Chapter 1

Low-level counting

1.1 Introduction

The technique of low-level counting is used to solve a large variety of problems, ranging from daily control of contamination around nuclear plants to studies of fundamental processes in physics and astrophysics. For this work a variety of detectors and systems are used, from small semiconductor devices to large liquid scintillation counters.

Low-level counting was invented and given a name by Williard F. Libby in his pioneering work at the University of Chicago when he developed the radiocarbon (^{14}C) dating method in the late 1940s, for which he was awarded the Nobel prize in 1960. He had concluded from measurements of cosmic-ray produced neutrons in the stratosphere that ^{14}C would be produced and subsequently mix into the whole atmosphere. First he demonstrated the presence of ^{14}C in natural carbon by measuring isotopically enriched biomethane. He thereafter developed a very sensitive counting system for the determination of ^{14}C in natural carbon without enrichment. This system is shown in Figure 1.1. The most important elements of the technique are already found here: heavy shielding, anticoincidence counters, selection of radiopure materials and sample arrangement securing optimum counting efficiency and maximum sample size. The technique developed quickly. Libby's screen wall counter was replaced by gas proportional counters (requiring smaller samples), improved anticosmic counters were developed, an inner shielding layer of mercury and a neutron absorbing layer were added.

For almost a decade, low-level counting was limited to radiocarbon dating. In the mid 1950s, huge quantities of radioactive materials spread all over our globe from tests with hydrogen bombs in the atmosphere. Monitoring the fall-out created a compelling demand for expanding the low-level counting technique to a large variety of radioisotopes that required new types of detectors. The need for high sensitivity intensified in the 1960s with increasing interest in cosmogenic radioactivity in surface layers of the Earth and of meteorites, and in studies preparing for the measurement of prospective lunar samples.

In this introductory chapter, various general aspects of the low-level counting technique will be discussed. Most of the topics are treated in more detail in later chapters.

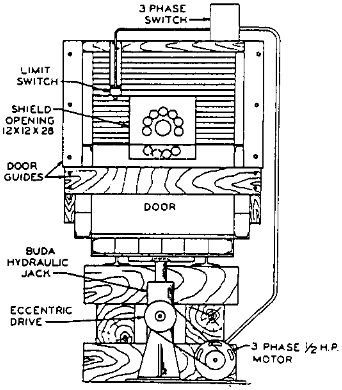

Fig. 1.1. Low-level counting started with this system. Libby's radiocarbon system with which he and his co-workers established the radiocarbon dating technique in 1947–1949.

1.2 Fields of application

The field of low-level counting started with radiocarbon dating where only gas counters were used. New radiation detectors were invented during this early period: scintillation counters with organic and inorganic crystals, liquid scintillation counters and, 15 years later, semiconductor radiation detectors. When these new detectors had reached maturity they were incorporated into systems based on the low-level counting technique, inherited from radiocarbon dating. To these radiometric methods, were later added mass spectrometric methods, where individual atoms of the radionuclides are counted rather than the few emitted decay particles, and sensitive optical methods (Chapter 16).

With new detectors and improved systems, the field of low-level counting gradually expanded. Today it is applied to a wide variety of studies, such as:

1. Radiocarbon dating.
2. Tritium in natural water.
3. Fall-out.
4. Contamination due to nuclear accidents.
5. Environmental control around nuclear establishments.
6. Distribution of primordial radioactivity in our environment.
7. General work with radioactive tracers.

8. Neutron activation analysis.
9. Tracing in hydrology and oil field exploration.
10. Mixing in the atmosphere and oceans.
11. Geochemical studies using primordial radioactive tracers.
12. Cosmogenic isotopes and extraterrestrial radioactivity.
13. Double beta decay and other rare processes.

This book is addressed to scientists working in these areas, except the last one.

It is difficult to define the field of low-level counting. Our interest starts when some special measures must be taken to increase the sensitivity of a system or a method, and it extends to sophisticated counting systems in general low-level laboratories. Expensive ultralow-level germanium spectrometers operating underground are on the border of the coverage of this book. They are, however, of interest to us as their development has brought us valuable information about the background sources of all low-level systems. Other, more specific systems, for example those for solar neutrino measurements, are not discussed here.

1.3 Detectors and systems

Today, the following detectors are of the greatest importance in low-level counting:

1. Gas proportional counters with internal samples.
2. Gas proportional and Geiger counters for external solid samples.
3. Liquid scintillation counters.
4. NaI scintillation counters.
5. ZnS scintillation counters.
6. Ge-diodes.
7. Si(Li) diodes.

