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Radioisotopes produced by neutron irradiation of food

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HIGHLIGHTS

- We show that neutron interrogation of food can produce many radioisotopes.
- We show a strong dependance between food and certain radioisotopes.
- Some isotopes are shown to have an energy dependence.
- Previous claims that 24Na is the main threat is shown to only apply in special cases.

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ABSTRACT

The use of neutrons for cargo interrogation has the potential to drastically improve threat detection. Previous research has focussed on the production of ^{24}Na, based on the isotopes produced in pharmaceuticals and medical devices. For both the total activity and the ingestion dose we show that a variety of isotopes contribute and that ^{24}Na is only dominant under certain conditions. The composition of the foods has a strong influence on the resulting activity and ingestion dose suggesting that the pharmaceuticals and medical devices considered initially are not a viable analogue for foodstuffs.

There is an energy dependence to the isotopes produced due to the cross-sections of different reactions varying with neutron energy. We show that this results in different isotopes dominating the ingestion dose at different energies, which has not been considered in the previous literature.

1. Introduction

Millions of containers pass through national borders every year, the port of Felixstowe alone sees approximately 3 million containers measuring approximately $6.1\,\text{m} \times 2.44\,\text{m} \times 2.59\,\text{m}$ annually. Inspecting the contents of these containers for content validation is vital for combating smuggling and for international terrorism prevention.

Current X-ray based security methods struggle to determine the presence organic material when shielded by high density, or disguised by other low density, materials. The potential for using neutrons in place of X-rays is gaining interest, with the potential for greatly increased threat detection (Liu et al., 2008). A variety of neutron techniques are available, the fast neutron technique Pulsed Fast Neutron Analysis (Brown and Gozani, 1995) has a lot of interest however there are techniques using all energies available. A detailed discussion of neutron scanning techniques is available in Runkle et al. (2009), Buffler (2004) and the references within.

When considering the use of neutron interrogation the efficacy for contraband identification is not the only consideration. The activation of irradiated goods is an unavoidable side effect of neutron irradiation and some research has been performed to identify the level of threat this may pose (Nelson, 2006; Tenforde, 2002; Giroletti et al., 2012).

For cargo interrogation 14 MeV $T(d, n)$ sealed neutron generators are typically used, e.g. in Nelson (2006), Giroletti et al. (2012) however 8.5 MeV $D(d, n)$ reactions are also considered, as in Tenforde (2002). ^{252}Cf fission sources can also be used for techniques requiring white or thermal spectra.

The authors of Nelson (2006) considered the effect of irradiating a broad range of materials, from jars of Ragu to metal sheets, with a 14 MeV neutron beam. The time required for the irradiated material to return to background was measured and used as the figure of merit. The time required for induced activity to decay away showed that the longest times were less than typical storage times at port. The results were limited however as only a single thickness of material, e.g. 1 jar of Ragu or 1 thin sheet of metal, was used leading to minimal moderation. As the beam

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Table 1
The elemental composition of the foods simulated. The relative mass (g/100 g) of each element (3 s.f.) in the simulated foods is given.

Element	Almonds	Banana	Brie	Cocoa	Corn	Potato
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}
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}
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}
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}
F	0	2.20×10^{-6}	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}
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}
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}
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}
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}
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}
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}
Mn	2.30×10^{-3}	3.00×10^{-4}	0	3.80×10^{-3}	5.00×10^{-4}	2.00×10^{-4}
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}
Cu	1.00×10^{-3}	1.00×10^{-4}	0	3.80×10^{-3}	3.00×10^{-4}	1.00×10^{-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}
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}

was almost entirely unmoderated there would have been very few neutrons in the thermal region where many cross-section resonances lie.

The authors of Tenforde (2002) showed that under 8.5 MeV neutron irradiation the dominant threat isotope in pharmaceuticals and medical devices was ^{24}Na. The method used in Tenforde (2002) was to calculate the induced activity based on a spectrum with fast and thermal components rather than a full Monte-Carlo approach.

The results of Tenforde (2002) were extended in Tenforde (2003) to include ^{24}Na production in food. As with Tenforde (2002) the conclusion of Tenforde (2003) was that no unacceptable level of activation would be seen. To determine if the induced activity would pose a problem the authors of Tenforde (2002), Tenforde (2003) calculated the ingestion dose for irradiated goods. The acceptable dose received by the public as a result of irradiation was set at 1 mSv/year, the greatest dose calculated was 1 μSv/year.

