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Radiological risks of neutron interrogation of food

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Abstract

In recent years there has been growing interest in the use of neutron scanning techniques for security. Neutron techniques with a range of energy spectra including thermal, white and fast neutrons have been shown to work in different scenarios. As international interest in neutron scanning increases the risk of activating cargo, especially foodstuffs must be considered.

There has been a limited amount of research into the activation of foods by neutron beams and we have sought to improve the amount of information available. In this paper we show that for three important metrics; activity, ingestion dose and Time to Background there is a strong dependence on the food being irradiated and a weak dependence on the energy of irradiation.

Previous studies into activation used results based on irradiation of pharmaceuticals as the basis for research into activation of food. The earlier work reports that ^{24}Na production is the dominant threat which motivated the search for $^{23}\mathrm{Na}(n,\gamma)^{24}\mathrm{Na}$ in highly salted foods. We show that $^{42}\mathrm{K}$ can be more significant than $^{24}\mathrm{Na}$ in low sodium foods such as Bananas and Potatoes.

Keywords: ingestion dose, cargo interrogation, food activation, induced activity, neutron activation

(Some figures may appear in colour only in the online journal)

1. Introduction

Over 90% of all global freight is transported through seaports [1], the port of Felixstowe alone handles over 3 million containers each year with typical dimensions 6.1 m × 2.44 m × 2.59 m. Apart from manual searches and intelligence gathering, the detection of contraband and security threats crossing national borders depends primarily on x-ray interrogation. X-ray interrogation can be defeated by shielding or disguising objects allowing smugglers to pass contraband through national borders undetected [2]. In addition x-ray interrogation systems often require a human operator to identify the presence of any threat or contraband. The difficulty of identifying threats leads to a large number of false-positive results, and of greater concern false-negative results. Singh [2] refers to a United States Federal Aviation Authority report finding that human operators of x-ray interrogation systems only identified approximately 20% of threats. The interactions between neutrons and matter are distinct from those of x-rays with matter, whilst x-rays interact with both the nucleus and the electron cloud neutrons only interact with the nucleus. Neutron interactions with matter are highly non-linear with both atomic mass (A) and atomic number (Z). Due to the nature of neutron interactions they can interrogate the contents of containers not suited to x-rays. X-rays are readily absorbed by high Z materials such as Fe and Pb whereas these are fairly transparent to neutrons. There are a variety of neutron security techniques which use either fast, thermal or white neutron spectra, and in some cases combinations.

The most commonly used neutron sources for interrogation are 14.1 MeV sealed DT fusion tubes (^{3}H(d,n)). In place of DT fusors any combination of projectile (proton, deuteron or α) and target nucleus, such as ^{7}Li(p,n) or ^{16}O(d,n), will provide a neutron flux and each combination will produce a different energy beam. In this paper we use a numerical, Monte Carlo, approach to study the impact of neutron beams with kinetic energy ranging from 1 MeV to 20 MeV irradiating commonly containerised foodstuffs. There is European Legislation relating to the irradiation of foods that governs aspects of the irradiation of food, however neutrons with energy below 14 MeV with an absorbed dose below 0.01 Gy are exempt from the legislation [3]. The European legislation does not prevent higher energies or doses being used, but requires justification for use outside the set limits.

As neutrons propagate through a container some of the contents, including food, will be activated raising the possibility of exposing the general public to radiation. When exposing individuals to radiation it is required to keep the dose As Low As Reasonably Achievable (ALARA). The ALARA principle does not require radiation to be removed altogether, but if a reduction can be made without compromising efficacy and with minimal cost that reduction should be made [4]. In addition to the ALARA principle it is necessary to consider the Justification Principle, which says that any change in radiation exposure must do more harm than good [4]. To date there has been little research into the relationship between source neutron energy, food composition and activation.