These detectors are the core of low-level counting systems where various measures have been taken to increase their sensitivity, mainly to reduce their background count rate. For alpha detecting systems, the radiopurity of materials close to the detector is of primary importance. These materials are therefore carefully selected. For beta and gamma detection systems, various further measures are taken:

1. The detector is surrounded by a thick shielding layer, usually 10 cm of lead.

2. An extra inner layer of very pure material may be added: old lead, completely free of ^{210}Pb contamination, or electrolytically refined copper.
3. Sometimes there is a 5–10 cm inner layer of paraffin/boron to thermalize and absorb neutrons, produced by cosmic-ray protons in the shield.
4. Frequently the sample counter is surrounded by a system of anticosmic counters for suppressing the background contribution of cosmic-ray muons and protons.
5. Background pulses are sometimes eliminated through pulse shape or time sequence analysis.

1.4 Selection of detection system

When a radioisotope is to be measured we can frequently choose more than one method. Many aspects then come into consideration. The following points can be considered as a useful check-list:

1. Counting efficiency.
2. Background.
3. Energy resolution.
4. Number of detectors.
5. Accuracy needed.
6. Mass of sample available.
7. Size of counting samples.
8. Radionuclide enrichment in counting sample.
9. Number of samples.
10. Equipment available.
11. Cost of a new system.
12. Cost and work per measured sample.
13. Personal experience.

Let us take radon (^{222}Rn) as an extreme example. Radon is either measured directly or its progeny are separated and counted. When radon is measured, it is usually in secular equilibrium with its decay products (Section 17.8). The main methods for determining its concentration are the following:

1. The alpha activity of radon and its progeny can be measured in (1) a ZnS coated scintillation chamber (Lucas cell) or (2) in a liquid scintillation counter.

2. Radon decay products can be electrically precipitated on the window of (3) a gas proportional counter, or (4) on the window of a Si(Li) diode.

3. The radon decay products in air can be collected on a filter paper and the alpha or alpha+beta activity determined with (5) a thin window gas proportional counter. (6) with a ZnS scintillation counter, (7) with a liquid scintillation counter.

4. Radon can be adsorbed on charcoal in a canister and (8) the gamma activity determined, usually with a NaI scintillation unit. The radon can also be released from the charcoal and its alpha activity determined with a (9) a gas proportional counter, (10) in a Lucas cell or (11) by a liquid scintillation counter.

Available low-level methods for other radioisotopes are much more restricted. Two general examples will be discussed here, the measurement of (a) alpha active samples and (b) radioisotopes emitting both beta and gamma radiation.

Alpha active samples. In the measurement of alpha activity we can generally choose among four different detectors:

1. *Si(Li) diodes.* The radioisotope must be carefully separated from the matrix of the collected sample and plated out in a thin layer on a metal disk in order to secure high energy resolution (about 15 keV FWHM, see Section 8.5.3). The absolute detection efficiency is about 30%, mainly determined by the source-diode geometry. The background is very low, a few pulses per day. The diameter of the diodes is limited to about 60 mm (area 28 cm^2).

2. *Liquid scintillation counters.* The detection efficiency is practically 100%, and the energy resolution can be about 250 keV. The background can be very low, of the order of one pulse per hour. Chemical separation is necessary, the final step being dissolving the sample in the scintillating liquid.

3. *ZnS screen scintillation counting.* Scintillations of alpha particles impinging on a ZnS screen are detected with a photomultiplier tube. This method gives no information of the alpha energy and is mainly used for gross alpha counting of large, thick samples, for example filter papers. The background depends on the diameter of the ZnS screen, it can be about one count per hour for a 7 cm diameter screen.

4. *Window gas proportional flow-counters.* The samples are of the same type as those for ZnS scintillation counting and the sensitivity is similar. The counting system, frequently of the multidetector type, is simpler and more compact than the ZnS scintillating system. Good gas proportional counters have a background comparable to that of a ZnS system.

