^3
^{137}Cs and ^{90}Sr	0.00006	mBq/m^3

with a total contribution of about 0.13 mBq/m^3.

The overall average measured TBA was 0.15 mBq/m^3 and given the uncertainties and assumptions in the above modelling it seems likely that atmospheric TBA is due almost entirely to natural cosmogenic radionuclides, ^{210}Bi and suspended dust, with contributions of about 60%, 25% and 15% respectively.

It follows that the measured TBA deposition would also be due to these naturally occurring radionuclides. Lead-210 deposition during 1993 (Table 4) was 44% of the measured TBA at Hokitika and an estimated 10% at Rarotonga.

4.3 Aerosol age

The $^{210}Po/^{210}Pb$ activity ratios (Section 3.1.3) were used to estimate aerosol age before collection, based on ^{210}Po ingrowth and assuming no alternative source of ^{210}Po. Mean aerosol ages (± standard error) during 1993 at Kaitaia, Hokitika and Rarotonga were estimated to be 30 ± 4 d, 41 ± 8 d and 46 ± 7 d, respectively. The similarity of these aerosol age estimates indicates that in regard to atmospheric ^{210}Pb, New Zealand can be considered as a remote oceanic site like Rarotonga, with negligible local input.

The annual trends in atmospheric ^{210}Pb levels at Rarotonga (Fig. 3) indicate a similar periodicity to those of ^{7}Be and TBA with spring maxima, suggesting that the ^{210}Pb is also largely of stratospheric (or at least upper tropospheric) origin. In New Zealand the annual variations in ^{210}Pb levels during the period 1987 - 1993 (Figs 1 and 2) were, on occasions, similar to those of the TBA but with little

similarity to ^{7}Be variations, although a greater stratospheric component would be expected at the higher New Zealand latitudes. It seems likely that ^{210}Pb in New Zealand is of mixed origin with tropospheric transport from Australia being a possible contributing source.

4.4 Trends in ^{7}Be and TBA levels

In the 1992 report[6] it was suggested that a downward trend in atmospheric ^{7}Be levels may be becoming evident at Kaitaia and Hokitika, coinciding with increased solar activity. If there was a trend, its continuation was not evident in 1993 when annual average levels were similar to those of the previous year, as indicated in Table 2. The variability of the 1987 - 1993 data (Table 2) is such that there are no statistically-significant differences between annual average concentrations during that period so it is doubtful that any downward trend existed.

Atmospheric TBA has displayed a significant increasing trend since 1987 as indicated in Figs 1 - 3 and Table 2, and noted earlier[6], though levels now appear to have levelled off with no significant difference between 1992 and 1993 results (Figs 1 - 3 and Table 2). This trend in TBA levels is presumably the result of a cyclic natural phenomenon, possibly involving cosmic radiation and solar activity cycles and their influence on cosmogenic radionuclide production rates.

5. RADIATION EXPOSURE FROM FALLOUT IN DIET

Monitoring results indicate virtually all aerosol radioactivity in the atmosphere is now of natural origin and due mainly to ^{210}Pb, ^{210}Po and cosmogenic radionuclides. Human radiation exposure from this source results mainly from the deposition of these nuclides and their resulting incorporation into diet.

Exposure due to the cosmogenic radionuclides is relatively uniform world-wide[7] with ^{14}C making the greatest contribution of about 12 μSv/y. The only other cosmogenic nuclides which give a quantifiable dose contribution are ^{22}Na, ^{7}Be, and ^{3}H with a total contribution of about 0.2 μSv/y.

While the distribution of cosmogenic radionuclides may be relatively uniform world-wide, atmospheric levels and deposition of natural ^{210}Pb and ^{210}Po may vary markedly depending on local geography. Atmospheric ^{210}Pb concentrations in New Zealand, for example, as reported here for 1993, are about one tenth of the level assumed in dose assessments in continental Northern Hemisphere areas[7].

In assessing dietary dose commitments due to ^{210}Pb and ^{210}Po deposition in New Zealand, estimates by UNSCEAR[7] (United Nations Scientific Committee on the Effects of Atomic Radiation) of concentrations of natural radionuclides in various diet components in relation to their atmospheric concentration were used to derive atmosphere-diet transfer factors (Bq/kg_{diet} per Bq/m^{3}_{air}) as indicated below. The New Zealand diet composition used in the dose assessment was based on that

published by Dick et al[13], with annual consumption data as listed below. The dose assessment was made assuming the mean annual atmospheric concentrations of 0.05 mBq $^{210}Pb/m^3$ and 0.007 mBq $^{210}Po/m^3$ as measured during 1993.

