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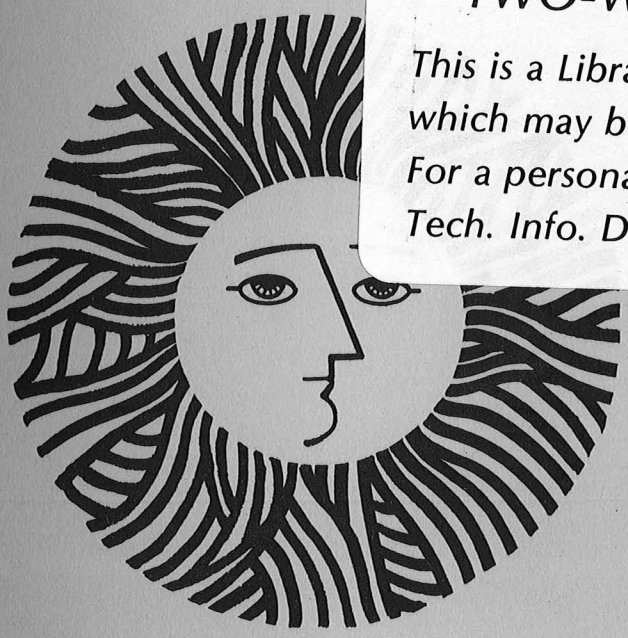

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ABSTRACT

As part of an overall Survey of Instrumentation for Environmental Monitoring, a survey was made of the instruments used for the determination of low energy beta radioactivity. Techniques commonly used are gas flow proportional counting, liquid scintillation counting, solid scintillation counting, and internal ionization chamber counting. Also used are solid state detector counting and radiochemical separation followed by counting using one of the preceeding techniques.

As a first step, the first four techniques were examined and compared with each other. The sensitivities of the techniques were compared on the basis of the detection limits quoted for instruments described in the technical and reviewed literature. The detection limits were then related to the occupational and public individual maximum levels for air and water given in Appendix B, Part 20 (Standards for Protection Against Radiation) of Title 10 of the Code of Federal Regulations. Attention is focussed primarily on the continuous monitoring of air for ^{3}H and ^{85}Kr, a medium energy β-emitter.

From a survey of U. S. commercial sources, it is clear that several continuous air monitoring instruments are readily available for measuring low energy β concentrations, even in presence of certain other activity, at occupational levels. However, these instruments do not typically have sensitivities comparable to the public individual levels. Moreover, their capabilities for giving results in real time and for differentiating among the radionuclides actually present is limited.

INTRODUCTION

Concurrent with an increasing use of energy, the realization has grown that environmental quality control is important. As a basis for such control, numerous monitoring programs have been developed, including a variety of instruments, some of which are elaborate and highly sophisticated. To aid monitoring organizations and analytical laboratories in choosing among the techniques and instruments available, Lawrence Berkeley Laboratory (funded by DOE and NSF) has for several years conducted a Survey of Instrumentation for Environmental Monitoring.[1] Instrumental methods covered are those suitable for monitoring and analyzing the conventional pollutants in air and water, as well as those for radiation measurements and analysis of biomedical samples.

As part of this work, a study was made of the instruments used for the measurement of low energy beta-emitters. The necessity for counting β particles lies in the fact that a number of natural or manmade radionuclides are β-emitters and either emit no gamma or X-rays, or emit them at so low a probability per decay as to make assay at low levels by photon detection difficult or impossible. Several such radionuclides with low β energies are listed in Table I, along with pertinent properties and sources of production. Not included are nuclides that decay by electron capture with the emission of conversion electrons. For this application semiconductor detectors can be particularly effective because of the low background attainable under the sharp conversion electron peaks.[2]

TABLE I. SELECTED LOW ENERGY β-EMITTERS AND PERTINENT PROPERTIES. DATA FROM REF. 3 EXCEPT WHERE NOTED.

