

#### **LIMITATIONS**

Highly colored samples may not be easily measureable. Either quench corrections have to be applied, or the sample may be diluted to minimize quench influence. Background of the instrument is low enough to use a dilution factor of ten and still achieve a reliable determination. The presence of hydrogen sulphide, easily detectable by its smell, had no effect on the analysis.

Of course, the direct determination of the radionuclide content in solid particles present in the water is hardly possible. Procedures such as acidification to dissolve the particles may be used. Maximum permissible concentrations of radionuclides in solid form are considerably higher than in soluble form.

#### **CONCLUSIONS**

A very fast and simple method for testing wastewater for compliance with government regulations has been described. No evaporation of samples is necessary. Results correspond much more to real gross  $\alpha + \beta$  determination than those obtained with gas counters.  $^3\text{H}$  and volatile radionuclides are not lost and can be monitored. We expect that this method will replace traditional methods.

#### **ACKNOWLEDGMENTS**

We thank Katharina Pock, Erich Findeis and Andreas Fuchshuber for collaboration in measuring and evaluating data.

## WASTEWATER CONTROL IN A NUCLEAR RESEARCH INSTITUTE BY LIQUID SCINTILLATION SPECTROMETRY

FRANZ SCHÖNHOFER

Federal Institute for Food Control and Research, Kinderspitalgasse 15, A-1090 Vienna, Austria  
and

KARL BUCHTELA

Atominstitut der Österreichischen Universitäten, Schüttelstrasse 115, A-1020 Vienna, Austria

**ABSTRACT.** We have used liquid scintillation spectrometry to determine gross alpha-beta activity, to detect  $\alpha$  emitters and to exclude the presence of highly toxic radionuclides in regulated discharges from a nuclear research installation. Our method is fast, simple and more reliable than traditional methods. Restrictions apply to highly colored samples;  $\alpha$  and weak  $\beta$  emitters enclosed in solid particles can hardly be measured.

### INTRODUCTION

Government legislation and regulations of releases of radioactive substances to the environment are usually based on radiation protection concerns. Severe restrictions apply to discharges of alpha and beta emitters. Many such radionuclides cannot be determined by simple gamma spectroscopic measurement. Therefore, it is useful to establish a simple method for determining the  $\alpha$  and  $\beta$  activity, of, for example, wastewater. Traditional methods involve evaporation of water and subsequent counting of the residue using a Geiger or proportional counter. One disadvantage is that volatile radionuclides and compounds are lost. Discrimination between  $\alpha$ - and  $\beta$ -radiation might also be a problem, and it is very difficult to estimate the energy of the betas. Measurement of low-energy  $\beta$  emitters, and especially  $\alpha$  emitters in thick sources, is a well-known problem. We investigated using liquid scintillation counting (LSC) to overcome these difficulties.

### METHODS

From previous experience, we knew that tritium ( $^3\text{H}$ ) can be measured in surface water or precipitation directly after mixing the water with a gel-forming cocktail. We took the same approach to measure  $\alpha$  and  $\beta$  emitters in wastewater from a nuclear research establishment in Vienna.

We measured samples in an ultra-low-level Wallac 1220<sup>TM</sup> Quantulus LS spectrometer after mixture with a gel-forming scintillation cocktail. The pulse-height spectra obtained from the instrument are first checked visually. If significant concentrations of  $\alpha$  emitters are present, they can be easily detected by their relatively sharp peaks, compared to the  $\beta$  emitters. Presence or absence of highly energetic and, therefore, highly radiotoxic  $\beta$  emitters, for example,  $^{90}\text{Y}$  in conjunction with  $^{90}\text{Sr}$ , can be easily verified visually. Less toxic, short-lived highly energetic  $\beta$  emitters, such as  $^{32}\text{P}$ , can be distinguished from long-lived radionuclides by repeated measurements over several days. In principle, a very large number of radionuclides may be present, but in practice, one may employ plausibility reasoning. High-resolution gamma spectrometry, which is used routinely, may give clues to radionuclides, such as  $^{137}\text{Cs}$  or other  $\beta$  emitters. Internal standards may also be useful for identifying radionuclides from their pulse-height spectra or estimating the energy of both  $\alpha$  and  $\beta$  emitters.

## REGULATIONS

Government regulations prescribe that the wastewater from each of three 20-m<sup>3</sup> tanks may be discharged in one day if it does not contain >64  $\mu\text{Ci}$  (2.4 MBq) of a mixture of “unknown  $\alpha$  and  $\beta$  emitters”. (<sup>3</sup>H is not explicitly exempted; pure  $\gamma$  emitters are not mentioned in the regulations because of their low toxicity.) If the presence of <sup>226</sup>Ra and <sup>228</sup>Ra can be excluded, the discharged activity may be higher by a factor of ten. The more highly toxic radionuclides can be exempted, the higher the discharged activity may be.

## APPLICATIONS

Figure 1 shows the spectrum of a real wastewater sample. The relatively sharp peak is due to an  $\alpha$  emitter. We used internal standards of various  $\alpha$  emitters to verify that uranium was present in the sample – much more hazardous radionuclides, such as <sup>239</sup>Pu or <sup>226</sup>Ra (Fig. 2), could be excluded because of their different pulse heights and spectra. We observed no high-energy  $\beta$  emitters, such as <sup>90</sup>Y (and thus, <sup>90</sup>Sr) or <sup>228</sup>Ac (and thus, its mother, <sup>228</sup>Ra). In the low-energy range (the pulse-height scale is logarithmic), we observed <sup>3</sup>H, which can be quantified easily after distillation of a small sample. Using traditional methods of evaporation and measurement with a gas flow counter, neither the  $\alpha$  emitter nor <sup>3</sup>H had been detected before.