Tenforde [5] showed that pharmaceuticals and medical devices irradiated by a mix of fast (8.5 MeV with narrow distribution) and thermal (Maxwellian distribution extending up to 0.1 MeV) neutrons would not produce effective doses above a recommended safe limit of 1 mSv per year. The results given by Tenforde [5] suggest that for an 8.5 MeV source irradiating various pharmaceuticals the production of Na^{24} in milk of magnesia (MgOH) is likely to produce the highest absorbed dose. Assuming 10 g of Mg ingested (recommended dose 2.7 gd^{-1}) the absorbed dose for a 50 kg person would be 6.84×10^{-8} mSv, far less than the recommended dose limit of 1 mSv.

Due to the conclusions of Tenforde [5] that only ^{24}Na need be considered for pharmaceuticals and medical devices Tenforde [6] considered only ^{24}Na production when analysing the results of neutron irradiation on food. As with pharmaceuticals the production of ^{24}Na by an 8.5 MeV neutron source was below safe levels.

The food irradiation studies of Giroletti [7] agreed with those of Tenforde [6] and showed that no significant production of ^{24}Na would be seen. Nelson [8] looked at the activation of various common cargo items ranging from jars of pasta sauce to sheets of aluminium. By measuring the time taken for irradiated goods to return to background Nelson [8] showed that the activation would reduce to safe levels within the typical storage time of transported goods.

The exclusive consideration of ^{24}Na by Tenford [6] and Giroletti [7] was based upon a study of pharmaceuticals and medical devices under 8.5 MeV neutron irradiation. As Giroletti [7] uses a 14 MeV source additional activation channels not considered by Tenforde [5] may have become available. Additionally it can be argued that the composition of pharmaceuticals and medical devices are not adequately representative to identify all possible hazards which might be produced in foodstuffs, which have highly varied composition.

The experimental method Nelson [8] used an unmoderated 14 MeV neutron beam incident on a single layer of material. In reality as a neutron beam passes through a container the spectrum will become moderated. The moderation will produce a significant thermal tail in the spectrum which will have an impact on activation. Despite the variety of the previous research the limitations prevent firm conclusions from being drawn.

Experimentally studying the relationship between neutron energy and cargo activation is complicated by the need for a variable energy, mono-chromatic, neutron source. An in-depth study is essential as there are multiple possible reactions a nuclide can undergo e.g. (n, α), (n, p), (n, γ), which may include multiple cross section resonances. In this paper we begin the process of performing this research using numerical simulations allowing any energy of neutron to be produced and a full analysis of the effect.

The foods chosen for the work presented here were; Almond, Banana, Brie, Cocoa Powder, Corn, Potato and Rice. These foods were chosen because they contain a range of elements in varying concentrations as shown in table 1. Whilst the list of foods simulated is far from exhaustive the variation in composition is broad and will show whether food composition plays a significant role in activation. In addition to the varied compositions all the foods are commonly imported and exported by a range of countries necessitating interrogation. We consider the γ activity, ingestion dose as a result of irradiation and the time required for samples to return to pre-irradiation ingestion dose.

We have expanded upon previous work in this area in a number of ways. By using the neutron spectrum seen after a beam has propagated through cargo, varying the source energy, and including all produced radio-isotopes, we ensure that all aspects of the system are considered.

2. Simulations

The results presented in this paper were produced with a combination of radiation transport and nuclear inventory simulations. The radiation transport was performed in MCNPX [9] and the nuclear inventory in Fispact-II [10].

MCNPX (Monte Carlo N-Particle eXtended) is a general purpose Monte Carlo transport code which tracks neutrons, protons, electrons, photons and a broad range of other particles and ions, however for this work only the neutron transport was required. MCNPX uses evaluated data tables, such as ENDF/B-VII. 0 [11] with thermal neutrons being described by both the free gas and $S(\alpha, \beta)$ models.

Fispact-II is a time dependant nuclear inventory code which models the activation and transmutation of nuclei under neutron, proton, α, deuteron or γ irradiation. Simulations can use a combination of irradiation and cooling phases during which the time dependent production and decay of nuclei is calculated. For known cross sections the European Activation File (EAF) [12] is used with the cross sections calculated in the Talys Evaluated Nuclear Data Libraries (TENDL) [13] used for those which are unknown.