Nuclide	Half-Life t 1/2	$\beta_{max.}$	$\beta_{ave.}$	Sources
^{3}H	12.33 yr	0.0186 MeV	0.0057 MeV	naturally occurring ternary fission[4] ^{6}Li (n,α)
^{14}C	5,730 yr	0.156 MeV	0.0467 MeV	naturally occurring ^{14}N (n,p)
^{35}S	87.4 d	0.167 MeV	0.061 MeV	^{34}S (n,γ)
^{79}Se	$\leq 6.5 \times 10^{4}$ yr	0.159 MeV	0.058 MeV[5]	fission
^{85}Kr[a]	10.7 yr	0.687 MeV	0.249 MeV[5]	fission ^{85}Kr (n,γ)
^{99}Tc	2.14×10^{5} yr	0.294 MeV	0.085 MeV[5]	fission
^{129}I	1.6×10^{7} yr	0.192 MeV	0.040 MeV[5]	fission
^{135}Cs	3×10^{6} yr	0.205 MeV	0.057 MeV[5]	fission daughter ^{135}Xe
^{147}Pm	2.6234 yr	0.225 MeV	0.0650 MeV	fission
^{151}Sm	90 yr	0.076 MeV	0.019 MeV[5]	fission
^{241}Pu	14.4 yr	0.0208 MeV	0.005 MeV[5]	multiple n-capture ^{238}U, ^{239}Pu, etc.

[a]Technically, ^{85}Kr is a moderate energy β-emitter,[6] but is included due to its widespread interest.

Of the nuclides in Table I the volatile and semivolatile long-lived manmade radionuclides ^{3}H, ^{14}C, ^{79}Se, ^{85}Kr, ^{99}Tc, ^{129}I and ^{135}Cs are of concern in connection with the operation of certain nuclear fuel cycle facilities. They are difficult and expensive to contain and once emitted to the environment many become permanent ecological constituents with both local and global distributions. Since volatile and semivolatile species are the most difficult to trap, they are the most likely to be released and transported long distances. Control of the release of these radionuclides to the environment and assessment of their long-term effects require sensitive sampling and measurement methods.[7] Measurements of both radionuclide levels and their chemical forms are needed. However, attention in this paper will focus on the measurement of the former.

Low energy β radioactivity, due to its limited penetration, tends to be an internal rather than external hazard. Internal exposure is usually assessed through estimation of the body burden based on measurements of the activity concentrations in biological samples (bioassay), or on measurements of activity in the body by use of external counters. Alternatively, the potential body burden or the potential dose equivalent in body organs is assessed by comparing measurements of the concentrations of radionuclides in air and water taken in by an individual with the maximum permissible concentrations that have been specified. NCRP Report No. 57 says, "In areas where frequent or continuous air contamination is likely, the air should be sampled continually during periods of personnel occupancy.* The potential for nonoccupational exposure to airborne contamination should be assessed by sampling the gaseous exhaust stream from the facility."[7]

Commonly used techniques for the measurement of β-emitters are gas flow proportional counting, liquid scintillation counting, solid scintillation counting, and internal ionization chamber counting. For the purposes of this paper, it is assumed that the reader is familiar with the details of the instrumentation and procedures involved in these counting techniques, and I will simply compare the different techniques in the measurement situations outlined later. For detailed information on techniques the reader is referred to the literature.[1, 2, 8-12] In this paper attention is focussed primarily on continuous air monitoring,

*In an alternative philosophy, the environment of personnel is controlled to levels well below the MPC and air sampling is done primarily to indicate when control is lost and remedial actions are needed.

although some mention will be made when a technique is applicable to continuous water monitoring as well.

DISCUSSION

First, a compilation of the detection limits quoted for instruments in the technical and reviewed literature will be presented to give an idea of the sensitivities obtainable with the aforementioned techniques. Then the results of a survey of U. S. commercial sources for off-the-shelf instruments will be presented to determine the extent to which commercially available equipment is able to measure low energy β-emitters at the various maximum permissible concentrations allowed by Appendix B of Title 10, Part 20 of the Code of Federal Regulations which are summarized in Table II.

TABLE II. MAXIMUM PERMISSIBLE CONCENTRATIONS IN AIR AND WATER. (FROM 10CFR20, APPENDIX B).