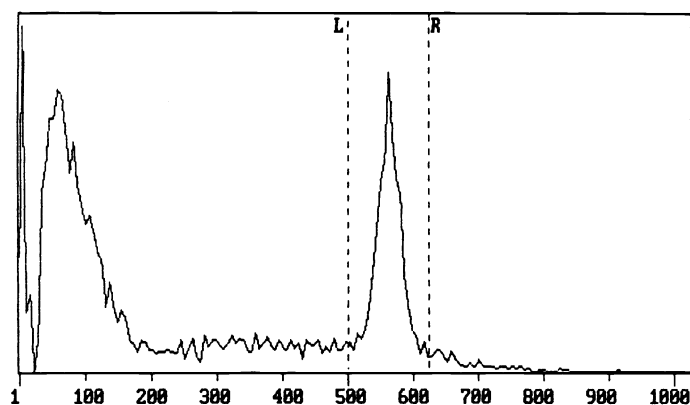


Fig. 1. Pulse-height spectrum of a wastewater sample

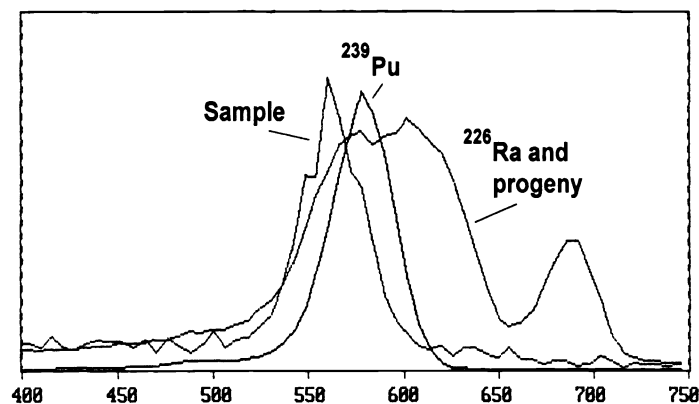


Fig. 2. Pulse-height spectra of the wastewater sample, a <sup>239</sup>Pu internal standard and a <sup>226</sup>Ra internal standard (pulse-height scale expanded)

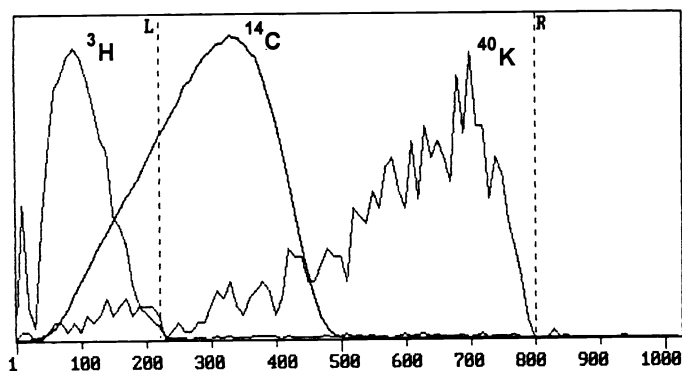


Fig. 3. Pulse-height spectra of standards of  $^3\text{H}$ ,  $^{14}\text{C}$  and  $^{40}\text{K}$

We used the following procedure for quantification: For quick determination, the activity in the obtained spectrum is compared to a  $^{40}\text{K}$  standard (Fig. 3). Because of the negligible radiotoxicity of  $^3\text{H}$ , we do not take it into consideration when determining the gross  $\alpha$  and  $\beta$  activity and set a window above the  $^3\text{H}$  spectrum. Here,  $^{40}\text{K}$  is measured with  $\sim 90\%$  efficiency; the count rate of the samples in this channel is calculated with this efficiency. In Table 1, typical values are shown for background, efficiency (as compared to  $^{40}\text{K}$ ) and the lower limit of detection (LLD) for different counting times. The LLDs shown are based on  $3 \times$  standard deviation of background.

TABLE 1. Specifications for Gross  $\alpha + \beta$  Measurements

Background (cpm)	Efficiency (%)	LLD (Bq liter <sup>-1</sup> )		
		500 min	60 min	10 min
2.65	87.1	0.52	1.51	3.7

Alpha emitters are counted with  $\sim 100\%$  efficiency, so that their activity is slightly overestimated. Beta emitters with similar energy are counted with similar efficiency. The activity of  $\beta$  emitters with lower energy will, on the one hand, be underestimated to an extent, depending on their energy, but on the other hand,  $\beta$  emitters with low energy are much less toxic than those with higher energy. For example,  $^{14}\text{C}$  (Fig. 3) is counted with only  $\sim 50\%$  efficiency in this window, but the maximum permissible concentration of  $^{14}\text{C}$  in water is higher by a factor of 2000 than for  $^{90}\text{Sr}$ . In principle, corrections may be applied after tests with low-energy  $\beta$  emitters, but in practice, this has never been necessary because the determined activities were far below the limits.  $^3\text{H}$  (Fig. 3) is determined either from the same spectrum after comparison to a  $^3\text{H}$  standard, or, if the contribution in the low-energy range from the other  $\beta$  emitters is too high, it can be determined precisely after distillation.

We measure samples as quickly as possible to enable the institute to empty a full wastewater tank as quickly as possible. Usually, we need only 10-min counting time due to the extremely low background of the LS spectrometer. We obtain a spectrum for a first estimate of the sample's activity to meet regulatory requirements. It takes less than half a day between sampling and clearance.