Table 1. The elemental composition of the foods simulated.

	Almonds	Banana	Brie	Cocoa	Corn	Potato	Rice
H	6.47×10^{0}	9.85×10^{0}	8.59×10^{0}	6.26×10^{0}	6.72×10^{0}	1.01×10^{1}	7.16×10^{0}
C	4.34×10^{1}	1.11×10^{1}	2.38×10^{1}	4.34×10^{1}	4.03×10^{1}	9.61×10^{0}	3.66×10^{1}
N	3.67×10^{0}	1.90×10^{-1}	3.59×10^{0}	2.93×10^{0}	1.63×10^{0}	1.30×10^{0}	2.55×10^{0}
O	4.44×10^{1}	7.85×10^{1}	6.16×10^{1}	4.42×10^{1}	5.05×10^{1}	7.83×10^{1}	5.21×10^{1}
F	0	2.20×10^{-6}	0	0	0	0	0
Na	1.00×10^{-3}	1.00×10^{-3}	6.30×10^{-1}	2.10×10^{-2}	3.50×10^{-2}	6.00×10^{-3}	1.30×10^{-2}
Mg	2.70×10^{-1}	2.70×10^{-2}	2.00×10^{-2}	5.00×10^{-1}	1.30×10^{-1}	2.30×10^{-2}	2.65×10^{-1}
P	4.80×10^{-1}	2.20×10^{-2}	1.90×10^{-1}	7.30×10^{-1}	2.10×10^{-1}	5.70×10^{-2}	6.16×10^{-1}
S	3.00×10^{-1}	1.60×10^{-2}	3.00×10^{-1}	2.40×10^{-1}	1.30×10^{-1}	1.10×10^{-1}	2.08×10^{-1}
Cl	1.50×10^{-3}	1.50×10^{-3}	9.70×10^{-1}	3.20×10^{-2}	5.40×10^{-2}	9.20×10^{-3}	2.00×10^{-2}
K	7.10×10^{-1}	3.60×10^{-1}	1.50×10^{-1}	1.52×10^{0}	2.90×10^{-1}	4.20×10^{-1}	$4.12 \times 10\ -1$
Ca	2.60×10^{-1}	5.00×10^{-3}	1.80×10^{-1}	1.30×10^{-1}	7.00×10^{-3}	1.20×10^{-2}	4.26×10^{-2}
Mn	2.30×10^{-3}	3.00×10^{-4}	0	3.80×10^{-3}	5.00×10^{-4}	2.00×10^{-4}	$6.90 \times 10\ -3$
Fe	3.70×10^{-3}	3.00×10^{-4}	5.00×10^{-4}	1.40×10^{-2}	2.70×10^{-3}	8.00×10^{-4}	2.7×10^{-3}
Cu	1.00×10^{-3}	1.00×10^{-4}	0	3.80×10^{-3}	3.00×10^{-4}	1.00×10^{-4}	5.00×10^{-4}
Zn	3.10×10^{-3}	2.00×10^{-4}	2.40×10^{-3}	6.80×10^{-3}	2.00×10^{-3}	3.00×10^{-4}	3.70×10^{-3}
Se	2.50×10^{-6}	1.00×10^{-5}	1.45×10^{-4}	1.43×10^{-4}	1.55×10^{-4}	3.00×10^{-7}	4.33×10^{-5}

Note: The relative mass per 100 g of food each element (3 s.f.) is given.

Neutron interaction cross-sections are strongly energy dependant typically with multiple resonances. MCNPX was used to provide the neutron spectrum which was then passed to Fispact-II to compute the nuclear inventory. For each food a 1 m^3 cube was simulated to ensure realistic levels of neutron spectrum moderation. A pencil beam of mono-chromatic neutrons was directed into one side of the volume and the spectrum recorded after 90 cm. Leaving 10 cm of food between the surface where the spectrum was measured and the end of the volume to ensure any reflected or scattered neutrons are included in the spectrum passed to Fispact-II. Tracking the spectrum through the simulated volume showed that the thermal component is rapidly populated and the distribution remains approximately constant throughout with only the total number of neutrons reducing with depth.