Diet component	Consumed kg/y	^{210}Pb transfer, Bq/kg_{diet} per Bq/m^3_{air}	^{210}Po transfer Bq/kg_{diet} per Bq/m^3_{air}	diet ^{210}Pb Bq/y	diet ^{210}Po Bq/y
Grain, cereals	120	200	2000	1.2	1.7
Meat	100	160	1200	0.8	0.8
Fish	20	400	40000	0.4	6.0
Dairy products	190	80	1200	0.8	1.6
Vegetables	210	60	600	0.6	0.9
Drink	700	20	100	0.7	0.5
			Total:	**4.5**	**11.5**

Total annual average dietary consumptions of ^{210}Pb and ^{210}Po were 4.5 and 11.5 Bq respectively. These intake values were converted to effective dose commitments using factors derived from ICRP[14] (International Commission on Radiological Protection) recommendations: 1.0 μSv/(Bq ^{210}Pb intake) and 0.2 μSv/(Bq ^{210}Po intake). Resulting estimated dietary dose commitments were:

^{210}Pb	4.5 μSv/y
^{210}Po	2.3 μSv/y

Although drinks are generally included in the above diet composition, the consumption of rainwater as normal drinking water represents a special case. The average concentration of ^{210}Pb in rainwater at Kaitaia and Hokitika during 1993 was 0.05 Bq/L, while the ^{210}Po concentration, estimated from the mean aerosol washout ratio (9.2×10^5 Bq/m^3_{rain} per Bq/m^3_{air}) measured at Hokitika[15] was 0.006 Bq/L. Assuming a consumption of 1 L/day, the annual intake in drinking water and resulting effective dose commitments were:

^{210}Pb	18 Bq/y	18 μSv/y
^{210}Po	2 Bq/y	0.4 μSv/y

Consumption of rainwater as normal drinking water would therefore be the greatest source of ^{210}Pb exposure. People in this category could be regarded as the "critical group" for this radiation exposure route. The measured rainwater concentration of ^{210}Pb was the same at Kaitaia and Hokitika so it is likely that the above drinking water dose commitment would apply in most areas of New Zealand. It is possible also that people consuming relatively large amounts of shellfish may receive a higher dose commitment due to ^{210}Po which is known to be concentrated in shellfish organs.

In order to put this natural fallout radiation exposure into perspective, the above dose estimates may be compared with estimated dose commitments due to dietary ^{90}Sr and ^{137}Cs resulting from weapons test fallout. Maximum fallout levels were

recorded in 1964 and 1965[1,2] and the dietary dose commitments peaked in 1965 at estimated[10] levels of:

^{90}Sr 2 μSv/y
^{137}Cs 25 μSv/y.

Annual dietary ^{210}Po exposure is therefore similar to the peak ^{90}Sr dose commitment, while dietary ^{210}Pb exposure is twice this level. Drinking water exposure to ^{210}Pb may give an annual effective dose commitment similar to the peak ^{137}Cs dose commitment, and an order of magnitude greater than the peak ^{90}Sr level. The present total dietary dose commitment[10] due to deposited ^{90}Sr and ^{137}Cs is about 2 μSv/y which is considerably less than the exposure due to natural ^{210}Pb and cosmogenic ^{14}C.

6. CONCLUSION

Measurements of artificial radioactivity levels in the atmosphere and rainwater in New Zealand and Rarotonga during 1993 indicated that the atmosphere contained only residual traces of global weapons test fallout, with no detectable influx of fresh fission products. Levels of ^{90}Sr and ^{137}Cs in milk continued to be measurable at trace levels only. The 1993 results were very similar to those reported for 1992[6] as indicated in the comparison of New Zealand site averages below:

	1993	1992	
TBA in air	0.15	0.14	mBq/m^3
TBA in rain	172	149	Bq/m^2
^{90}Sr in rain	0.1	0.1	Bq/m^2
^{90}Sr in milk powder	0.3	0.4	Bq/kg
^{137}Cs in milk powder	1.5	2.7	Bq/kg

Caesium-137 was detected on yearly aggregates of air filters at Kaitaia and Hokitika during 1993 at an average level of 0.04 $\mu Bq/m^3$. A retrospective analysis of filter aggregates for earlier years indicated that the detected ^{137}Cs was of Chinese weapon test origin with no input from the Chernobyl disaster, validating atmospheric pollution dispersion models used earlier to predict that radioactive debris from the Chernobyl reactor were not likely to reach detectable levels in New Zealand.