Beta/gamma active samples. Beta emitting radionuclides usually leave their daughter nuclei in an excited energy level. The nuclei get rid of the excessive energy through emitting a gamma-photon, usually instantly. These radioisotopes can be measured by three different methods:

1. *Germanium spectrometer*, detecting the gamma radiation. This is usually the preferred method, as large samples, in the order of one kilo, usually needing little or no preparation, can be measured because of the penetrating power of the photons. The high energy resolution leads to low background, gives positive identification of the radiation and allows the simultaneous determination of the concentration of individual nuclides in a mixture of radioisotopes. The detection efficiency of the germanium diodes is, however, rather low, typically a few percent.
2. *Liquid scintillation counter.* The beta detection efficiency is usually over 90% and the background of a low-level system is typically 0.5 cpm.
3. *Thin window gas proportional counter.* With thin samples the detection efficiency is typically 35–45%. The background depends on the diameter of the counter window, for a diameter of 25 mm it is about 0.2 cpm. These counting systems are usually of the multidetector type, where 4–8 samples are measured simultaneously.

1.5 Sensitivity comparison

When selecting a system for a particular job, one sometimes needs to compare quantitatively the sensitivity of two or more different systems, based on their background, counting efficiency and number of detectors. Earlier, the number of detectors was not taken into consideration, but with increasing use of multidetector systems, it is now natural also to include this parameter. Systems can be compared by the time needed to reach a given percent accuracy when a standard reference sample is measured. The relative standard deviation ε of net sample count rate is given by (see Chapter 18)

$$\varepsilon^2 = [(S+B)/T_S + B/T_B]/S^2 \tag{1.1}$$

where S and B are the net standard sample and background count rates, T_S and T_B the counting times of sample and background. The background count rate B is generally measured regularly and it will, after proper corrections have been made, stay constant during long periods. It is therefore usually known with better accuracy than is sought for the unknown sample. The term B/T_B in (1.1) can then be omitted. The sample counting time is then

$$T_S = (1 + B/S)/(\varepsilon^2 \cdot S) \tag{1.2}$$

The number of samples that can be measured per unit time to a given accuracy, i. e., the system's counting capacity, is a natural measure of the merit of the system. This number is inversely proportional to the counting time, so the *factor of counting capacity*, FCC, is defined by (Theodorsson 1991):

$$FCC = S/(1 + B/S) \tag{1.3}$$

The system giving the highest value of FCC is obviously most suitable. When the net count rate S of an unknown sample is large compared to the background B, FCC can be approximated by S (or rather by $S - B$). When, however, S is small compared to B, S^2/B gives a good approximation for the FCC. The most suitable system is then the one that gives the highest value of S^2/B. This is another form for the conventional *figure of merit* (FOM) of low-level systems, which is based on $1/\sqrt{T}$ rather than on $1/T$ or

$$FOM = S\sqrt{B} \tag{1.4}$$

For dimensional reasons, it is an unfortunate choice to base the widely used FOM on $1/\sqrt{T}$ rather than $1/T$.

When comparing a single sample detector system with a comparably priced multicounter system, it is natural to take into account the number of detectors N and not base the comparison on S and B of a single detector element. In this case the FCC of the multidetector system will have N times the FCC value of a single detector in the system.

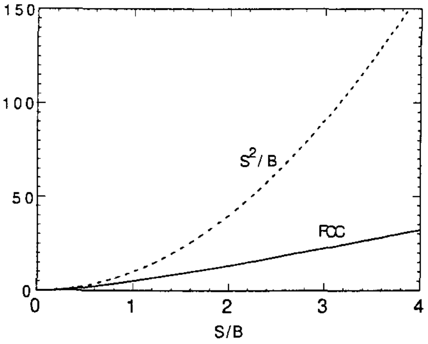

Fig. 1.2. Factor of counting capacity.

1.6 Recipes in low-level counting

In the design of low-level counting systems, as well as in their use, we depend on a number of recipes that we have inherited from our predecessors. Some of these were established by scientists short of time, setting up new low-level counting systems, wanting to start their studies with the least possible delay. Furthermore, the equipment used was expensive and cumbersome and offered limited possibilities for detailed data collection and processing compared to present day systems. Experimental work in low-level counting is usually time consuming because of the long counting times needed. Under these circumstances the scientists frequently had to settle on a procedure that seemed good enough, rather than to spend valuable time to find the best one. Various sectors of the low-level counting technique therefore still depend on lore and recipes. The description of the situation regarding scintillations cells for radon counting, as described by Cohen et al., (1983) is still valid. According to their description on the state of the technique, little information was available on these cells in the literature.