The Fispact-II simulations were run with a flux of 10^8 n cm^{-2} s^{-1} and a fluence of 10^9 n cm^{-2}. The fluence used is comparable to that used by the authors of [7]. The fluence will be the dominant factor in the level of activation, with flux only influencing the number of very short lived isotopes left at the end of irradiation. We do not seek to identify an optimum fluence, and the level of activation will be approximately linear with fluence, therefore the differences between foods and energies will be approximately flux and fluence independent.

The masses of trace elements in foodstuffs along with the water and protein mass are well known. Two important approximations were included, the mass not included in trace elements, water or protein was simulated as cellulose and the sodium was assumed to be in the form of sodium chloride.

To calculate the mass of H, C, N, O and S the mass of water, protein and cellulose were used. In water the mass ratio of H:O was assumed as $1:8$, cellulose has the chemical composition $C_6H_{10}O_5$ giving C:H:O mass ratios of $7.2:1:8$. There are a variety of proteins found in nature however Torabizadeh [14] calculated a generic formula $C_nH_{1.85n}N_{0.28n}O_{0.3n}S_{0.01n}$ which gave mass ratios of C:H:N:O:S in protein as $12:1.85:3.92:4.8:0.32$.

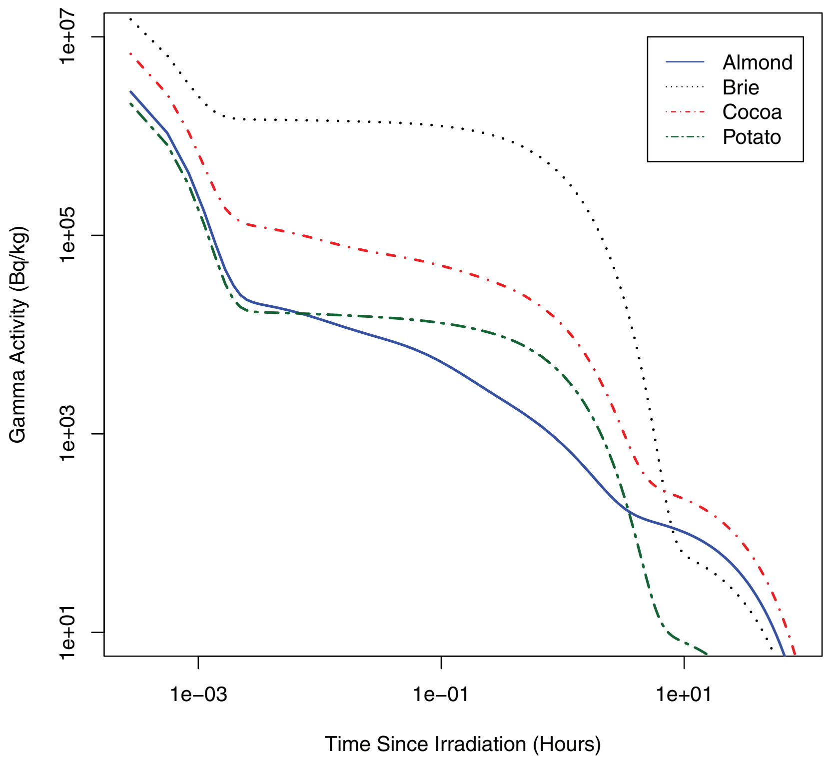

Figure 1. Decay in γ activity with time starting immediately after irradiation and continuing to 83 h (3.5 d) after irradiation. The γ activity for Almond, Brie, Cocoa and Potato irradiated by a 14 MeV neutron source is shown in (Bq kg^{-1}).

For each of the foods simulated; Almond, Banana, Brie, Cocoa, Corn, Potato and Rice the relative mass per 100 g of food of each element is given in table 1, these foods were chosen because they cover a range of compositions and are commonly containerised. The foods were simulated at a density of 1 g cc^{-1}, the results are given and discussed in the following section.

