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The Radium Legacy: Contaminated land and the committed effective dose from the ingestion of radium contaminated materials

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Abstract

The manufacture and use of radium in the early to mid-20th century within industrial, medicinal and recreational products has resulted in a large number of contaminated sites across a number of countries with notable examples in the USA and Europe. These sites, represent a significant number of unregulated sources of potential radiological exposure that have collectively and hitherto not been well characterised. In 2007, the Radioactive Contaminated Land (RCL) regulations came into force in the UK, providing the statutory guidance for regulators to classify and deal with RCL. Here we report on results derived from digestion experiments to estimate committed effective dose, a key aspect of the RCL regulations, from the ingestion of radium contaminated sources that can be found in the environment. This case study includes particles, clinker and artefacts that arise from past military activities on a site that was once an airfield at Dalgety Bay on the Firth of Forth, UK. Since 2011 the number of radium contaminated finds have increased by one order of magnitude on the foreshore areas of Dalgety Bay. The increase in finds may in large part be attributed to a change in monitoring practice. A sub sample of sixty sources were selected, on the basis of their activity and dimensions, and subjected to digestion in simulated stomach and lower intestine solutions. The study demonstrated that more radium-226 (^{226}Ra) and lead-210 (^{210}Pb; driven by Polonium solubility) is dissolved from sources in artificial 'stomach' solutions compared with 'lower intestine' solutions. The combined 'gut' solubility for ^{226}Ra and apparent ^{210}Pb varied from less than 1 % to up to 35 % ICRP 72 conversion factors were used to convert the activities measured in solution to committed effective dose. A little over 10 % of the sources tested dissolved sufficient radioactivity to result in 100 mSv committed effective dose to an infant. Using the solubility of 35 % as a worst case, minimum source activities necessary to deliver 100 mSv to the full age range of users of the foreshore were estimated. All the estimated activities have been detected and recovered through routine monitoring.

1. Introduction

Radium, which is an alpha particle emitter, was frequently used in the early to mid-20th century as a luminescent paint (Harvie, 1999, Pratt, 1993). Research into the health effects of radium contamination in the human body started in the US in the mid 1920s, following the significant increase in morbidity and mortality amongst radium dial workers in New Jersey (Stebbings, 2001). The history of the radium dial painters, primarily women, has been well documented along with the follow-up health studies (e.g. Stebbings, 2001; Fry, 1998) and Rowland (1994) provides a classic review of the impact and health related research undertaken in the US. In the UK, radium production ended by the mid 1930's although imported radium salts continued to be used for medicine, industry and military purposes into the 1960s in the UK (Harvie, 1999).

In the UK, a number of possible radium contaminated sites was recognised by Government in 1990 (House of Commons, 1990). A recent UK Government report (DECC[1], 2012) estimated that there are between 150-250 contaminated sites across the UK associated with Ministry of Defence (MOD) activities and acknowledged that there may be as many as 1000 sites. These sites represents an important source of potential radiological exposure situations compared with the limited number of conventional nuclear sites, although there is uncertainty as to the number of sites and the consequential hazards they pose.

In 2007 the RCL[2] Regulations (Scottish Statutory Instrument 2009) came into force in the UK, providing the environmental regulatory agencies with the statutory instrument to classify radioactively contaminated land and the duty to implement appropriate enforcement strategies. The Scottish Environment Protection Agency (SEPA) has used the criteria in the statutory guidance (Statutory Guidance to support the Radioactive Contaminated Land (Scotland) Regulations 2007) to describe amount of contamination that may give rise to significant harm to humans (Dale et al., 2012; 2011; 2009). For *homogeneous contamination,* significant harm is defined when lasting exposure gives rise to an individual dose exceeding one or more of the following criteria:

(a) An effective dose of 3 mSv per annum;
(b) An equivalent dose to the lens of the eye of 15 mSv per annum;
(c) An equivalent dose to the skin of 50 mSv per annum.

Contamination in the environment that is *heterogeneous* and associated with particles or artefacts complicates the assessment of contaminated land. Dale *et al.* (2008) proposed that the nature of the hazard along with the probability of encounter should also be considered within a risk assessment. For *heterogeneous contamination* the statutory guidance (Statutory Guidance to support the Radioactive Contaminated Land (Scotland) Regulations 2007) provides thresholds above which significant harm is being caused irrespective of the probability of encounter. These thresholds are:

(d) The potential total effective dose is greater than 100 mSv; or
(e) The contact with contamination would result in a dose to the skin greater than 10 Gy hr[-1].