Atmospheric radioactivity was dominated by naturally occurring radionuclides, with cosmogenic ^{7}Be making the greatest contribution to aerosol radioactivity. Atmospheric TBA was made up of contributions from cosmogenic radionuclides (60%), $^{210}Pb/^{210}Bi$ (25%) and suspended dust (15%). Aerosol $^{210}Po/^{210}Pb$ activity ratios indicated aerosol ages of 30 - 40 days in New Zealand.

During 1993 the trend of summer maximum and winter minimum ^{7}Be levels continued in New Zealand while the gradual upward trend in atmospheric TBA

seems to have ceased with levels measured in 1993 being similar to those recorded in 1992.

Accumulated caesium-137 is present in New Zealand soils at concentrations of 300 - 1200 Bq per square metre of soil surface and is still largely retained in the upper 20 cm of the soil profile.

Radiation exposure to fallout in diet is due mainly to natural cosmogenic radionuclides (particularly ^{14}C), and ^{210}Pb and ^{210}Po. In New Zealand, fallout ^{210}Pb and ^{210}Po in diet contribute estimated annual average effective dose commitments of 4.5 and 2.3 μSv respectively, compared to the total dose commitment of ^{90}Sr and ^{137}Cs, of about 2 μSv/y. The consumption of rainwater as normal drinking water would increase the ^{210}Pb contribution to about 22 μSv/y.

7. OTHER ENVIRONMENTAL WORK

7.1 Quality assurance

Quality assurance is an essential feature of any measurement laboratory's operations. The National Radiation Laboratory has for many years participated in international analytical intercomparisons performed by agencies in other countries including the WHO, IAEA, EPA and more recently SPERA.

During 1993 the Laboratory participated in 11 intercomparisons involving 45 analyses. One of these was organised by the Marshall Islands Nationwide Radiological Study and included only 4 laboratories, compared with the EPA intercomparisons which normally include about 170 laboratories worldwide.

Results for 1993 are presented in Table 7 which lists known and measured values.

Analytical performance was generally very satisfactory with a mean measured/known ratio of 0.96. The only significant problem encountered was with a particular ^{226}Ra in water analysis, and the analytical procedure for such analyses is being revised.

7.2 Commercial and advisory services

There was a continuing demand for export certificates and radioactivity tests in 1993. During the year 464 export certificates were issued and 328 commercial analyses performed.

The number of enquiries in 1993 from members of the public and commercial interests regarding radioactivity in consumer products and foodstuffs and environmental issues was normal.

7.3 Publication

During 1993 a major review of milk contamination monitoring results was completed[10].

ACKNOWLEDGEMENT

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Table 1: A summary of atmospheric radioactivity levels measured during 1993

Monitoring site:	KAITAIA	HOKITIKA	RAROTONGA

Atmospheric radioactivity, weekly measurements

TBA

Range, mBq/m^3	0.05 - 0.25	0.04 - 0.27	0.05 - 0.27
Mean, mBq/m^3	0.15	0.14	0.15

Cosmogenic ^{7}Be

Range, mBq/m^3	1.6 - 5.9	1.2 - 4.4	0.7 - 6.4
Mean, mBq/m^3	3.2	2.4	3.2

Radon decay products

^{210}Pb, range $\mu Bq/m^3$	9 - 200	3 - 110	3 - 110
mean $\mu Bq/m^3$	58	46	46
^{210}Po, range $\mu Bq/m^3$	1 - 15	<1 - 17	1 - 17
mean $\mu Bq/m^3$	7	6	7

Atmospheric radioactivity, annual measurement: annual average concentration

Fission product ^{137}Cs

^{137}Cs, $\mu Bq/m^3$	0.02 ± 0.01	0.05 ± 0.01	< 0.02

Cosmogenic ^{22}Na

^{22}Na, $\mu Bq/m^3$	0.31 ± 0.03	0.22 ± 0.02	0.28 ± 0.03

Table 2: Annual average atmospheric concentrations of ^{7}Be, TBA and ^{210}Pb measured at Kaitaia, Hokitika and Rarotonga, 1987 - 1993