3. Results

To understand the relationship between activation, neutron energy and composition three figures of merit have been considered; Time to Background (TtB), activity and ingestion dose. In many respects the ingestion dose is most important as a low activity α emitter could be more harmful than a high activity γ emitter when ingested. The activity and TtB are important when considering the effect of irradiation on those handling goods, and the TtB is particularly important when considering ingestion after irradiation.

Immediately after irradiation the γ activity of goods is the highest threat as any α and β activity will be blocked by container walls and packaging. If the activity is too high it may necessitate storage in a radiation controlled area and it is important to know how long a container may need such measures and if this time can be reduced.

The results in figure 1 show the decay in γ activity in Bq kg^{-1} for Almond, Brie, Cocoa and Potato, the other foods fit within the range covered by these four. The percentage uncertainty in the activity for the four foods shown peaked at 12.6%, 18%, 11.8% and 15% but this

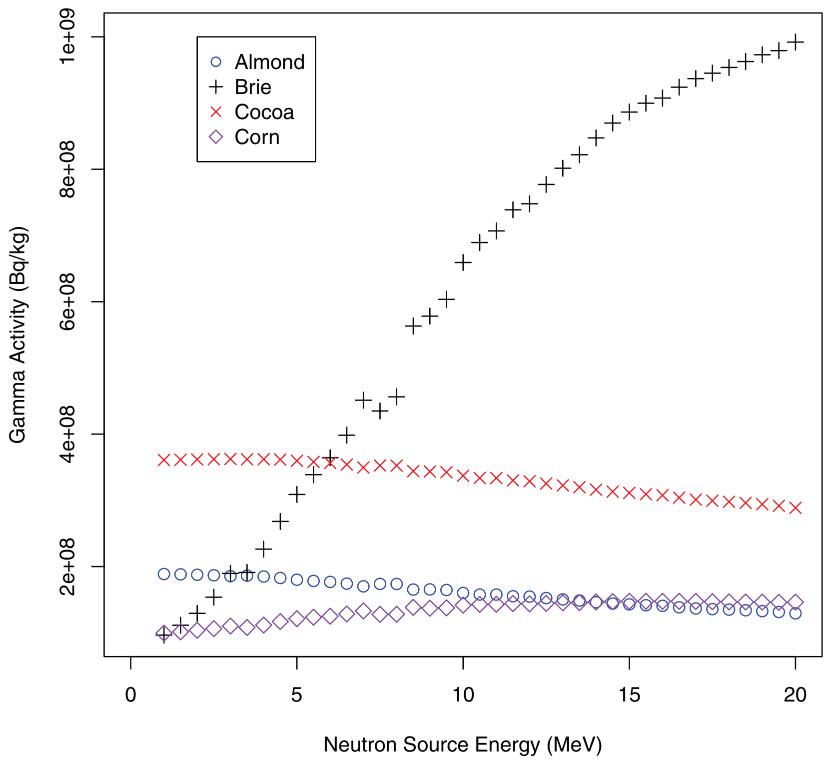

Figure 2. The dependence of the γ activity of Almond, Brie, Cocoa and Corn on the energy of the irradiating neutron source, neutron energy ranging from 1 MeV to 20 MeV. Almond, Cocoa and Corn show insignificant dependance but Brie shows approximately an order of magnitude variation.

included contributions from all decay modes, not just the γ activity shown in figure 1. Figure 1 shows that food composition can have a substantial effect, in this case the γ activity varies by more than 2 orders of magnitude approximately 10 h after irradiation.

According to the analysis of Tenforde [6] and Giroletti [7] high salt foods undergoing ^{23}Na$(n, \gamma)^{24}$Na reactions are the primary activation threat for foods. The sodium content of Brie is approximately 600 mg per 100 g of cheese and so ^{23}Na$(n, \gamma)^{24}$Na reactions will contribute significantly to the activation. In foods with low levels of salt and magnesium the production of ^{24}Na by ^{23}Na$(n, \gamma)^{24}$Na and ^{24}Mg$(p, n)^{24}$Na reactions is never a dominant process. In the case of Bananas and Potatoes the activity from ^{42}K is higher than that of ^{24}Na on the time scales where the activity of ^{24}Na is dominant in other foods.

