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Rapid screening of radioactivity in food for emergency response

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ABSTRACT

This paper describes the development of methods for the rapid screening of gross alpha (GA) and gross beta (GB) radioactivity in liquid foods, specifically, Tang drink mix, apple juice, and milk, as well as screening of GA, GB, and gamma radioactivity from surface deposition on apples. Detailed procedures were developed for spiking of matrices with ^{241}Am (alpha radioactivity), ^{90}Sr/^{90}Y (beta radioactivity), and ^{60}Co, ^{137}Cs, and ^{241}Am (gamma radioactivity). Matrix stability studies were performed for 43 days after spiking. The method for liquid foods is based upon rapid digestion, evaporation, and flaming, followed by gas proportional (GP) counting. For the apple matrix, surface radioactivity was acid-leached, followed by GP counting and/or gamma spectrometry. The average leaching recoveries from four different apple brands were between 63% and 96%, and have been interpreted on the basis of ion transport through the apple cuticle. The minimum detectable concentrations (MDCs) were calculated from either the background or method-blank (MB) measurements. They were found to satisfy the required U.S. FDA's Derived Intervention Levels (DILs) in all but one case. The newly developed methods can perform radioactivity screening in foods within a few hours and have the potential to capacity with further automation. They are especially applicable to emergency response following accidental or intentional contamination of food with radioactivity.

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1. Introduction

Radiological safety of food is of paramount importance for protection of the general population from internal radiation exposures. The source of radioactivity in food can be natural radionuclides or contamination resulting from accidents or intentional actions. Radioactivity in food can be incorporated either within the food volume or surface-deposited. The most well-known documented case of radioactivity entering the food chain is the Chernobyl accident (Baratta, 2003). A response to this accident was creation of the networks of monitoring laboratories for radiological protection of food (e.g., Varga et al., 2006).

An important function for laboratories to effectively analyze food for radioactivity and report meaningful results is participation in proficiency testing, intercomparison studies, and emergency exercises. The European community has recently conducted a comprehensive intercomparison study of ^{137}Cs, ^{40}K, and ^{90}Sr in milk powder (Spasova et al., 2008; Wätjen et al., 2008). Recently, the U.S. Food and Drug Administration's (FDA) Food Emergency and Response Network (FERN) supported an extensive Radiological Capability and Capacity Inter-laboratory Comparison Exercise (MENU, 2010). Realistic food matrices were chosen for the exercise: milk, Tang fruit drink, surface-spiked apples, and tuna fish. Several radionuclides of concern were selected: ^{90}Sr/^{90}Y, ^{131}I, ^{137}Cs, ^{239}Pu, and ^{241}Am (EPA, 2008). We do not report (MENU, 2010) results in this paper, only the results obtained at New York and Wisconsin states' health laboratories. (MENU, 2010) preliminary results have been recently reported in After Action Report (AAR, 2010).

Most known methods of environmental radioactive sample analysis are also applicable to the analysis of food products. However, these methods are usually time-consuming and not applicable to emergency situations. Relatively little information is available on rapid methods of analysis for both environmental samples and food samples, and there exists a strong need for development of such methods. The analysis of food samples is especially complicated due to its chemical complexity. One rapid method of determination of Am and Pu in liquid foods was developed using chemical separation and liquid scintillation counting (Healey et al., 2010). Rapid methods for gross alpha (GA) determination in Tang fruit drink were developed using either microwave digestion (Chu et al., 2010) or coprecipitation (Parsa, 2010), followed by gas proportional (GP) counting. The existing method for rapid leaching of radioactive strontium from

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vegetation (ASTMI, 2009) exhibited low recoveries (30%), which could be presumably due to low leaching volumes and insufficient acidity of the leaching solution.

The present work focuses on the development of rapid GA screening of liquid foods: Tang drink mix, apple juice, and milk, assuming incorporation of radioactivity into the volume of the food samples. Such incorporation can be caused by either passage through the food chain or intentional spiking. Another scenario by which radioactivity can enter the foodstuff is surface deposition, which may result from the explosion of a radiological device or from fallout following a nuclear reactor accident or a nuclear detonation. The latter scenario is addressed in the second part of this work, which focuses on the determination of GA, gross beta (GB), and gamma radioactivity from surface-spiked apples.