Year	Kaitaia			Hokitika			Rarotonga		
	^{7}Be	TBA	^{210}Pb	^{7}Be	TBA	^{210}Pb	^{7}Be	TBA	^{210}Pb
1987	2.74 ± 0.81	0.07 ± 0.04		2.25 ± 0.65	0.06 ± 0.03		3.14 ± 1.00	0.07 ± 0.03	
1988	3.94 ± 1.29	0.10 ± 0.04		2.89 ± 0.91	0.09 ± 0.04		2.84 ± 1.46	0.06 ± 0.04	
1989	2.97 ± 1.15	0.10 ± 0.04	0.056 ± 0.032	2.43 ± 1.05	0.09 ± 0.04	0.062 ± 0.034	2.28 ± 1.24	0.08 ± 0.04	0.049 ± 0.032
1990	2.87 ± 0.91	0.11 ± 0.04	0.067 ± 0.030	2.21 ± 0.79	0.08 ± 0.03	0.052 ± 0.013	2.83 ± 1.11	0.08 ± 0.04	0.051 ± 0.023
1991	2.89 ± 0.92	0.12 ± 0.05	0.056 ± 0.032	2.13 ± 0.75	0.09 ± 0.04	0.051 ± 0.039	3.05 ± 1.32	0.11 ± 0.07	0.051 ± 0.029
1992	2.92 ± 0.94	0.14 ± 0.06	0.056 ± 0.035	2.35 ± 0.74	0.13 ± 0.05	0.048 ± 0.036	3.40 ± 1.02	0.14 ± 0.06	0.055 ± 0.036
1993	3.21 ± 1.04	0.15 ± 0.04	0.058 ± 0.033	2.42 ± 0.69	0.14 ± 0.04	0.046 ± 0.030	3.17 ± 1.21	0.15 ± 0.05	0.046 ± 0.023

Annual means ± 1 standard deviation

Table 3: Annual average atmospheric ^{137}Cs concentrations at Kaitaia determined by analysis of aggregated air filters, together with 95% confidence intervals

Year	Annual average ^{137}Cs level $\mu Bq/m^3$
1986	0.25 ± 0.03
1987	0.14 ± 0.03
1988	0.07 ± 0.02
1989	0.04 ± 0.02
1990	0.03 ± 0.01
1991	0.02 ± 0.01
1992	0.02 ± 0.01
1993	0.02 ± 0.01

Table 4: A summary of ^{90}Sr, ^{210}Pb and total beta activity deposition measurements at Kaitaia, Hokitika and Rarotonga during 1993

	Deposited radioactivity (Bq/m^2)		
	Kaitaia	Hokitika	Rarotonga
TBA		172 ± 7	334 ± 7
^{90}Sr	0.1 ± 0.2	< 0.1	< 0.1
^{210}Pb	48	115	33
Rainfall, cm	111	260	184

Table 5: Caesium-137 and strontium-90 levels in cows' milk during 1993

Results are expressed as Bq ^{137}Cs per gram potassium, Bq ^{90}Sr per gram calcium, and as Bq per kilogram of milk powder, for each quarter year period in the 3 regions. The 95% confidence intervals are of the order of 10% and 20% of the stated results for ^{137}Cs and ^{90}Sr respectively.

Caesium-137

Qtr	Auckland		Taranaki		Westland	
	Bq/g K	Bq/kg	Bq/g K	Bq/kg	Bq/g K	Bq/kg
1	0.18	2.3	0.35	3.4	0.07	0.9
2	0.07	0.7	0.26	2.6	0.13	1.1
3	0.07	0.9	0.21	1.9	<0.04	<0.4
4	0.06	0.8	0.26	2.8	0.05	0.5
Mean	0.10	1.2	0.27	2.7	0.07	0.7

Strontium-90

Qtr	Auckland		Taranaki		Westland	
	Bq/g Ca	Bq/kg	Bq/g Ca	Bq/kg	Bq/g Ca	Bq/kg
1	0.02	0.21	0.02	0.26	0.02	0.32
2	0.01	0.18	0.02	0.27	0.03	0.34
3	0.02	0.29	0.03	0.33	0.03	0.35
4	0.02	0.28	0.02	0.29	0.03	0.34
Mean	0.02	0.24	0.02	0.29	0.03	0.34

Table 6: Cosmogenic radionuclides normally present in the atmosphere[10]

Nuclide	Half-life	Decay mode	Beta energy, keV
^{10}Be	1.6×10^6 y	β	556
^{7}Be	53.3 d	EC, γ	
^{36}Cl	3.0×10^5 y	β	709
^{14}C	5730 y	β	156
^{32}Si	104 y	β	225
^{3}H	12 y	β	18
^{22}Na	2.6 y	β^+, γ	
^{35}S	87.5 d	β	167
^{33}P	25.3 d	β	248
^{32}P	14.3 d	β	1710

β beta emission
EC electron capture
β^+ positron emission
γ gamma emission

Table 7: A summary of performance in international analytical intercomparisons during 1993

The table compares the Laboratory's analytical result and the "known" value as stated by the organiser. Units were varied and are not given in the table.