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Should condition (a) to (e) not be met then the probability of encounter should be taken into consideration and the possibility of harm is significant when the product of the dose and probability exceeds the thresholds described in (a) to (c) above. Current site management arrangements are also considered prior to any designation of contaminated land.

The spatial extent of radium contamination can be characterised by *in-situ* and mobile gamma spectrometric methods (e.g. Tyler, 2008) and techniques have been developed to detect and isolate spatially *heterogeneous* radioactive contamination including particles (Tyler *et al.*, 2010). However, until the sites are identified, characterised and where necessary remediated, the risk of exposure to society remains. Whilst many routes of exposure are possible, ingestion of contaminated artefacts, materials and food products remains one of the primary areas of concern. The literature on the gastro-absorption of radium across the gut is limited (Rundo, 1999), with early studies based on soluble radium salts indicating the fractional absorption of gastro-absorption of between 0.25 and 0.35 (Seil *et al.*, 1915). However, the availability of radium on contaminated sites is likely to be highly dependent on the physical and chemical form of the radium, which may be controlled by the site history and subsequent environmental influences on the contaminated material.

Dalgety Bay (Figure 1), on the north side of the Firth of Forth, is linked with ^{226}Ra contamination as result of the wartime and post-war activities of the airfield operated by the RNAS Donnibristle and HMS Merlin. The airfield closed in 1959 and the new town of Dalgety Bay was developed across the entire area in the 1960s and 1970s. The first ^{226}Ra contaminated particle was discovered and recovered from the foreshore at Dalgety Bay in 1990 as part of the baseline monitoring campaign by Babcock Engineering Services for the Rosyth Naval Base. Once it was identified as ^{226}Ra, its origin was attributed to the historical operations on the Donnibristle airfield. The National Radiological Protection Board (NRPB) undertook two surveys of the Bay in 1990 and 1991 and recovered 220 and 354 sources. Subsequent surveys have been undertaken by various contractors for the MOD during which sources are removed from the foreshore to provide some protection to beach users. Figure 2 charts the annual finds of sources until September 2011. SEPA then initiated their own survey work at Dalgety Bay and in the period October 2011 to October 2012 and recovered over 800 sources from 36 surveys. The activity of sources recovered ranged from around 1 kBq to 76 MBq and Figure 3 provides a Log_{10} distribution of the ^{226}Ra source activities. Over the same period since, several hundred additional sources were recovered by the MOD contractors over the 800 m coastline.

The physical origin of the radium sources is part of an on-going site investigation. Figure 1 shows the locations where the majority of the sources have been recovered and is centred around the site of the sailing club. Contaminated materials range sources (typically < 3 mm in dimension), clinker (up to a few 10s mm in dimension) from incineration occasionally associated with ash deposits. Wilson and Tyler (2012), describes SEM-EDS analysis on a small sub sample of particles that shows some sources to have been incinerated whilst others appear to be intact and all sources have high concentrations of Zn and S, which can be attributed to radium paint.

Site contamination and any subsequent designation requires an assessment of the likely doses that could be received by individuals, children or adults, through the accidental ingestion of contaminated material on the foreshore areas. An *in vitro* methodology is described to assess the solubility of sources. Results are presented from three separate batches of digestion experiments to assess the possible

committed effective dose to an individual. The experiment is designed to mimic the acidic, enzymatic and temperature conditions of the stomach and small intestine. Whilst the approach to estimate solubility is not specifically novel, the complexity of the sources raises a number of measurement challenges and the results provide an important approach to enable the RCL regulations to be interpreted.