The threat isotopes considered by Tenforde (2003) may not be readily applied to all foodstuffs as the compositions of foods and pharmaceuticals are not necessarily equivalent. Additionally the target elements considered by Tenforde (2002) have omissions which may not matter for pharmaceuticals but may be significant for some foods. Activation reactions based on Ca(n,X) were not included in Tenforde (2002), but Ca is found in significant quantities in a variety of foods, including dairy and tofu.

The results of Tenforde (2003) were further extended by the authors of Giroletti et al. (2012) who considered ^{24}Na production by 14 MeV neutron irradiation. As with Tenforde (2003) only ^{24}Na production was considered, however the increased neutron energy may enable additional reactions and the applicability of pharmaceuticals as an analogue of food is still to be verified.

In this paper we show that the induced activity and ingestion dose are caused by a variety of isotopes. Furthermore we show that the conclusion that ^{24}Na is the dominant threat isotope is only valid under certain conditions. Finally we show that there is an energy dependence for induced activity and ingestion dose, which may justify consideration of the interrogation energy.

2. Simulations

By performing numerical simulations we are able to produce any energy neutron beam desired. In this paper we consider the radioisotopes produced by neutron beams with energies from 1 MeV to 20 MeV incident upon a variety of foods, we then show how the dominant cause of ingestion dose and activity varies as a function of time, energy and on the food type irradiated.

We used two numerical techniques to identify the radioisotopes produced, MCNPX (Forster et al., 2004) and Fispact-II (Sublet et al., 2012). MCNPX is a Monte-Carlo radiation transport code, which uses point wise data sets and numerical models to track particles through materials. Fispact-II is a nuclear inventory code, which provides time dependent production, and decay, of radionuclides under arbitrary irradiation conditions.

Neutron transport in MCNPX was used to generate the neutron spectrum, the spectrum was then passed to Fispact-II to simulate the isotope production. The MCNPX simulations used a pencil beam source of perfectly monochromatic neutrons. The neutrons were directed at a cubic volume, 1 m on each side, of material and the spectrum was recorded 10 cm before the far face of the volume. The Fispact-II simulations were run with a fluence of 10^{9} neutrons and irradiation time of 60 s to provide the activation products.

The results obtained from these simulations are specific to the flux and fluence used. Changing the flux and/or fluence will influence the levels of activation after irradiation. For example at higher flux the interrogation time will be reduced meaning that short lived isotopes will be at higher levels immediately after irradiation.

In both MCNPX and Fispact-II the foods were simulated with the same elemental ratios and natural isotopic composition. The trace elements, protein and water content per 100 g provided the elemental composition, with the remaining mass simulated as cellulose. The protein composition was calculated using the generic formula $C_nH_{1.85n}N_{0.28n}O_{0.3n}S_{0.01n}$ (Torabizadeh, 2011) and the trace elements considered were Ca, Fe, Mg, P, K, Na, Cl, Zn, Cu, Mn, Se and F. The masses of each element used in g/100 g are shown in Table 1.

The foods used were Almond, Banana, Brie, Cocoa, Corn, Potato and Rice, they were chosen as they cover a broad variety of compositions and are commonly containerised for import/export. The elemental ratios used in these simulations was chosen to approximate the average composition of distributed foods. The elemental composition will vary with country of origin and cellulose is not the only organic component however these approximations are sufficient to highlight any significant effects.

3. Results

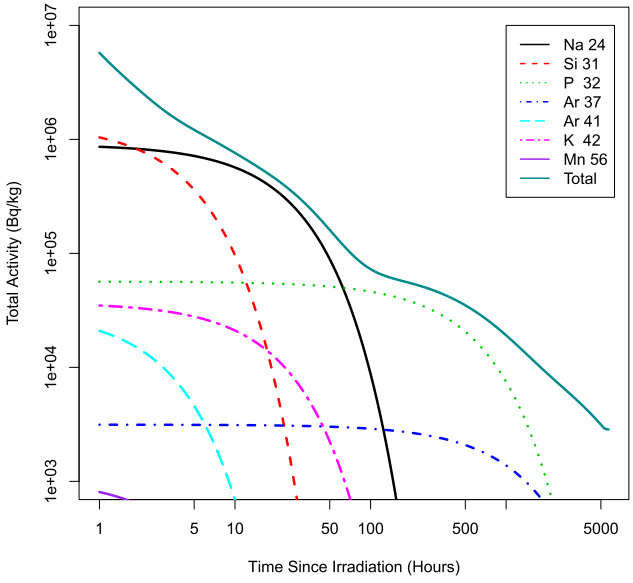
The increase in both the total activity and the ingestion dose of food after irradiation should be considered. Activity is important during packaging and handling, whereas ingestion dose is important when food is consumed. An α emitter will pose no threat whilst containerised but have significant ingestion dose, whereas γ emitters are potentially hazardous at all times.