Organiser	Sample	Type	Nuclide	NRL mean	Known
EPA	Air filter	Alpha	TAA	32	30
		Beta	TBA	69	69
			Sr-90	24	25
		Gamma	Cs-137	18	18
EPA	Milk	Beta	Sr-90	13	15
		Gamma	I-131	102	100
			Cs-137	12	15
			K-40	1720	1750
WHO	Wine	Gamma	Cs-134	195	191*
			Cs-137	477	480*
			K-40	17	18*
EPA	Water	Beta	TBA	145	155*
			Sr-90	28	29
		Alpha	TAA	95	95
			Uranium	28	29
			Ra-226	12	25
		Gamma	Cs-134	23	27
			Cs-137	33	32
			Co-60	39	39
EPA	Water	Alpha	Uranium	154	152
EPA	Water	Alpha	Uranium	74	76
EPA	Water	Gamma	Co-60	14	15
			Zn-65	104	103
			Ru-106	96	119
			Cs-134	5	5
			Cs-137	5	5
			Ba-133	89	99
EPA	Water	Alpha	TAA	12	15
		Beta	TBA	27	43
EPA	Water	Gamma	I-131	113	117
EPA	Air filter	Alpha	TAA	20	19
		Beta	TBA	47	47
			Sr-90	17	19
		Gamma	Cs-137	10	9
Marshall Is.	Coral	Gamma	Cs-137	968	857*
				103	97*
				13	12*
			Co-60	120	105*
				17	17
				4	4
			Bi-207	7	7*
				4	4*
		Alpha	Pu-239	2375	2443*
				613	668*
				34	37