Radionuclide		Occupational MPC		Unrestricted MPC	
		Air[a,b]	Water[a,b]	Air[a,b]	Water[a,b]
^{3}H	S	5×10^{-6} μCi/ml	1×10^{-1} μCi/ml	2×10^{-7} μCi/ml	3×10^{-3} μCi/ml
	I	5×10^{-6} μCi/ml	1×10^{-1} μCi/ml	2×10^{-7} μCi/ml	3×10^{-3} μCi/ml
	Sub	2×10^{-3}		4×10^{-5}	
^{14}C	S	4×10^{-6}	2×10^{-2}	1×10^{-7}	8×10^{-4}
	(CO_2) Sub	5×10^{-5}		1×10^{-6}	
^{35}S	S	3×10^{-7}	2×10^{-3}	9×10^{-9}	6×10^{-5}
	I	3×10^{-7}	8×10^{-3}	9×10^{-9}	3×10^{-4}
^{85}Kr[c]	Sub	1×10^{-5}		3×10^{-7}	
^{99}Tc	S	2×10^{-6}	8×10^{-2}	7×10^{-8}	3×10^{-4}
	I	6×10^{-8}	1×10^{-2}	2×10^{-9}	2×10^{-4}
^{129}I	S	2×10^{-9}	1×10^{-5}	2×10^{-11}	6×10^{-8}
	I	7×10^{-8}	6×10^{-3}	2×10^{-9}	2×10^{-4}
^{135}Cs	S	5×10^{-7}	3×10^{-3}	2×10^{-8}	1×10^{-4}
	I	9×10^{-8}	7×10^{-3}	3×10^{-9}	2×10^{-4}
^{147}Pm	S	6×10^{-8}	6×10^{-3}	2×10^{-9}	2×10^{-4}
	I	1×10^{-7}	6×10^{-3}	3×10^{-9}	2×10^{-4}
^{151}Sm	S	6×10^{-8}	1×10^{-2}	2×10^{-9}	4×10^{-4}
	I	1×10^{-7}	1×10^{-2}	5×10^{-9}	4×10^{-4}
^{241}Pu	S	9×10^{-11}	7×10^{-3}	3×10^{-12}	2×10^{-4}
	I	4×10^{-8}	4×10^{-2}	1×10^{-9}	1×10^{-3}

[a] To convert to pCi/cm^3 divide numerical value by 10^{-6}.
[b] To convert to $\mu Ci/m^3$ divide numerical value by 10^{-6}.
[c] Technically not a low energy β-emitter.

TABLE III. INSTRUMENTS APPEARING IN TECHNICAL AND REVIEWED LITERATURE

Reference Source	Detector Type	Detector Characteristics	Nuclide Detected	System Response Time	Detection Limits[a]	γ Response	Other Nuclide Response	Remarks
Howell et al[14]	gas flow proportional	250 cm^3 volume 250 cm^3/m flow	^3H	∿5 min.	∿2.5 nC/l		Decreased by lowering concentration	Semipermeable membrane used to enhance HTO over HT and Kr.
Ehret[15, 16][b]	gas flow proportional	1 ℓ volume methanet + 20 - 30% air counting gas, flow 25 l/hr	^3H	few minutes	0.1 pCi/cm^3		Used anti-coincidence proportional chamber	Memory effects due to tritiated water vapor absorption in plastic tube
Block et al[16, 17][b]	Two thin window gas flow proportional chambers	0.2 ℓ volume	^3H		1 pCi/m^3	Compensation such that detection limit held in 3MR/hr field	Compensated against ^{41}Ar and ^{85}Kr	Thin Formvar window allows detection ^3H; aluminum mylar window opaque to ^3H.
Gregory and Parnell[18]	proportional	multiwire multiplane proportional chamber; 112.5 cm^2 active area	^{85}Kr	200 s count time	4×10^{-3} pCi/cm^3	anticoincidence between planes		Technology used in high energy physics
Osborne and Coveart[19]	internal ion chamber with sealed ion chamber inside	1330 cm^3 vol., flow rate 30 cm^3/s, solid state electronics	^3H	70 s.	$5\text{-}10^7$ μCi/m^3	>95%	2.2 times more sensitive to ^{41}Ar	Portable air monitor for tritiated water vapor; ion trap.

[a] Taken as stated by the author; for precise meaning the reader is referred to the source.
[b] The data actually quoted is taken from ref. 16.

TABLE III (Cont'd.)