Figure 2 shows the energy dependence of γ activity immediately after irradiation for Almond, Brie, Cocoa, and Corn, the results of the other foods fall below the data of Cocoa also with low energy dependence. The energy dependence of Almond, Cocoa and Corn are very small and may not be experimentally measurable however Brie shows a very strong energy dependence of approximately an order of magnitude across the energy range. As with the results shown in figure 1 the uncertainties are only available for the total activity however in this case they vary with energy, low energy results have higher uncertainties peaking at 24.7%, in Brie at 1 MeV, and dropping to 10.7%, in Cocoa, at 20 MeV. The energy dependence shown in figure 2 is representative of all samples and continues with time but decreases and becomes inconsequential after the first 1 to 2 h.

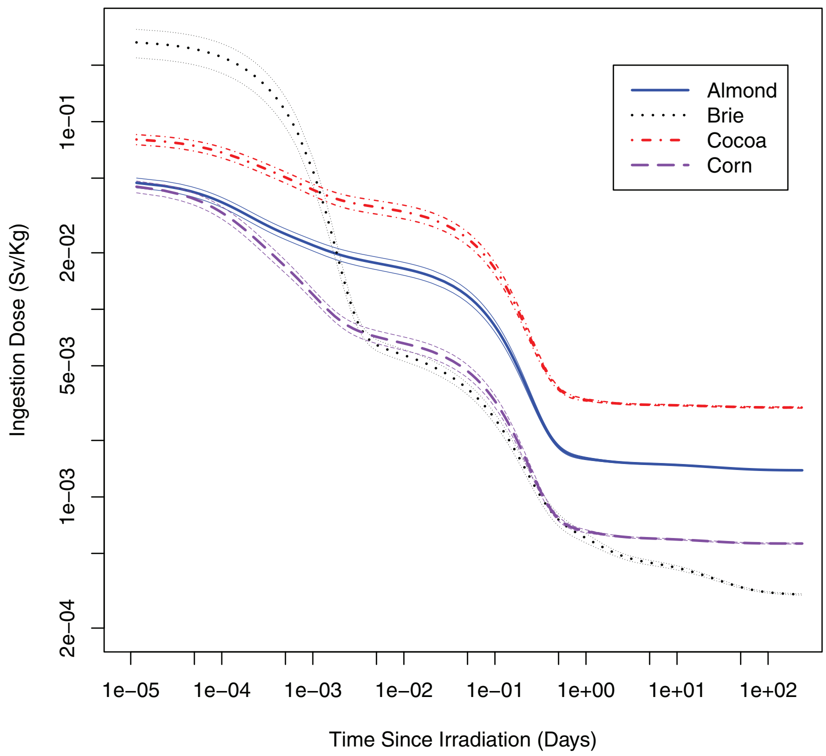

Figure 3. Decay with time of the ingestion dose induced in Almond (solid blue line), Brie (dotted black line), Cocoa (dashed-dotted red line) and Corn (dashed purple line) under 14 MeV neutron irradiation shown in Sv kg^{-1}. The decay is shown starting 10^{-5} days (0.8 s) after irradiation through to 30 d (1 month) after. The uncertainties are given by the faint lines which bracket each main line.

The results in figure 3 show the decay in ingestion dose of Almond, Brie, Cocoa and Corn. As with the γ activity shown in figure 1 the ingestion dose shown in figure 3 indicate a strong food dependence. The uncertainties are shown by the faint lines bracketing the thicker lines.