2. Radioactivity in liquid foods

2.1. Matrix selection, preparation, and spiking

Some of the ingredients of the liquid food matrices used in this work are listed in Table 1, reproduced from the manufacturers' labels. When reported, the Percent Daily Values from the labels were converted to mass according to DA (2010). All matrices contain considerable quantities of carbohydrates, particularly apple juice. Milk also contains protein and fat. Both Tang drink and apple juice contain some ascorbic acid. The presence of these chemicals is challenging to GA assay. Tang fruit drink is sold as a powder, which needs to be dissolved in water. There are some inconsistencies in the manufacturer's (volumetric) packaging instructions for the dilution. For our purposes, we adopted a dilution of 6.25 g of powdered Tang in 100 mL of deionized water. We noticed that Tang has not dissolved completely in water and left some white residue at the bottom of the container, so it was necessary to shake the container well before taking an aliquot.

Appropriate quantities (e.g., 10, 15, 20, 25, and 30 mL) of liquid samples were taken for analysis. The samples of Tang drink, apple juice, and milk were spiked immediately before analysis with 11.9 Bq of ^{241}Am or 6.82 Bq of ^{90}Sr/^{90}Y per sample, for the analysis or to determine the efficiency of the GP counter for alpha and beta particles. The samples for method blank (MB) were not spiked. All radionuclides used in this work were NIST-traceable.

For the dry matrix stability test, as required by the (MENU, 2010) exercise, the samples of Tang powder were spiked first and then dissolved in water as needed. We spiked each of several vials containing 6.25 g of Tang powder with ^{241}Am. Then, each dry sample was dissolved in 100 mL of water after a chosen delay time of up to 34 days (Table 2). A 25 mL aliquot of Tang drink was taken for the analysis, containing 11.9 Bq of ^{241}Am.

2.2. Digestion and evaporation of liquid samples

As can be seen from Table 1, the liquid foods analyzed contain substantial quantities of carbohydrates and, in the case of milk, fat, and protein. Direct evaporation of Tang and apple juice causes formation of thick, sticky, spattering residue inappropriate for direct counting. In addition, heating and evaporation of milk cause formation of lipid layers as well as coagulation of proteins. Consequently, acid digestion during evaporation is necessary. It was quickly realized that H_2SO_4, HCl, and *aqua regia* are not appropriate for use, since they corrode stainless steel planchets preventing accurate residue mass determination for detector efficiency calculations. Therefore, three strategies for digestion were investigated:

- Step 1: HNO_3; Step 2: H_2O_2.
- Step 1: HNO_3; Step 2: HCl; Step 3:HNO_3.
- Step 1: HNO_3; Step 2: HNO_3.

Another observation was that the rapid evaporation from a small beaker causes losses of residue mass, presumably carried away with aerosols and vapors. Therefore, 250 mL boiling flasks (24/40, available from Kimble Chase, Vineland, NJ) were used. Also, distillation glass beads were added to flasks to prevent violent boiling.

The HNO_3/H_2O_2 digestion proceeded as follows. In Step 1, an aliquot of Tang solution or apple juice was transferred into the boiling flask, and the volume was reduced by approximately one half through boiling on a hot plate. Then, a volume, equal to the volume of the Tang aliquot, of concentrated (70%) HNO_3 was added. For milk, HNO_3 was added before heating, and the solution was allowed to cold-digest for 10 min before placing on a hot plate. This step is critical, since it prevents formation of a thick film of fat on the surface of milk during evaporation.

The liquid sample was evaporated to near dryness. This evaporation took about 30 min, depending upon the quantity of the liquid sample. Next, the flask was removed from the heating plate and allowed to cool briefly. Following this, H_2O_2 (35% in H_2O) was carefully added in Step 2, equal in volume to the original sample volume. Addition of H_2O_2 must proceed slowly; otherwise the reaction can be violent. Then, the solution was evaporated to a small volume (3–5 mL). The second evaporation took about 20 min. It is important not to evaporate to dryness in order to prevent possible explosion in the presence of hydrogen peroxide.

We also tried a concentrated $HNO_3/HCl/HNO_3$ digestion, where HCl was added drop wise in Step 2 and was driven off with HNO_3

Table 2
Test of ^{241}Am tracer stability in Tang powder.

Delay time (d)	^{241}Am recovery (%)
1	91.1
1	106.2
12	106.9
12	107.2
21	92.6
21	102.5
34	116.9
34	109.2
Average recovery	104 ± 9 (1σ)

Table 1
Some of the ingredients of liquid foods in the study.

Liquid food	Brand	Serving size (mL)	Na (mg)	K (mg)	Ca (mg)	Carbohydrates (g)	Protein (g)	Fat (g)	Ascorbic acid (mg)
Tang drink[a]	Kraft	237	0	NA	120	9	0	0	90
Apple juice	Hannaford	237	10	240	NA	29	0	0	117
Milk (1%)	Crowley	237	130	NA	360	13	8	2.5	2

[a] "Sensible Solution" variety with half the sugar of 100% juice.