The results presented here consider only the non-natural isotopes in each food. We do not consider the contribution from naturally occurring radioisotopes, for example ^{40}K, as only activation above background is a concern for public health. We show the produced isotopes dominating both the total activity and the total ingestion dose under 14 MeV irradiation and then show how the production of these isotopes varies with energy.

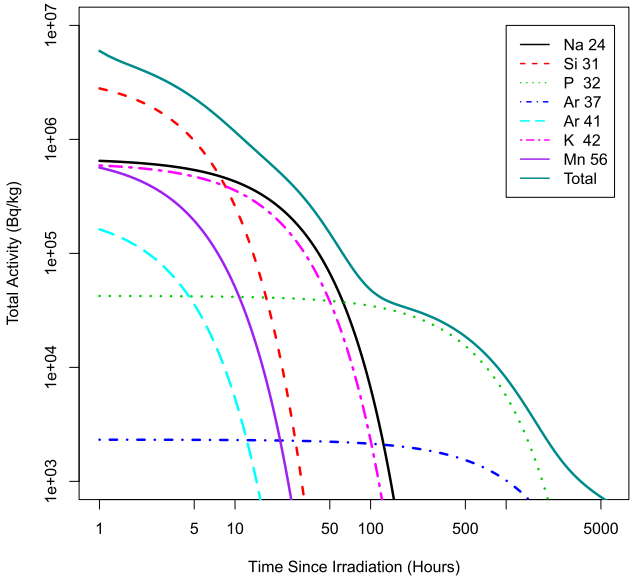
The production of ^{24}Na through ^{23}Na$(n,\gamma)^{24}$Na and ^{24}Mg$(n,p)^{24}$Na reactions were identified by Giroletti et al. (2012), Tenforde (2003) as the greatest threat to health when irradiating food for security. In our study the highest Na content was found in Brie with 629 mg per 100 g and the highest Mg content was in Cocoa Powder with 499 mg per 100 g. The Na and Mg content of Banana is very low at 1 mg and 27 mg per 100 g respectively.

From the simulations used the activity induced in Brie and Cocoa are shown in Figs. 1(a) and (b). The Brie results show strong ^{24}Na dominance from approximately 5–75 h after irradiation, with other isotopes dominating outside this range. Cocoa also shows a ^{24}Na dominance, for slightly less time than Brie, however the activity of ^{42}K is nearly as high as that of ^{24}Na. Finally Banana, shown in Fig. 1(c) shows no ^{24}Na dominance, however ^{42}K is dominant for a substantial time.

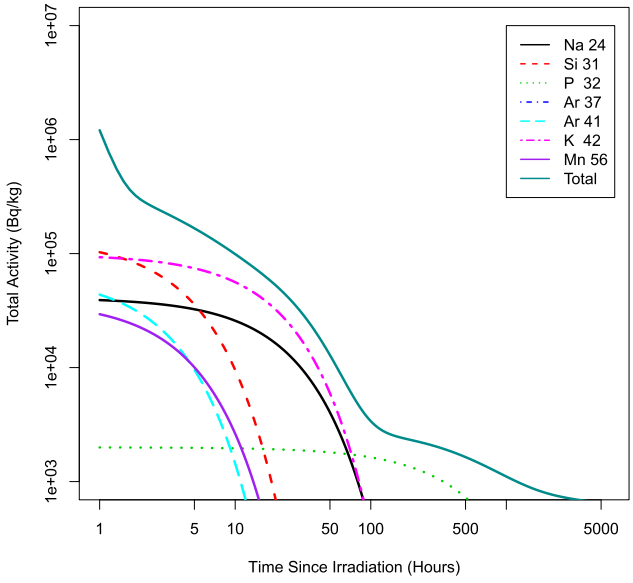
The induced activities show that, as previously claimed in Tenforde (2002), ^{24}Na can be the most important isotope, however this only applies to a narrow time window and for foods high in Na and/or Mg. Whilst the activity is an important consideration, and a



(a) Brie



(b) Cocoa



(c) Banana

Fig. 1. Activity of ^{24}Na, ^{31}Si, ^{32}P, ^{37}Ar, ^{41}Ar, ^{42}K and ^{56}Mn produced in Brie, Cocoa and Banana after interrogation with 14 MeV neutrons. The total induced activity is also given.