Along with the magnitude of the activity and the ingestion dose discussed previously the time required for a sample to return to background should be considered. Figure 4 shows the time required for the ingestion dose of Brie, Cocoa, Corn and Rice to return to within 5% of naturally occurring pre-irradiation levels of each food, the other foods show minimal energy dependence and fall below the level of Rice. The ingestion TtB shows a strong food dependence and a weak energy dependence. In descending order the means of the ingestion TtBs are: Brie, 106 d; Almond, 30 d; Rice, 26 d; Corn, 18 d; Cocoa, 4 d; Potato, 4 d; Banana, 1 d varying by approximately 10% across the energy range used. The TtB for activity of each sample was comparable to that of the ingestion dose but the energy dependence was weaker. The 5% threshold was chosen as it allows the short lived isotopes to decay, typically within 1 to 2 d, and leaves only the longer lived and potentially threatening isotopes.

The uncertainties in the simulations are dominated by the cross sections used in Fispact-II. For the simulations we used the 616 group EAF-2010 neutron activation cross sections in which unknown cross sections are calculated resulting in some isotopes having very large uncertainties.

The molecular structure in which the produced radio-isotopes are found is not considered in the Fispact-II simulations when calculating ingestion dose. The molecule a radioisotope is

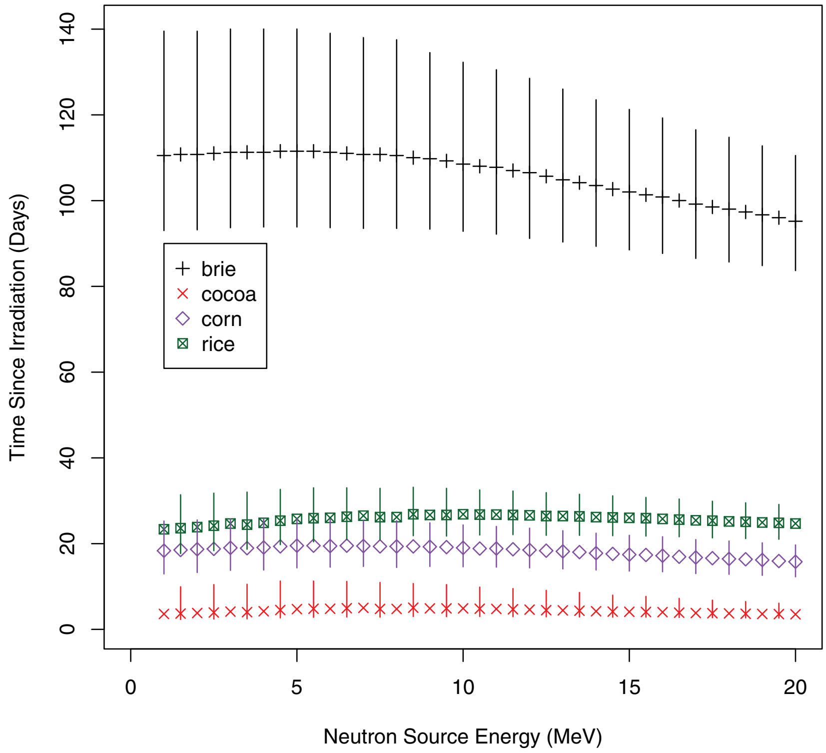

Figure 4. The time required for the ingestion dose of the 7 irradiated samples to return to background after irradiation by a neutron beams ranging from 1 MeV to 20 MeV. Background is taken as the ingestion dose prior to irradiation and the plot shows time in days required to reach background+5%.

part of can have a significant impact on the biological half-life, and therefore the radiotoxicology [15]. The biological half-life may have a dramatic effect on the results presented however significant research would need to be performed to determine if that was the case.

4. Conclusion

In this paper we have shown that composition of food has a strong effect on three relevant metrics; Time to Background (TtB), ingestion dose and activity and that the energy of the irradiating neutrons can also have an effect. The precise effect of the neutron irradiation is strongly dependent upon both the food being irradiated and the time since irradiation.

Previous research [5, 7] has concluded that the build up of ^{24}Na though ^{23}Na$(n,\gamma)^{24}$Na and ^{24}Mg$(n,p)^{24}$Na reactions in irradiated food is the only concern. In the case of Banana and Potato, which have a very low salt content, the gamma activity cannot be explained through ^{24}Na but is in fact due to ^{42}K predominantly from ^{41}K$(n,\gamma)^{42}$K reactions, though others are also contributors.