2. Materials and Methods

2.1 Source Selection

The radium sources selected for digestion were initially selected on the basis of their ingestible size (Lovitz *et al.,* 2010) and activity. In 2011, a total of 45 samples were collected from the MOD's contractor at Dalgety Bay, isolated from the surrounding sedimentary matrix and radium content estimated via gamma spectrometry. The size was measured by placing the sources on 1 mm graph paper and estimated via digital photography. From the 45 samples, a sub sample of ten sources was selected for digestion within simulated stomach solutions. The selection covered a range in ^{226}Ra activities with source dimensions less than 5 mm to allow for ingestion. One exceptional item was a copper aircraft dial (Figure 4), with 166 kBq of ^{226}Ra, to estimate the general availability of radium from such artefacts irrespective of size. Following the large number of finds made by SEPA later in 2011, a second batch of 30 physically smaller sources were selected. These were all less than 5.5 mm in the longest axis with ^{226}Ra activities ranging from 2 kBq to 50 kBq. Following the analysis from the first two batches, a third batch of 18 sources was selected in early 2012, focused around the estimated threshold activity likely to deliver a 100 mSv dose to an infant, all with dimensions less than 5 mm in the longest axis. To this set of 18, an additional two sources were added: a 150 kBq of ^{226}Ra (2x2 mm) particle, resembling a centre pin head of a dial (Figure 5); and a 1.26 MBq ^{226}Ra (10 x 7 mm) piece of clinker (Figure 6). A total of 60 radium sources were digested and each batch of digestions included a blank sample.

2.2 Source Digestion

The committed dose is considered from *in vitro* digestion of sources using simulated solutions for the stomach solution and small intestine solution.   To ensure consistency with previous investigations on Dounreay hot particles (DPAG, 2006), the method for digestion was based on Harrison *et al.,* (2005). The composition of the 'stomach' solution and 'small intestine' solution are presented in Tables 1 and 2 respectively.   The chemicals listed in Table 1 were dissolved in 0.5 litre of distilled water and adjusted to a pH 2 using hydrochloric acid for a final volume of 1 litre. For the small intestinal solution, the chemicals listed Table 2 were dissolved in 0.5 litre of distilled water, adjusted to pH 7 with 5 M sodium hydroxide solution to produce a final volume of 1 litre. Both solutions were pre-warmed to body temperature (37 °C).

All sources, with the exception of the instrument dial, were placed into prewashed 100 ml beakers. The mass of the source was measured by weighing the source in its storage container and by reweighing the container, once the source had been removed, on an ISO17025:2005 calibrated 5-figure mass balance. The containers were checked for radium contamination using a Thermo BP19 RD beta/gamma detector coupled to an Electra. Where gross beta/gamma counts were above background, the container was swirled with a small amount of the stomach solution and added to the source until the count rate returned to background. If the source had to be removed from the container by rinsing, then the containers were dried before being weighed empty.   The instrument dial was kept in its polystyrene container (Figure 4). Each source was photographed, prior to each stage of the

digestion process. Each beaker was filled to 30 ml with the 'stomach' solution and placed in an incubator for 8 hours maintained at 37 °C. A total of 100 ml of 'stomach' solution was added to the instrument dial so that it was completely immersed. Source immersion was staggered by 5-10 minutes to allow sufficient time for filtering and transfer to the 'lower intestine' solution. The first 10 sources were agitated hourly by hand. An orbital incubator was used for the remaining two batches of samples to maintain constant gentle agitation. The pH of the solution was checked with pH paper 5 minutes after immersion and then every hour during the digestion period. Where pH was observed to drift, the pH was adjusted to the appropriately.

Following 8 hours of immersion the solution containing the source was filtered under vacuum through 5 µm Whatmann filter papers (48 mm diameter). All the glassware was rinsed first with 'stomach' solution and then distilled water through the filter paper and checked for contamination and rinsed again until no contamination was detected. The filtrate from the 'stomach' solution was carefully poured into suitably labelled Marinelli containers for counting by gamma spectrometry and the volume was made up to the calibrated 350 ml volume with distilled water. The filter paper and source were used for the final phased of digestion within 'lower intestine' solution and the experiment procedure was repeated.

2.3 Gamma spectrometry analysis

Gamma spectrometry analysis was used to determine the activities of the filtered 'gut' solutions and the individual sources.