> "This does not imply that such information has not been collected, but such work has generally been done by groups with practical development goals and little taste for publication in the scientific literature. As a result the use of scintillation cells is surrounded by a lore propagated largely by private conversations and heavily influenced by isolated experience and hearsay evidence. While this evidence has been valuable, it is also somewhat variable among different practitioners and not always reliable."

We should take a critical attitude towards many of these recipes. Today, procedures would be simpler and systems better if the policy of the research laboratories of the Philips concern had guided development work in the low-level counting technique. The director of the laboratories, Hendrik Casimir, described it the following way:

> "It was an accepted policy of the research laboratories to try to really understand empirical procedures and not be satisfied with a recipe that worked well in practice, but was not understood."

Lore and recipes are, however, an inevitable part in all scientific work. In low-level counting we should show extra care in their use for the reasons given above. Five examples will be described in order to demonstrate the nature of these recipes, how they have been introduced, and what the consequences can be when we follow them uncritically. This is discussed in some detail here in the hope that it will encourage scientists to study critically some of our recipes.

1. Libby's group reported that contamination in the lead tested for the shield contributed 30 cpm to the background of the screen wall counter. Iron was therefore chosen and a 20 cm thick layer reduced the background to 10 cpm (Anderson et al., 1951). For almost twenty years, all dating laboratories using gas counters followed Libby's example and chose iron for the main shield, although lead has distinct advantages over iron, as it is easier to stack, offers greater flexibility and has higher density. With information we have today (Section 10.4.1), we can estimate crudely that Libby's lead must have had a contamination of about 8000 Bq of ^{210}Pb per kg of lead in order to contribute 30 cpm in a counter with a cathode area of 600 cm^2. This contamination level is improbable, as the ^{210}Pb concentration in recent lead is generally 100–500 Bq/kg,

although higher concentrations are met. A more probable explanation of these 30 cpm can be found in the paper describing the system (Anderson et al., 1951). Discussing the suppression of the background component due to external natural gamma radiation, the paper states:

> "Two inches of lead are sufficient for this purpose, but, unfortunately, the laboratory lead bricks themselves have been found to contribute about thirty cpm of gamma impurities. Various other materials were considered; iron was chosen as the best compromise between price, radiochemical purity, and high atomic number."

Two inches (5 cm) of lead are far from enough to suppress the contribution of external gamma radiation. Lead has a high atomic number and is very efficient in absorbing X-rays and gamma-photons with energies below one MeV, where photoelectron absorption is strong or predominating. For natural gamma radiation above 1 MeV, Compton scattering is the dominating gamma attenuating process, even in lead, and it is independent of the atomic number of the absorbing medium (see Figure 5.9). Actually, 12 cm of lead are needed to give the same gamma absorption as 20 cm of iron. It can be estimated at 300 cpm for Libby's unshielded counter from the information given in the paper. When the counter is shielded with 5 cm of lead, this count rate is reduced by a factor of about 12, so we should expect this component to have been 25 cpm, not far from the 30 cpm that was ascribed to contamination in the lead. Today we know that lead is actually the best shielding material, provided that lead with high ^{210}Pb contamination is avoided.

2. The second example of a hasty recipe is the procedure recommended by the Chicago group that developed the technique of assaying low levels of tritium in natural waters. Before the measurement, the water samples were enriched by a factor of 100 or more by electrolysis. In the description of the work it was stated that the volume reduction of the samples had been kept below 10 in order to avoid corrosion of the anode at high electrolyte concentration. It is easy to understand that scientists, needing to measure a limited number of samples in establishing the existence of cosmogenic tritium in natural waters, preferred to play safe in order to avoid possible complications due to corrosion. The samples were therefore enriched in three stages when the water was distilled between stages to remove excessive electrolyte. This recipe was followed for more than 20 years. Some laboratories, however, evaded one of the usual three stages by periodically adding water to the cells. Later it was shown that a volume concentration factor up to 300 did not affect the enrichment process and caused only mild corrosion.

3. The third example concerns the 5–10 cm thick layer of paraffin/boron inside the main shield in gas proportional counting systems, introduced by de Vries (1955) in the Groningen radiocarbon laboratory, in order to lower the neutron background component. This extra layer adds 30–40% to the mass of the main shield and therefore does not

come cheap. The Groningen laboratory only had a thin roof, giving minimum attenuation of the flux of cosmic protons that produce the neutrons in the shield. As discussed later, there is a serious doubt about the benefit of this layer in laboratories that have a few floor plates above them. Gas proportional counters in the dating laboratory at Trondheim, with four storeys above, have no neutron shield, but their background is lower than that of other counters of similar volume.