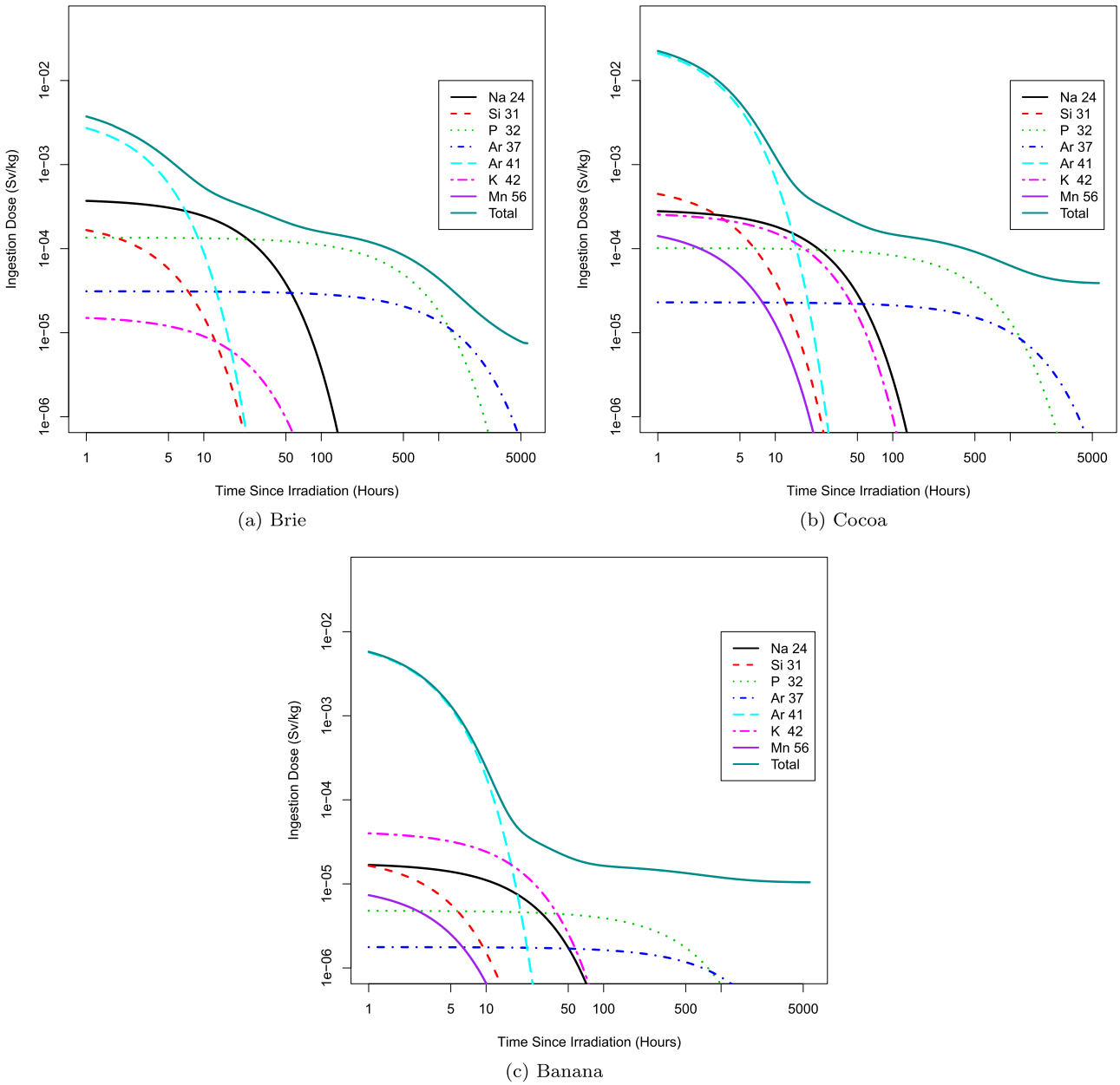


Fig. 2. Ingestion dose of ^{24}Na, ^{31}Si, ^{32}P, ^{37}Ar, ^{41}Ar, ^{42}K and ^{56}Mn produced in (a) Brie, (b) Cocoa and (c) Banana after interrogation with 14 MeV neutrons. The total induced ingestion dose is also given.