Reference Source	Detector Type	Detector Characteristics	Nuclide Detected	System Response Time	Detection Limits[a]	γ Response	Other Nuclide Response	Remarks
Jalbert[20]	two concentric ion chambers	1.6 ℓ sampling volume	^3H	18 s.	10^{-6} μCi/ml to 10^{-4} μCi/ml		w/background β of 2×10^{-5} μCi/ml to 0.4 μCi/ml, respectively	Sensitive to temperature or pressure changes; iontrap.
Jalbert and Hiebert[21]	internal ion chamber and sealed ion chamber	1 ℓ volume flow rate 10 ℓ/m	^3H	50 s	25 μCi/m^3	in 50 mR/hr field	no selectivity	Ion trap; chambers orbit about common axis.
Osloond et al[22]	liquid scintilation	5 g. gel 2 ml H_2O 18 ml scintillator solution	^3H	167 hr-sampling 20 min.-counting	4×10^{-11} μCi/ml low humidity 6×10^{-10} μCi/ml high humidity	inherently none	noble gases pass sampling gel	250 g. silica gel, with sample flow rates 6 1/hr; result must be calculated.
Osborne[23]	liquid scintillation		^3H	10 hr at 10 cm^3/min. flow rate, or flow at 100 cm^3/min.	0.1 pCi/cm^3 minimum	inherently none	selectivity against noble gas to <.013% and also HT	Conjectured; uses bubbler system to sample air flow.
Osborne and Tepley[24]	liquid scintillation	LS flow ∿2 mm^3/sec.	^3H	several minutes	∿.3 nCi/m^3 minimum	inherently none	those gases not soluble selected against	HTO collected directly from air in LS; water monitoring possible with detection limit 2 μCi/kg.
Colmenares et al[25]	CaF_2(Eu) scintillator and photomultiplier	6.35 cm. dia. x 0.37 cm.	^3H gas	60 s.[b]	minimum 48 pCi/cm^3			Uses amplifying pulse shape analyzer; ∿.05 efficiency.

[a] Taken as stated by the author; for precise meaning the reader is referred to the source.
[b] Sample & background counting time used to calculate minimum detectable activity.

TABLE III (Cont'd.)

Reference Source	Detector Type	Detector Characteristics	Nuclide Detected	System Response Time	Detection Limits[a]	γ Response	Other Nuclide Response	Remarks
Osborne[26]	plastic NE 102 scintillator flow counter	Sheets 0.125 mm apart w/300 cm^2 surface; water flows through	^3H water vapor	200 s.	1 $\mu Ci/m^3$ minimum	1 mr/hr[27] corresponds to .5 $\mu Ci/m^3$	<0.7% for same conc. ^3H	Air stream vapor collected in water stream, purged; .05% of ^3H disintegration detected.
Moghissi et al[28]	anthracene scintillator	ground scintillator coating 100 parallel 3 mm diameter plexiglas rods, 45 mm long viewed by two PMT.	^3H		minimum ~6nCi/ℓ			Signal out of SCA grated coincident in 30-50 nsec. and in energy window of interest.
Takamatsu[29] et al	plastic scintillator	polyvinyl-toluene plate discs stacked 10 cm and thickness 0.5 mm viewed by 2 PMT	^{85}Kr	1 hr.	min. 10^{-10} $\mu Ci/ml$	anti-coincidence shielding + shielding	selectively enriches Kr over Xe and eliminates Co_2 and H_2O.	Uses silicone rubber membrane and 2 molecular sieves.

[a] Taken as stated by the author; for precise meaning the reader is referred to the source.

The results of the literature survey, as summarized in Table III, indicate that attention has focussed on instruments that determine ^{3}H and ^{85}Kr concentrations.

Since occupational monitoring is generally required when there is a probability that individuals will receive a dose in excess of 10% of the standard,[30] then it is desirable that available monitoring techniques measure as low as 0.1 MPC (occupational). Although this would be greater than that set forth by ANSI N 13.10 - 1974, it says itself, "These values represent current minimum standards. Improved sensitivities are always encouraged and should be used when improved state-of-the-art and commercial availability are realized."[31] In any case instruments should be available to measure at the 0.1 MPC (occupational) level, since the public MPC is even lower.

From perusing Table III it seems clear that all of the techniques are sensitive enough to at least be able to measure down to the occupational MPC_a. Only two of the techniques (without special sampling procedures) are adequate for measuring 0.1 occupational MPC_a. These are liquid scintillation and gas flow proportional counting. Using special sampling techniques the solid scintillator may become adequate for measuring even lower than 0.1 public individual MPC_a, although a rapid response is sacrificed. From the results obtained by Gregory and Parnell,[18] it seems possible that a specially designed gas flow proportional chamber can also measure down to these levels, and with the advantage of a shorter response time. This particular instrument is based on technology used in experimental high energy physics and should be tested further in practical applications for low energy β-emitters to determine its true potential. The techniques that give reliable readings at occupational MPC_a even when other β-activity is present (>10 times) are internal ionization chamber counting with a compensating chamber, liquid scintillation coupled with a bubbler, and either solid scintillation counting or internal ionization counting using permselective membranes for sampling. The technique with the fastest response time at occupational MPC_a is internal ionization chamber counting.

