Toward a Standard Method for Determination of Waterborne Radon

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Abstract:
When the USEPA specifies the maximum contaminant level (MCL) for any contaminant, a standard method for analysis must be simultaneously stipulated. Promulgation of the proposed MCL and standard method for radon in drinking water is expected by early next year, but a six-month comment period and revision will precede final enactment.

The standard method for radon in drinking water will probably specify that either the Lucas cell technique or liquid scintillation spectrometry be used. The latter technique will probably predominate, and this paper reports results which support a standard method with the following features: (1) samples should be collected by an explicitly stated technique to control degassing, in glass vials with or without scintillation cocktail, and possibly in duplicate. (2) samples should be measured by liquid scintillation spectroscopy in a specified energy "window", in a glass vial with particular types of cocktails. (3) radium standards should be prepared with controlled quench levels and specified levels of carriers, but radium-free controls prepared by a specified method should be used in interlaboratory comparison studies.

Three elements of a proposed standard method for the determination of waterborne radon will be discussed in detail: Standard preparation, sampling methodology, and liquid scintillation spectral region selection.

Standard Preparation: Standards prepared from Radium solutions of known activity exhibit very complex behavior because they are subject to several chemical as well as several radiologi-
cal equilibrium processes. Liquid scintillation spectra will be displayed which provide a basis for explanation of the changes which are observed with temperature, agitation, and changes in vial composition. Standards show strong temperature dependence when prepared in plastic vials because changes in temperature cause chemical disequilibria between the aqueous layer, the cocktail layer, the vial, and the cap, all of which have different affinities for radon and its progeny. All chemical equilibria are, of course, overlain with the trend toward radiological equilibrium. The temperature dependence is complex, and reestablishment of equilibrium may take days. As a result, environmental samples will usually be at a different state of equilibration than the standards. Measurement in glass vials is therefore recommended.

Even if glass vials are used, the interference of aqueous phase radionuclides must be quantified. We have found that cocktail-insoluble radionuclides contribute only 1-2% of the total activity of standards. At the extremely low chemical concentration levels necessary for measurable counts, even ionic compounds are somewhat soluble in the mineral oil-, pseudocumene-, or alkylbenzene-based cocktails typically used. Solubility is demonstrated by a carrier effect in which barium nitrate/nitric acid carrier added during standard preparation actually decreases counting efficiency.

Most of the cocktails that are commercially available show similar counting efficiencies, but they behave differently, as would be expected, in chemical equilibration processes. The rates of attainment of equilibrium for newly prepared standards can be used to diagnose these differences among cocktails. Because organic (two-phase) cocktails allow the use of aqueous radium for standards, they are preferable to detergent type (single-phase) cocktails which allow only the use of radium-free, radon "controls". These "controls" serve the purpose of a useful check on determinations made with organic cocktails.
Sampling: The sampling vial type critically affects the determination of waterborne radon. A comparison of plastic and glass vials over time reveals loss through adsorption into plastic vials, and strong loss through adsorption into rubber septa on glass vials. Bubbles in glass vials have an effect which is predictable from the distribution coefficient for radon between air and water. Plastic vials containing scintillation cocktail or glass vials with aluminum or Teflon septa are acceptable sampling containers, although the latter may be preferable.

Spectral Region Selection: Most interesting effects are observed when the entire liquid scintillation spectrum of samples containing radium and its progeny is obtained. Glass or plastic vials in which radon or radium solutions had been stored can be emptied and filled with cocktail to reveal the processes of radiochemical equilibration between solution and container phases. Peaks are readily identified by observing their behavior under radiological or chemical disequilibrium, and peak selection can lead to dramatic improvements in sensitivity and concomitant reductions in LLD. Most background counts are fortuitously outside the region of interest for radon and progeny.

It is possible to determine radon concentrations as low as 3.7 Bq dm\(^{-3}\) after a 150 hour delay using glass vials and other optimal conditions. Water samples in plastic vials counted after 20 hours without peak selection as described above give results below the LLD.