* mean of participating laboratories' results

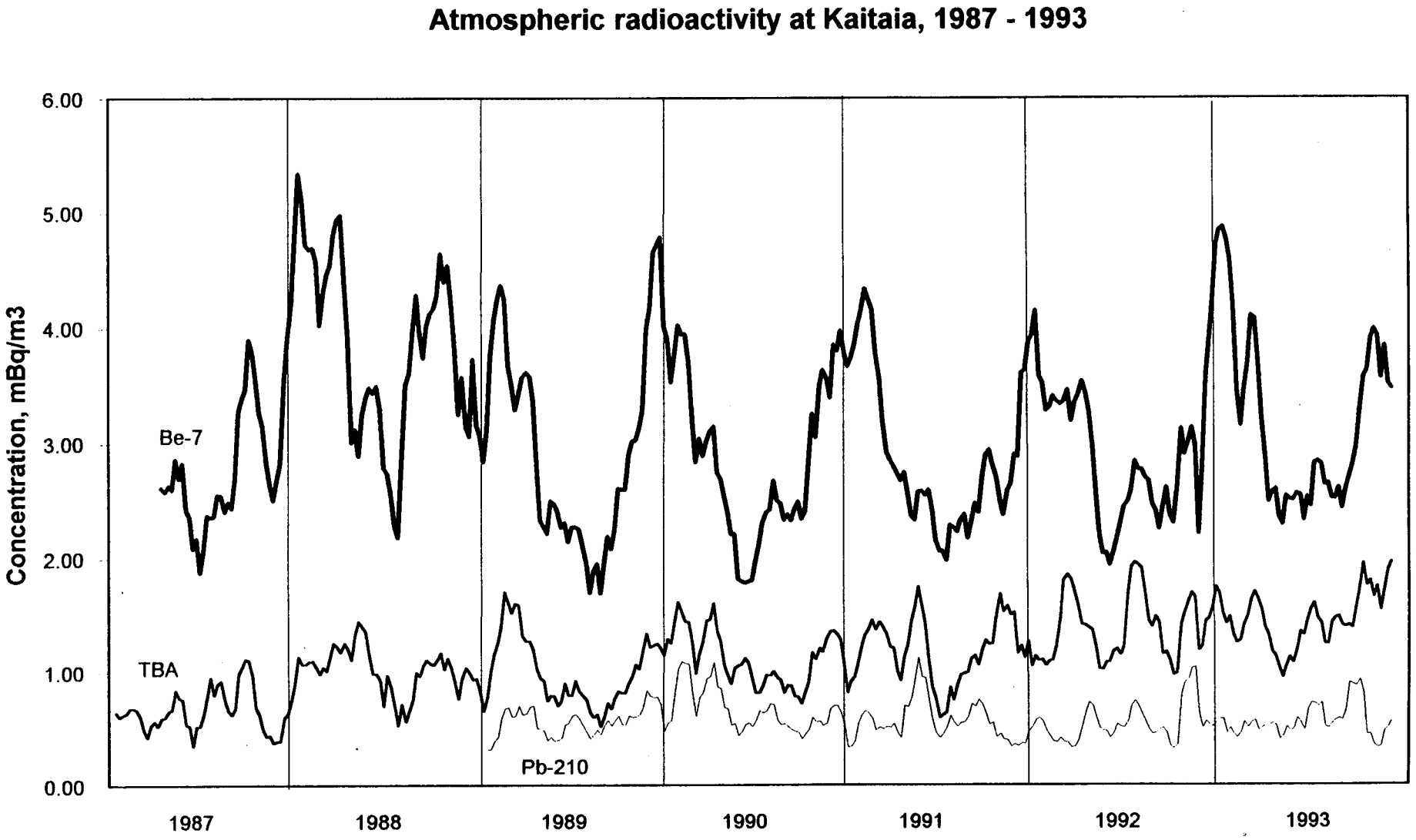

Fig 1. Five-week moving-average atmospheric concentrations of beryllium-7, lead-210 and total beta activity (TBA) at Kaitaia during the period 1987 to 1993