The Marinelli beakers were counted on one of four Ortec HPGe detectors calibrated under ISO17025:2005 accredited procedures within the Environmental Radioactivity Laboratory at the University of Stirling (UKAS testing lab 2751). Each detector is shielded with 100 mm of lead with a copper cadmium lining. The detectors were calibrated with a range of environmental matrices of varying densities and compositions spiked with Isotrak QCY48 spike with the additional of a DKD RBZ B44 ^{210}Pb spike. Spectra were processed using Gamma Vision 32 and spectral libraries are regularly updated with data from the Bureau International des Poidset et Mesures (BIPM). All calibrations were traceable to international standard reference materials and the laboratory is a regular participant in performance testing trials. Radionuclide activities are reported as total activity (Bq).

The irregular shape and size of the radium sources and artefacts required a calibration that was as independent of the effects of geometry as possible. To minimise the effect of geometry, an IAEA RGU (uranium reference material) sample was pelletized into a disk 48 mm in diameter and 11 mm in thickness with a nominal activity concentration of 5000 ± 300 Bq kg^{-1}. The pellet was placed within a petri dish of similar geometry and sealed with epoxy resin. The sealed RGU pellet was then set aside for over three weeks to allow radon equilibration. The RGU pellet was then supported at known distances from the face of a GEM HPGe detector and an efficiency calibration calculated using the natural series lines from ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{234}Th and ^{210}Pb. The efficiency calibration was then tested with a ^{137}Cs point source of known activity. The optimal calibration distance was determined to be at 300 mm from the face of the detector, beyond which the Pb shielding housing the detector and sample could not be closed. At 300 mm distance the activity of a ^{137}Cs point source was over-estimated by 25 % of the reference value. The 25 % systematic uncertainty was incorporated into the uncertainty budget and is likely to be a reasonable uncertainty in the activity determination of the radium artefacts for ^{214}Bi and ^{214}Pb. Lower energy gammas may be influenced by self-absorption influences, especially in larger heterogeneous objects and the uncertainties may be greater than

the 25% estimated. However, geometrical effects may be less important at lower gamma energies than higher energies. These two effects in part cancel each other out for more complex source geometries. Nevertheless an uncertainty of 25% due to geometrical variation was assumed.

For source activity estimation, all samples were typically counted to single percentage 1 sigma Poisson counting uncertainties on the key [214]Bi and [214]Pb gamma photon full energy peaks. Gamma Vision 32 was used to estimate each source activity with the efficiency calibration derived from the IAEA RGU pellet.

2.4 Calculation of the committed effective dose

The calculation of committed effective dose is necessary for SEPA's interpretation of the RCL Regulations. The *in-vivo* digestion experiment provides indicative estimates of the solubility of the radium sources. The potential doses are calculated using standard ICRP 72 (ICRP, 1996) where dose coefficients were derived from age specific biokinetic models and take account for changes in gut uptake, body mass, geometry with age of the individual and includes excretion pathways. Gut adsorption coefficients f_1 were also used from ICRP 72 as these, together the dose coefficients, have been adopted by the International Atomic Energy Agency for their International Basic Safety Standards for protection against Ionising Radiation (1996) and Euratom Directive (EC, 1996).

Assuming that the [226]Ra daughters are in equilibrium, the contributions from [226]Ra, [214]Bi, [214]Pb, ^{210}Pb, [210]Bi and [210]Po are included in the calculation. It is assumed that [210]Bi and [210]Po are in equilibrium with ^{210}Pb, which is an important assumption as 66 % of the dose for a < 1 year old infant is dominated by [210]Po and 55 % for an adult. This assumption of equilibrium has been validated by results from a recent IAEA TEL 2011-03 intercomparison exercise and from the UK's Health Protection Agency (Brown, pers comm)

3. Results and Discussion

The source activities measured at the start used for this work are presented in Appendix A. The percentage activity in solution following the experimental work is given in Appendix B for the 'stomach acid' and Appendix C for the 'lower intestine'. The total activity lost to solution is presented in Appendix D. Each Appendix is split into three parts of each individual batch of digestions. Here, we present the results and discuss the key findings.

3.1 Radium Dissolution

The results tabulated in Appendix A provide the details of the activities measured from each source, and show good agreement between [226]Ra and the daughters through to ^{210}Pb, indicating equilibrium in the decay series below [226]Ra. However, analysis of the gut solutions (Tables in Appendix B and C) show significant disequilibrium in the decay series with [226]Ra and ^{210}Pb having similar activities with a ratio of around 1:1. In contrast, the [214]Bi and [214]Pb activities were very much lower. The obvious disequilibrium between ^{210}Pb and [214]Pb indicates that ^{210}Pb is being preferentially supported by [218]Po and [214]Po. The 'stomach' solution consistently has much higher amounts of [226]Ra and ^{210}Pb compared with 'lower intestine' solution, largely reflecting the acidity of the 'stomach' solution and this is shown in Figure 7.