4. Graded shielding has been recommended for a long time for low-level counting systems, i.e., lining the inner surface of the main lead shield with a thin layer of cadmium and copper to absorb characteristic X-rays produced in the lead by cosmic rays. Most low-level germanium spectrometers have a shield of this type. It has recently been shown (Section 12.8) that this only helps when photons below 100 keV are being measured, as this layer increases the background in the energy range 100–500 keV by about 30%.

5. The final example concerns the anticosmic counter system inside the shield enclosing the sample detector. It eliminates the muon background contribution. It was pointed out by Scholz (1961), based on solid experimental work, that anticosmic counters on the outer sides of the shield would give the lowest background, as it would eliminate, in addition to the muon component, also a large part of the secondary radiation produced by muons and protons in the main shield. Furthermore, external anticosmic counters would reduce the mass of the main shield drastically as it makes the neutron absorbing layer superfluous and removes the anticosmic counters from the inner shield (Theodorsson and Heusser, 1991). Over 20 years elapsed until this proposal was implemented. This arrangement of guard counters is now used in a number of low-level and ultralow-level germanium spectrometers, giving high background reduction.

These examples show that we should not be satisfied with a recipe that works well in practice, but is not understood. We should continuously strive for a better understanding of all parameters and use new information for improving and simplifying our systems and procedures.

1.7 Reporting low-level systems

Today, there are still many loose ends in the low-level counting technique. As long as we cannot give a good quantitative estimate of all background components, we cannot design optimal systems.

Systematic studies, carried out with the aim of increasing our understanding of factors affecting background and counting efficiency of detectors, are naturally a major source of information. Descriptions of low-level counting systems, where their use is the main focus, are another important source of information. It is frustrating when the usefulness of good work of this type is seriously limited by not giving the value of important parameters, or when the results are described in a way that may be good enough for the purpose of the author, but not to those interested in the technique itself. It would help us in advancing the low-level counting technique if authors gave somewhat more detailed information about their systems. The following points can be considered as a check list for authors of articles on low-level counting to consult before they write their reports:

1. Overburden, mwe.
2. Thickness of all shielding layers.
3. Total mass of shield.
4. Dimensions and mass of detector.
5. Detection efficiency.
6. Background count rate.
7. Counting time.
8. X-axis of background spectra preferably linear.
9. Y-axis of background spectra in cpm/keV (not counts/channel), X-axis in keV (not channel number).
10. Match number of spectrum channels to energy resolution.
11. Quantitative reporting of ^{210}Pb in lead, Bq/kg.

A few remarks will explain the purpose of this list.

- A few floor plates above the laboratory room will significantly attenuate the nucleonic component of the cosmic rays. In order to be able to compare the background count rate of similar systems, we must know their overburden.
- In background comparisons, the mass of a germanium diode is more significant than its relative efficiency.
- The basis of the minimum detection-limit should be given, i.e., sample size, background and detection limit; the reader may want to calculate the detection efficiency using other values for the parameters or compare different systems or methods.
- The pulse height spectra of liquid scintillation counting systems are usually displayed on a logarithmic (usually uncalibrated) energy scale. This usually makes it difficult to use the information.

- Liquid scintillation systems usually have multichannel analyzers with 1000 channels. This does not match their poor energy resolution and often leads to a small number of pulses in each channel, with high scattering from one channel to the next. Fewer channels will display the results more comprehensively. It would be useful if the software offered the option of combining, for example, every four channels into a single one.
- The Y-axis of energy spectra is frequently not calibrated, but given in counts per channel. In order to make direct comparison possible the Y-scale should give cpm per keV. It is sometimes useful to know the width of the channels (in keV). It would also help if scientists would stick to one time unit, preferably minutes, not seconds, hours or days.
- Often it is useful to display the count rate of the spectra on a logarithmic scale, but only when the numbers span a wide range. In other cases it is better to use a linear scale.
- When the background of germanium spectrometers is reported, a spectrum gives the fullest information. Often we have to compress this information into a few numbers. It is then highly desirable to do this in a standardized way in order to facilitate comparison of the performance of different systems. It is recommended that the following count rates are given:
  (i) Total count rate from 50–1500 keV, in cpm
  (ii) The peak at about 150 keV, in cpm/keV
  (iii) The count rate at 500 and 1000 keV, cpm/keV.
  (iv) The fast neutron broadened peak at 691 keV, cpm.
- When scientists compare their spectra with those of other systems, an easy transfer of a spectrum into some general computer spreadsheet, such as EXCEL, is desirable. The multichannel analyzer software generally used today does not make this simple operation easy.