The activation of a food is more strongly dependant on the composition than the energy however the radiological risk may be reduced in some cases with careful selection of irradiation energy. A variable energy neutron source could enable safer security scanning, with neutron energies tailored to have the lowest possible impact on a given food, although in practice imposing an upper limit based on the response of certain foods may be the most practical option.

Although the results in this paper show that varying neutron energy has an impact on the three figures of merit considered (Time to Background, Activity and Ingestion Dose) it is clear that the food being irradiated has a much bigger impact.

Due to the accuracy limitations of the numerical models, in particular those used in Fispact-II, it is essential that experimental work is performed to verify the results in this paper. In addition to the limitations of the numerical models for production of radionuclides the resulting ingestion dose is also not a trivial problem, Puncher [16] did a detailed analysis of the uncertainties associated with inhalation and ingestion doses. This paper shows that there is an energy dependence for activation of food however all other containerised goods will potentially be irradiated by neutrons as part of a security system. An understanding of how more goods, e.g. clothes, computers and machinery are affected by neutrons, and what role the energy plays in that effect would be beneficial.

Acknowledgments

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References

[1] Bergantino A, Musso E and Porcelli F 2013 Port management performance and contextual variables: which relationship? Methodological and empirical issues *Res. Transp. Bus. Manag.* **8** 39–49
[2] Singh S and Singh M 2003 Explosives detection systems (EDS) for aviation security *Signal Process.* **83** 31–55
[3] European Parliament 1999 Directive 1999/2/EC of the European Parliament and of the Council of 22 February 1999 on the approximation of the laws of the member states concerning foods and food ingredients treated with ionising radiation *Official J. Eur. Communities* **42** 16
[4] Valentin J and 2007 *The2007 Recommendations of the International Commission on Radiological Protection* (Oxford: Elsevier)
[5] Tenforde T 2002 Letter report on radiation protection advice for pulsed fast neutron analysis system used in security surveillance
[6] Tenforde T 2003 Presidential report on radiation protetion advice for the pulsed fast neutron system used in security surveillance: part II. The ALARA principle and related issues
[7] Giroletti E, Bonomi G, Donzella A, Viesti G and Zenomi A 2012 Radiological risks from irradiation of cargo contents with EURITRACK neutron inspection systems *Radiat. Phys. Chem.* **81** 758–65
[8] Nelson M 2006 Investigation into the feasibility of highly enriched uranium detection by external neutron stimulation (expanded study)
[9] Forster R, Cox L, Barrett R, Booth T, Briesmeister J, Brown F and Bull J 2004 MCNP version 5 *Nucl. Instrum. Methods Phys. Res.* B **213** 82–6
[10] Sublet J, Eastwood J and Morgan J 2012 *The Fispact-II User Manual*
[11] Chadwick M, Oblo P, Herman M, Greene N M, McKnight R, Smith D and Young P 2006 ENDF/VII.0: next generation evaluated nuclear data library for nuclear science and technology *Nucl. Data Sheets* **107** 2931–3060
[12] Sublet J, Packer L, Kopecky J, Forrest R, Koning A and Rochman D 2010 The european activation file: EAF-2010 neutron-induced cross section library
[13] Koning A and Rochman D 2012 Modern nuclear data evalution with the TALYS code system *Nucl. Data Sheets* **113** 2841–934
[14] Torabizadeh H 2011 All proteins have a basic molecular formula *World Acad. Sci. Eng. Technol.* **54** 961–5
[15] Takeda H 1982 Comparative metabolism of tritium in rat after single ingestion of some tritiated organic compounds versus tritiated water *J. Radiat. Res.* **23** 345–57
[16] Puncher M 2014 An assessment of the reliability of dose coefficients for intakes of radionuclides by members of the public *J. Radiol. Prot.* **34** 625–43