A scaling factor of 10 has been applied to ^{210}Pb and TBA levels

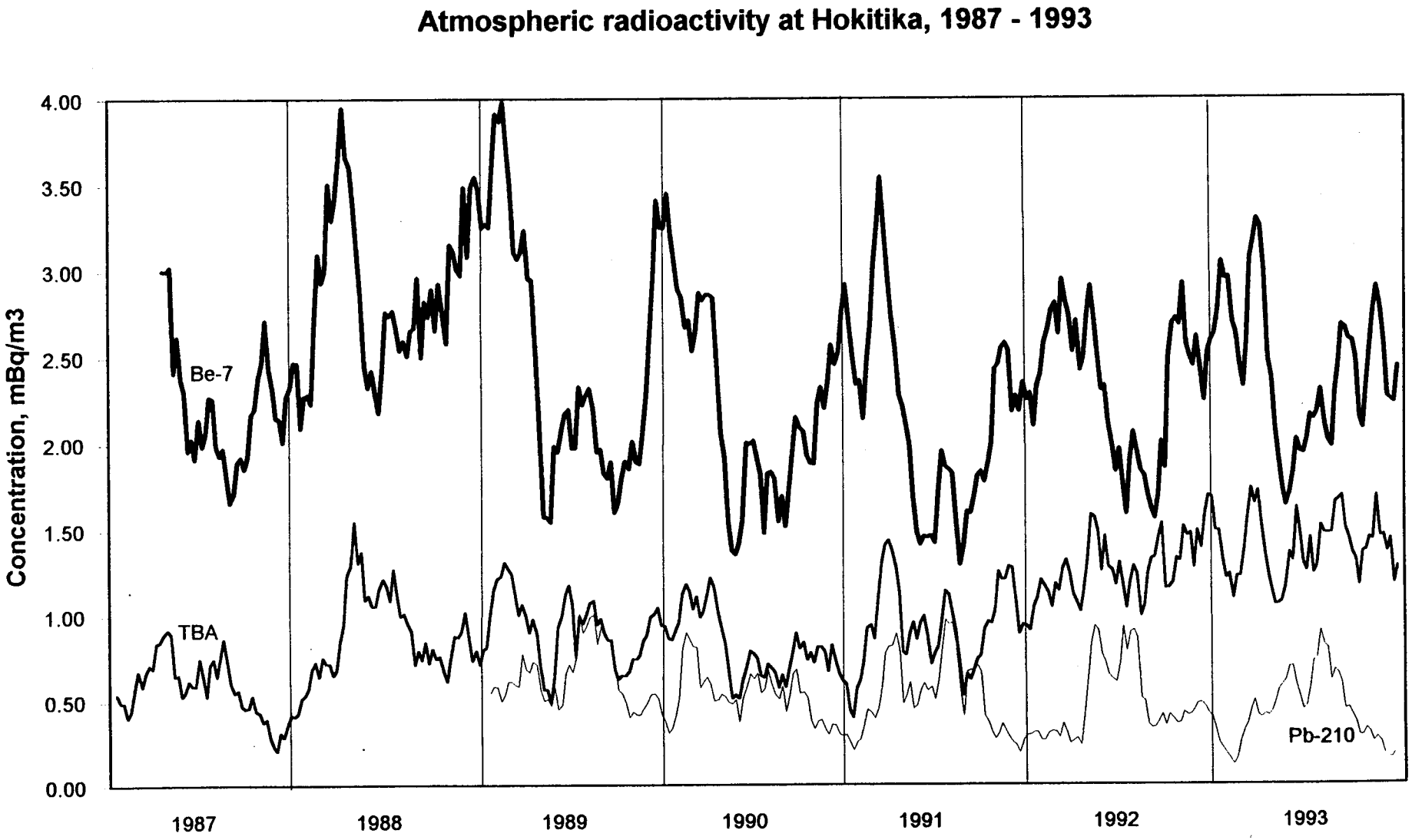

Fig 2. Five-week moving-average atmospheric concentrations of beryllium-7, lead-210 and total beta activity (TBA) at Hokitika during the period 1987 to 1993

A scaling factor of 10 has been applied to ^{210}Pb and TBA levels

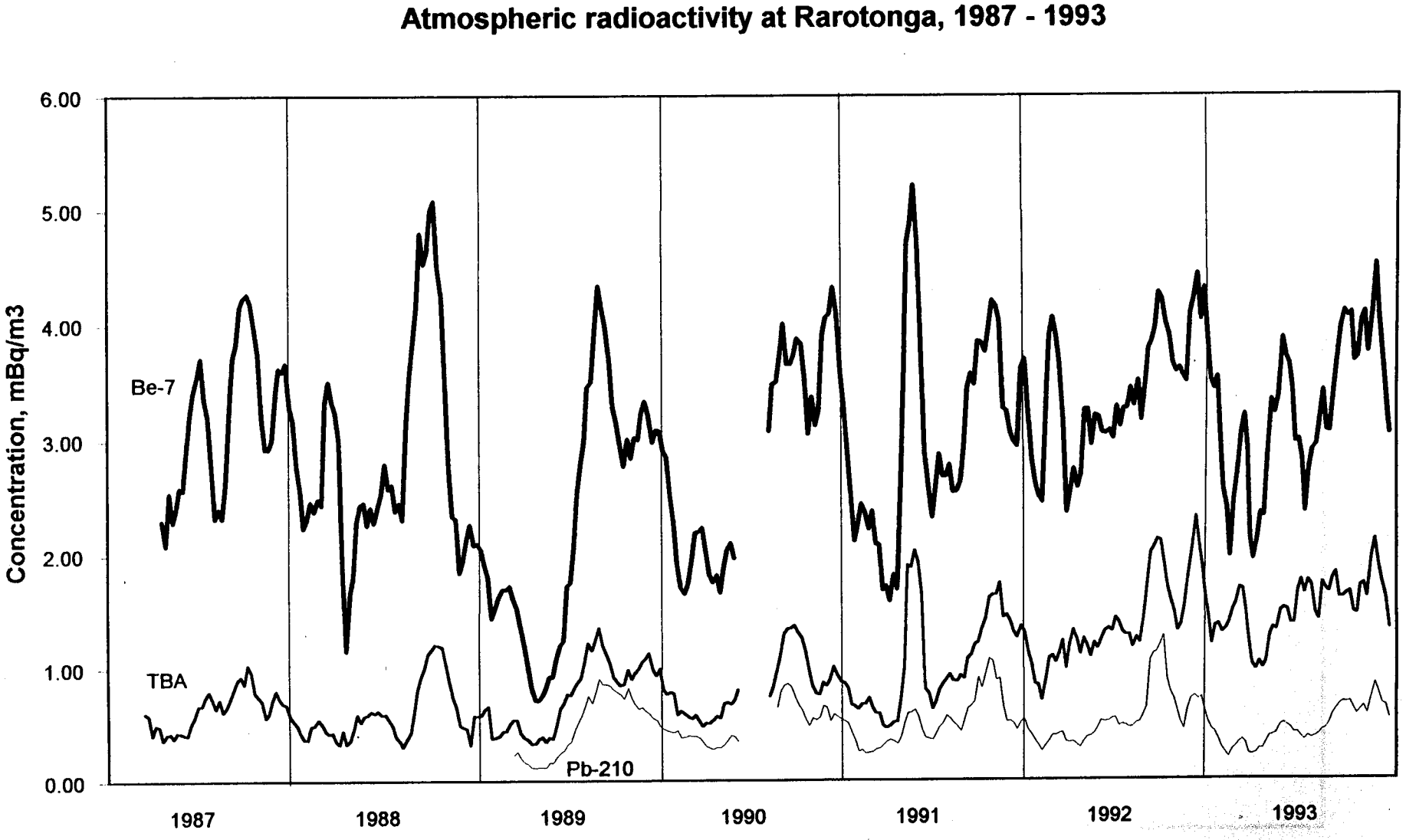

Fig 3. Five-week moving-average atmospheric concentrations of beryllium-7, lead-210 and total beta activity (TBA) at Rarotonga during the period 1987 to 1993