Finally, it should be noted that the recommendations given here will not significantly increase the length of the reports, but will substantially increase their usefulness.

1.8 Literature on low-level counting

A book was published in 1964 describing the technique of low-level counting. Today, it is naturally obsolete. Various review articles have been written, usually with main emphasis on a certain sector of the technique. Some of these are listed below, especially the more recent ones. The technique and its applications have been the subject of a number of specialized conferences, starting with a meeting of experts and scientists interested in radiocarbon dating in Copenhagen in 1954. Radiocarbon conferences have been held regularly ever since, now every three years, and their proceedings volumes have been published, in later years in Radiocarbon. The last Radiocarbon conference was held in 1994.

The first general conference on low-level counting and its applications was held by the International Atomic Energy Agency in Vienna in 1960. It has been followed up by a number of both general conferences as well as meetings for specialized sectors of the field. In most of these conferences, both techniques and applications have been discussed. Three general low-level counting conferences were held in Czechoslovakia, the first in 1975 and the last in 1989. The proceedings of these conferences have been published.

Conferences have regularly been held on the liquid scintillation counting technique, the last one in 1994. Low-level systems have been discussed at these meetings. The proceedings of the two last conferences have been published as special issues of Radiocarbon.

The International Committee for Radionuclide Metrology has held a number of conferences on various aspects of radioactivity measurements. Valuable information about low-level counting has been presented at some of these conferences. The latest, the Conference on Low Level Measurement Techniques, was held in October 1995, and its proceedings volume will be published in a special issue of Applied Radiation and Isotopes.

The University of Sevilla has arranged three times an international summer school for scientists using techniques of low-level counting. The lectures given there have been published by World Scientific.

One of the most active fields of low-level counting is the study of primordial and man-made radioactivity in our environment. A number of conferences have been held on the subject. Finally, health physics unions regularly hold conferences, usually centred on some main theme. For example, in 1993 the German Fachverband für Strahlenschutz e.V. held a conference on environmental radioactivity, radioecology and effects of radiation.

Recommended Reading

- *High Sensitivity Counting Techniques* D. E. Watt, D. Ramsden, Pergamon Press, 1964.
- *Low Level counting techniques*, H. Oeschger and M. Wahlen, 1975, Ann. Rev. Nucl. Sci., 25, 423-463. A broad general review of the technique, with emphasis on gas proportional counting.
- *Methods of Low-Level Counting and Spectrometry*, Proceedings of an IAEA symposium, IAEA 1981.
- *Radon Monitoring in Radioprotection, Environmental Radioactivity and Earth Sciences*. Proc. of an international workshop 1989, World Scientific 1990.
- *Proceedings of the ICRM Symposium on Low-Level-Radioactivity Measuring Techniques and Alpha Particle Spectrometry*, held in Monaco June 1991, ed. W. B. Mann, Pergamon Press, 1992.
- *Low-Level Measurements and their Applications to Environmental Radioactivity*, proceedings of the First International Summer School, held in Huelva, Spain, 1987. Ed. M. Garcia-Leon and G. Madurga, World Scientific, 1988.
- *Low-Level Measurements of Man-Made Radionuclides in the Environment*, Proceedings of the Second International Summer School, held in Huelva, Spain, 1990. Ed. M. Garcia-Leon and G. Madurga, World Scientific, 1991.
- *Low-Level Measurements of Radioactivity in the Environment*, Proc. of the Third International Summer School, held in Huelva, Spain, 1993. Ed. M. Garcia Leon and R. Garcia-Tenorio, World Scientific, 1994.
- *Umweltradioaktivität Radiokologie Strahlenwirkungen*, proceedings of the meetings of Fachverband für Strahlenschutz e.V. September 1993. Verlag TÜV Rheinland.
- *Low Radioactivity Background Techniques* G. Heusser, 1995. Ann. Rev. Nucl. Sci. A review article with a detailed analysis of background components and description of methods to suppress the background and description of some modern ultralow level studies. Gives 162 references.