The results of a survey of U. S. commercial sources for off-the-shelf instruments based on three of these techniques is summarized in Table IV. The instruments have been identified by letters and in any case the inference should not be drawn that we recommend one over the other, or that one is inherently superior to another.

TABLE IV. OFF THE SHELF U. S. COMMERCIAL INSTRUMENTS (COMPILED AS A RESULT OF SURVEY CONDUCTED NOVEMBER - DECEMBER 1978)[32]

Indentifier	Detector Type	Detector Characteristics	Nuclide Detected	System Response Time	Detection Limits[a]	γ Response	Other Nuclide Response	Remarks
A	ion chamber	2 ℓ volume, other sizes available	^{3}H	∿10 sec. time constant	1 μCi/m^3 to .2 Ci/m^3	compensated to as high as 10 mR/hr.	no inherent selectivity	ion trap; dust trap; α selectivity possible.
B	2 internal ion chambers and 2 sealed	air flow 2-10 ℓ/min.	^{3}H	15, 45 sec. time constant	1 μCi/m^3 to 10^4 μCi/m^3	compensated up to 5 mR/hr.	no inherent selectivity	electrostatic precipitator submicron filter
			^{14}C	15, 45 sec. time constant	0.2 μCi/m^3 to 2000 μCi/m^3	compensated up to 5 mR/hr	no inherent selectivity	
C	ionization chamber	∿1 ℓ	^{3}H		5 pCi/ m^3 10 μCi/m^3		no inherent selectivity	portable; shock sensitive; available on contract basis only.
D	scintillation	sampling volume 192 $in.^3$ flow rate 4 CFM	^{133}Xe		3 x 10^{-7} μCi/cc min.		no selectivity	very spotty specs; unclear whether it has a response below .30 MeV.
			^{85}Kr		2 x 10^{-7} μCi/cc min.			
E	2 ion chamber	2ℓ volume	^{3}H		3 x 10^{-6} μCi/cc to 3	compensated	no selectivity	dust and ion elimination portable
F	2 ion chambers		^{3}H		10 μμCi/cm^3 to 15,000	compensated to few mr/hr	no selectivity	portable; dust filter and electrostatic precipitator

[a]Taken directly from manufacturers quoted specifications

TABLE IV (Cont'd.)

Indentifier	Detector Type	Detector Characteristics	Nuclide Detected	System Response Time	Detection Limits[a]	γ Response	Other Nuclide Response	Remarks
G	GM detector	thin window pancake 2" dia.	^{133}Xe	40 sec. time constant	2×10^{-5} μCi/ml		no selectivity	a background subtract circuit, separate operation
H	scintillator and photo-multiplier	10 mil thick plastic, sampling volume 100 ml	noble gas	1 min	2×10^{-7} μCi/cc	heavily shielded	no selectivity	unclear whether it has a response below .30 MeV.
I	ion chamber	2 ℓ/m air flow, 2ℓ volume	^{3}H	15 sec. constant	7.5 μCi/m^3-5000	comp. up to 5 mR/hr.	no selectivity	filter and ion trap portable
J	GM detector	wall thickness 30 mg/cm^2	noble gas		10^{-7} μCi/cc -10^{-4} μCi/cc		no selectivity	can't detect ^{3}H or ^{14}C

[a] Taken directly from manufacturers quoted specifications

CONCLUSION AND RECOMMENDATIONS

It seems clear from a study of Table IV that instruments are readily available for measuring low energy β-emitters at occupational MPC_a. However, these are typically not capable of measuring concentrations at the public individual MPC_a for low energy β-emitters. These conclusions agree in a qualitative fashion with other work.[33-35] In order to change this situation it seems clear that several sampling techniques from research would have to be applied commercially. The one with the easiest and most general application would be permselective membranes. In addition, there are three fundamental areas where improvement might be sought: 1) reducing background radioactivity in materials used for construction, 2) improving particular components in a detection system, and 3) paying critical attention to proper design of the system geometry. A specific example of the second factor is improvements in photomultipliers used for scintillation counting. The pulse height resolution capabilities of a photomultiplier are important for the detection and measurement of low-level scintillations in which only a few electrons are produced, as is particularly true for scintillations produced by ^3H. The recent significant high electron resolution improvements observed for prototype high-gain microchannel plate photomultipliers permit the elimination of almost all single-electron dark pulses that accompany low-level scintillations.[36] An obvious potential usage is in tritium counting. In order to make significant advances in all three areas for the future, significant development effort is required. One can only hope that this is done.

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