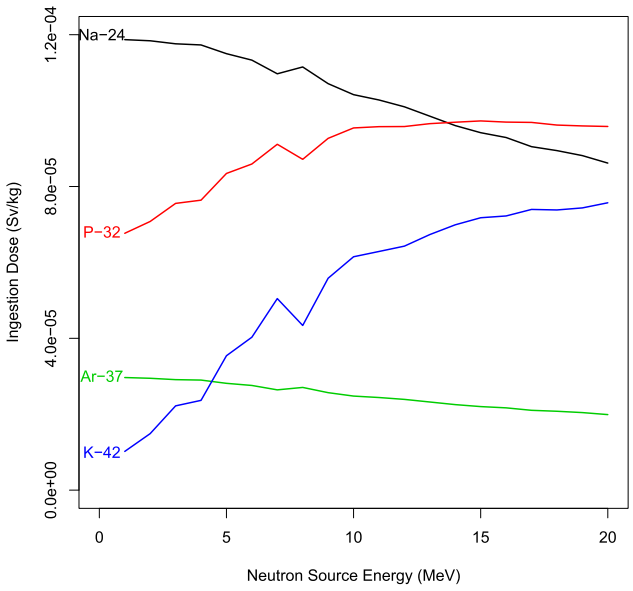
useful parameter to measure, the most important consideration for foods is the ingestion dose. As there is no direct relationship between the activity and ingestion dose of a nuclide the ingestion dose was taken from the results of the Fispact-II simulations.

The ingestion dose of the dominant produced radioisotopes is shown for Brie, Cocoa and Banana in Figs. 2(a), (b) and (c) respectively. The plotted ingestion dose results show that whilst ^{24}Na can be very significant for the activity it's significance is considerably reduced in the ingestion dose. Particular attention should be paid to the ingestion dose of ^{41}Ar and ^{32}P, in both cases the difference between the activity compared to ^{24}Na is much larger than the ingestion dose compared to that of ^{24}Na.

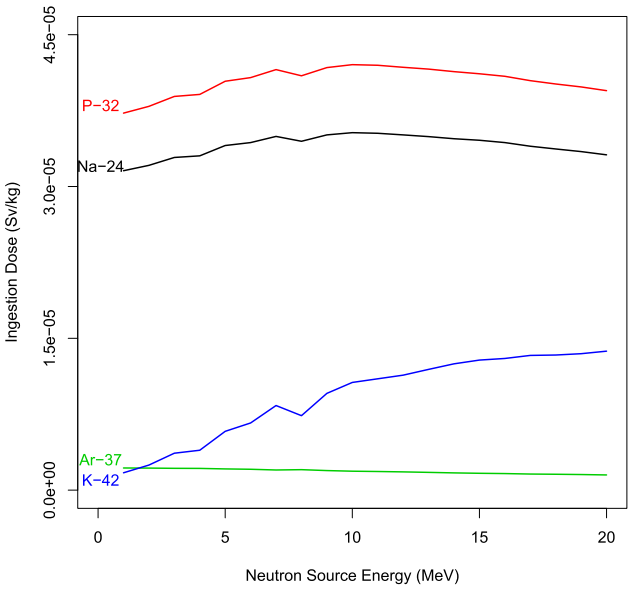
The production of ^{41}Ar is primarily through ^{41}K(n,p) and ^{44}Ca(n,α) reactions, with a small contribution from other interactions. The original calculations used in Tenforde (2002) considered only ^{41}K(n,γ)^{42}K reactions for K isotopes and did not use any reactions from Ca. As can be seen in Fig. 2a–c there is a very significant contribution to the ingestion dose from ^{41}Ar for approximately 10 h after irradiation. The production of ^{41}Ar demonstrates the importance of including all isotopes in the calculations of produced isotopes. The limited considerations of K and Ca reactions may have been reasonable for Tenforde (2002), however when considering activation of food, as in Tenforde (2003) and Giroletti et al. (2012), it is evidently problematic.