A scaling factor of 10 has been applied to ^{210}Pb and TBA levels

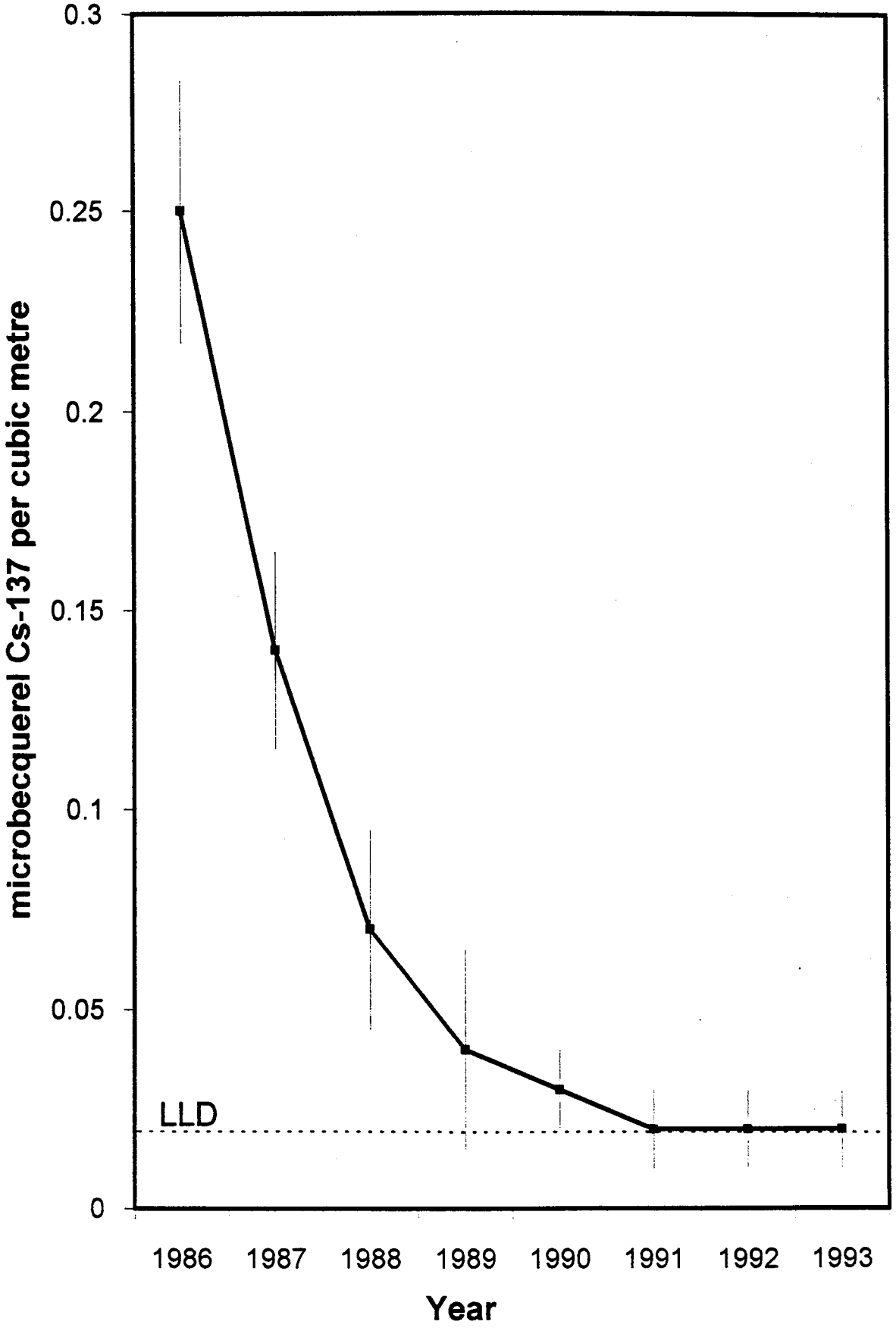

Fig 4. Annual average atmospheric ^{137}Cs concentrations at Kaitaia during the period 1986-1993

The 95% confidence intervals for each measurement and the limit of detection (LLD) are indicated