In the first phase of ten samples, four samples released more than 1 kBq of ^{226}Ra and/or ^{210}Pb into the combined gut solutions. This included 18 kBq of ^{210}Pb from the dial, confirming that ^{226}Ra and ^{210}Pb and ^{210}Po are biologically available from contaminated aircraft artefacts. Of importance here however, was a 4 x 5 mm source of 16 kBq that released 4 kBq of ^{226}Ra and 3.2 kBq ^{210}Pb, providing the highest solubility of about 25 ± 10 % (Figure 8; Table D1).

The second phase of thirty samples focused entirely on sources that were digestible, with a mean size of 1.7 mm (maximum 4 mm) in the smallest dimension and 2.3 mm (maximum 5.5 mm) in the longest dimension with activities ranging from 2 to 50 kBq. In this sample set, only 4 sources released 1 kBq of ^{226}Ra and/or ^{210}Pb in the combined simulated gut solutions (Figure 8). However, the maximum solubility was again around 25 ± 10 % although in this case from a sub 1 mm sized 2 kBq particle (Figure 8; Table D2).

From the first two experiments, the maximum 25% dissolution for ^{226}Ra and ^{210}Pb could result in 100 mSv from a 20 kBq source for a young child. With this in mind, eighteen of the final phase of source digestions focused on small digestible sources with an activity around this value. In addition, the 1.26 MBq source (Figure 5) and 150 kBq pin head (Figure 6) were also digested. From these 20 samples, five resulted in activities greater than 1 kBq of ^{226}Ra and/or ^{210}Pb in the combined 'gut' solution. A total of 13 kBq of ^{226}Ra and 10 kBq of ^{210}Pb was measured in the combined gut solution from the 1.26 MBq source, representing about 1 % of the total activity. For the 150 kBq pin head, 3 kBq of ^{226}Ra and 10 kBq of ^{210}Pb were measured in the gut solution, representing 8% of the ^{210}Pb activity. Notably, the 13 kBq source, which resembled a small piece of clinker, yielded 4.5 kBq of ^{226}Ra and 3.9 kBq of ^{210}Pb in the combined gut solution. This represents a dissolution of about 35 ± 10 % (Figure 8; Table D3).

The results show that the amount of activity that is removed from the sources during digestion is highly variable. The two artefacts that represent parts of an instrument panel lost about 10 % of their activity in the digestion process. However, it is not known whether the effect of being exposed to environmental conditions over *ca.* 60 years has influenced this value. Some radionuclide activity from the instrument dial remained within the pot prior to the digestion process indicating that the Radium was still mobile, which may also have been the case for the dial in the environment. The amount of radium available for dissolution may also depend on where the radium is located within the sample, i.e. buried within the clinker or on the surface, or its chemical form if incinerated where burning was undertaken with aviation fuel or wood. The factors influencing radium availability require further investigation. These values were considered to represent the worst-case scenario when interpreting the results for the RCL regulations and the requirements for targeted source removal and site remediation.

3.2 Calculation of Committed Effective Dose

The results from the combined simulated 'gut' solutions were used to calculate the committed effective dose for the different age groups using ICRP 72 fractional absorption values (f_1) for the GI tract and dose coefficients. Figure 9 shows the variation in committed effective dose from each of the 60 sources digested. Of these 60 sources, seven exceed the 100 mSv threshold for infants (3 months), two for 1 year olds and one for 5 year olds. Several additional sources lie close to the 100 mSv threshold especially for the younger age ranges. The lowest source activity resulting in 100 mSv is a 10 kBq source for an infant. The number of sources on the foreshore at any one time is not established yet, but is currently estimated to be at

least one hundred and perhaps as many as several hundred. From the data presented, it is possible to suggest that around 10 % of this population may have the characteristics necessary to release sufficient activity to provide in excess of 100 mSv to an infant on the foreshore.