The number of produced isotopes is proportional to the cross-section(s) for the reaction(s) producing that isotope. As the range of energies were simulated it is possible to observe any significant energy dependence in the resulting ingestion dose and activity. The results in Fig. 3 show the energy dependence of the ingestion doses for dominant produced isotopes in Rice, Cocoa and Brie 24 h after irradiation. After 24 h many isotopes have decayed to essentially 0 leaving the longer half-life isotopes, mainly ^{24}Na, ^{32}P, ^{37}Ar and ^{42}K.

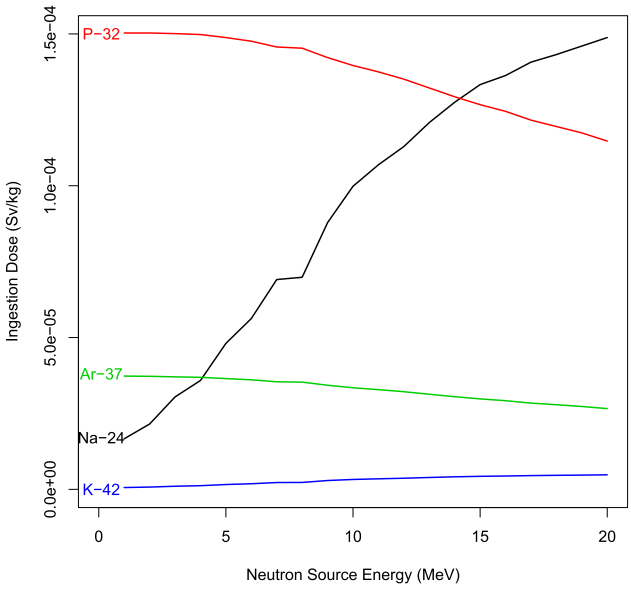
The results in Fig. 3 show the energy dependence for the dominant ingestion dose contributing isotopes in Cocoa, Corn and Brie. Comparison of Cocoa (Fig. 3(a)) with Brie (Fig. 3(c)) shows an



(a) Cocoa



(b) Corn



(c) Brie

Fig. 3. The ingestion dose of ^{24}Na, ^{32}P, ^{37}Ar and ^{42}K 24 h after irradiation of Cocoa, Corn and Brie by neutrons with source energy from 1 to 20 MeV.

increase in ^{24}Na with energy for Cocoa and a decrease with energy for Brie. Conversely the ^{32}P ingestion dose of Coca increases with increasing irradiation energy, but decreases with energy in Brie. Corn, shown in Fig. 3(b) shows no significant energy dependence, unlike Cocoa and Brie.

The differing energy dependencies shown in Fig. 3 demonstrate the potential influence of irradiation energy in an interrogation system. Whilst the energy dependence is not universal it does suggest that careful consideration of the source energy may enable reduced exposure of the public to additional radiation.

4. Conclusion

The conclusion that ^{24}Na is the primary threat isotope was based on research into induced activity in pharmaceuticals and medical devices under 8.5 MeV irradiation (Tenforde, 2002). Food was first considered by Tenforde (2003) at 8.5 MeV, and later Giroletti et al. (2012) at 14 MeV. The applicability of pharmaceuticals as an analogue of food was not considered, nor was the potential for 14 MeV neutrons to stimulate different reactions, and have different cross-sections, to 8.5 MeV neutrons considered.

In this paper we have shown that there are multiple isotopes influencing both the activity and the ingestion dose of irradiated food and that ^{24}Na is only the dominant threat under certain conditions. Some reactions that were omitted from Tenforde (2002), such as Ca(n, X) reactions, can be significant. Excluding Ca reactions may have been justified for pharmaceuticals and medical devices however as a source of ^{41}Ar this has a significant effect on the ingestion dose for approximately 24 h after irradiation.

The change from 8.5 MeV considered by Tenforde (2002) to 14 MeV considered by Giroletti et al. (2012) will have an influence on the produced isotopes. The energy dependence of different isotopes, shown 24 h after irradiation, indicates that a change in

source energy necessitates a detailed analysis of the induced activity and ingestion dose.

We have shown that the compositions of the foods considered have a significant effect on the quantities of isotopes produced. Despite the composition of all foods being above 93% H, C and O there is still a significant influence from the trace elements.

Contrary to Tenforde and Giroletti, who state ^{24}Na is the primary threat isotope, in this work we have shown that neutron energy, and the trace elements present in the foods considered have a significant effect on the types/quantities of isotopes produced. We believe that our paper demonstrates that before recommendations on legislative limits can be made further work is necessary to understand the impact of neutrons on a wider range of foods.

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