If we assume that 35 % of a source's ^{226}Ra and ^{210}Pb (including ^{210}Po) can be dissolved when digested, and that ^{214}Bi and ^{214}Pb is only present to around 1% of the source activity, then we can calculate the source activity threshold for each age group and this is shown in Figure 10. These results indicate that doses of 100 mSv may be delivered by a source of around 8 kBq to a 3 month infant, 22 kBq to a toddler (1 yr) and 40 kBq to a 5 year old child. This increases to around 140 kBq for an adult. Figure 3 shows that all these source activities have been detected and recovered from the foreshore areas since September 2011, and that in excess of 25 % of sources exceed 8 kBq in activity. Sources with activities greater than around 20 kBq could provide a committed effective dose in excess of the 100 mSv value for young children known to occupy the area.

The approach using the maximum solubility may be argued to be highly cautionary. The solubility data are non-normal and a log transformed distribution yields a corrected mean solubility of 7.59 %, with an upper 95 percentile of the distribution giving a solubility of 20 % using the approach by Gilbert (1987). If we take the corrected mean solubility value, then 100 mSv can be delivered by a source of 33 kBq to an infant, a source of 100 kBq to a toddler (1 yr), a source of 250 Bq to a 10 year old child and source of 600 kBq to an adult. Figure 3 also shows that these increased source activities are also present on the foreshore, although they are proportionately fewer in number.

The assumptions behind some of the ICRP values should also be acknowledged. Importantly, the absorption across the GI tract assumes that no other foods are being co-digested. ICRP 56 (ICRP, 1989) states that elements may be more readily absorbed when co-digested with other foods. In this scenario, 100 mSv may be achieved with lower activity sources. In addition, ICRP 72 assumes complete f_1 absorption of ^{210}Po and 0.6 for ^{226}Ra compared with absorbed fractions of 0.5 and 0.2 for the older age groups. Although this is based on animal data and supported by limited human data, these values will carry some uncertainty.

Nevertheless, within the context of SEPA's RCL regulations, whether or not the corrected mean or maximum solubility is considered and accounting for the uncertainty with the ICRP gut absorption conversion coefficients, these results indicate that the probability of encounter need not be taken into consideration. In contrast to other situations, such as at Sellafield and Dounreay, where hot particles are routinely detected and recovered from the foreshore, the probability of encounter for the Dalgety Bay sources cannot be purely described by chance. At Dalgety Bay beachcombers proactively seek out items of interest, such as that shown in Figure 4 and 6. The monitoring and recovery of these sources is therefore an important component of the interpretation of the RCL regulations until the sources of these sources can be established and the site remediated. As with the Dounreay example (Tyler et al., 2010), the number of sources on the foreshore is likely to change in relation to processes of sediment accumulation and erosion, and the frequency of sediment turnover (Tyler *et al.,* 2010). Access to the site is now restricted to the public and fences demarcate the areas of greatest concern, i.e. where the largest numbers of sources have been recovered.

Comment [AT1]: Expand here

4. Conclusion

It has now become widely acknowledged that land contaminated with radium due to 20th century civilian and military pursuits has the potential to result in significant radiological harm. This paper presents the results from one aspect of the interpretation of the RCL in relation to the potential committed effective dose from radium contaminated particles and artefacts at a coastal site at Dalgety Bay that had once been a military airfield. Significant numbers of sources have been recovered from the foreshore areas. We show that the solubility of the sources in simulated 'gut' solutions is highly variable, but the highest value is about 35 %. In this study, ^{210}Po was assumed to be in equilibrium with ^{210}Pb and committed effective dose of 100 mSv could be received from a 20 kBq source to a toddler (1-2 years old) and 130 kBq from an adult. This range of source activities has been recovered from Dalgety Bay. As with the Dounreay and Sellafield examples, source numbers on the foreshore are likely to be change in relation to the state of the foreshore. Unlike the hot particles detected and recovered on Sellafield and Dounreay beaches, some sources are collectable military artefacts such as instrument dials and are actively sought by beachcombers, which must be factored into the chance of encounter and risk.

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TABLES

Table 1 Composition of the 'stomach' solution, after Harrison *et al.,* (2005)

Table 2 Composition of the 'small intestine' solution, after Harrison *et al.,* (2005)

FIGURES