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Radionuclide partitioning to sewage sludge - A laboratory investigation

Science Report – SC020150/SR1

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Steve Killeen

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Head of Science

Executive summary

This project, *Radionuclide partitioning during sewage treatment – A laboratory investigation*, was undertaken by Enviros Consulting Ltd with support from WRc and the University of Plymouth. It is one of two pieces of work commissioned by the Environment Agency to investigate the radiological implications of authorised discharges of liquid radioactive waste to sewer.

This first report outlines a series of laboratory experiments to generate robust sludge retention factors for the Environment Agency's Initial Assessment Tool, a spreadsheet model to assess the radiological impact of discharges to sewer on a number of candidate critical groups.

This study explored the solid-solution behaviour of a range of radio-elements (Br; Ca; Co; Cu; Fe; Ga; I; In; La; Mn; Ni; P; Re (as an analogue for Tc); S; Sr; Th; U; V; and Y) using stable isotope tracers and high resolution ICP-MS for the analysis. The elements were chosen for their relatively high radiological risk (a function of the amount discharged and the radiotoxicity of the radioisotopes of each element) and the existence and validity of literature data on their behaviour during sewage treatment.

Experiments were conducted using specialist laboratory facilities and sewage materials sourced from a domestic housing estate. These assessed:

- partitioning processes likely to occur during transport through the sewer;

- the transfer of tracer to primary solids during primary settlement (the primary settlement retention factor, PSRF);

- the transfer of tracer to solids during secondary (activated sludge) treatment (the activated sludge retention factor, ASRF);

- the overall sludge retention factor, SRF, likely to be achieved in a typical sewage works.

The study found good agreement between the SRFs determined here and those in the Environment Agency tool for Br, Co, Fe, Ga, In, P, Re (as Tc), S, Th, U and V. However, for others agreement was less good, particularly for Ca, I, La, Mn, Sr and Y. For these elements, SRF values in the Environment Agency tool may need updating.

Overall, the study has demonstrated that not all values in the Environment Agency model are fully underpinned and that further work is required to assess these.

Acknowledgements

Contents

1   Introduction

This project, *Radionuclide partitioning during sewage treatment – A laboratory investigation*, was undertaken by Enviros Consulting Ltd with support from WRc and the University of Plymouth. It is one of two pieces of work commissioned by the Environment Agency to investigate the radiological implications of authorised discharges of liquid radioactive waste to sewer (see Punt *et al.*, 2007).

1.1   Background

The Environment Agency has a statutory responsibility to assess and authorise radioactive discharges to air, water or sewer under the Radioactive Substances Act 1993 (RSA 93). Within England and Wales, the Environment Agency has issued about 130 permits for the discharge of radioactive substances to sewer (the primary disposal route for liquid effluent from non-nuclear licensed sites). These discharges primarily come from hospitals, universities and research laboratories ('small users'), although there is also a small contribution from nuclear licensed sites.

1.2   Regulatory assessment tools

Liquid effluent discharges to sewer and subsequent treatment of sewage results in two distinctly different waste streams that may have elevated levels of radioactivity. These are treated sewage effluent discharged to rivers or the sea, and digested and/or dewatered sewage solids that may be spread onto land, incinerated for energy recovery or disposed of to landfill.

As part of this authorisation process, the Environment Agency must assess the potential radiological exposure to sewer workers and members of the public from treated effluent discharged to surface waters or the sewage solids spread onto farmland.

The Environment Agency has developed a suite of Initial Assessment models (Allott and Titley, 2005; Allott *et al.*, 2006; Lambers and Thorne, 2006) which estimate the dose to a range of critical groups from discharges to sewer. These include:

- workers in sewage treatment works exposed to radioactivity in untreated, solid and effluent phases (via inadvertent ingestion, inadvertent inhalation and external exposure routes);

- farming families exposed through the application of sewage sludge to land (via inadvertent ingestion, inadvertent inhalation, external exposure and consumption of foodstuffs grown or raised on the land);

- children exposed whilst playing in streams receiving treated sewage effluent (via inadvertent ingestion of river water and external exposure on banks or through water immersion);

- families exposed as a result of angling activities in rivers receiving treated sewage effluent (via consumption of fish);

- consumers of foodstuffs contaminated from irrigation with river water which has received treated sewage effluent;

- estuarine/coastal fishermen who catch and consume fish from areas that receive treated and untreated sewage effluent, where the fish may bioaccumulate activity from the water within which they live.

A key factor within this assessment is the proportion of activity discharged to sewer that is likely to become associated with sewage sludge and solids, and hence may enter the human food chain though the spreading of these solids onto land (terrestrial exposure pathways), or which may remain dissolved and be discharged to surface waters (aquatic exposure pathways).

Within the Initial Assessment model for discharges to sewer, the partitioning of activity between particulate and dissolved fractions (and hence terrestrial and aquatic exposure pathways) is expressed as a single value, the sludge retention factor (SRF). The SRF is the proportion of total activity entering the sewage works that becomes associated with sludge and solids, the remainder being discharged with treated effluent. This assessment method also considers the loss of activity during decay. Although radioactivity that remains predominantly in the dissolved phase will pass rapidly through a treatment plant (in 24 hours), activity associated with solids may reside on site for weeks or months before being applied to land. In this instance, for relatively short-lived radionuclides (those with a half-life of less than 10 or a few 10s of days), decay can lead to greater than 50 per cent loss of activity in solids before they leave the sewage works.

Sewage sludge may also be used as waste-derived fuel (WDF) and co-incinerated for energy recovery. In this instance, sludge is dewatered and transferred for incineration within a few days, with less opportunity for loss of activity by decay.

1.3   Previous studies

A number of authors have estimated the SRF and a review is provided by Ham *et al*. (2003). Values reported by Ham *et al*. (2003) were derived from a range of studies which assessed different end points and employed different sampling or analytical methods. As a result, many of the SRFs quoted have a wide range of values. In addition, for some radionuclides no data exist at all. There is therefore a need to derive, through experimental work where practicable, a more comprehensive and realistic dataset on the partitioning of radionuclides between the sludge and liquid effluent fractions of treated sewage. A further review of the literature is given in Appendix 1.

1.4   Project aims

This project aimed to derive partition factors from laboratory experiments to describe the solid-solution partitioning of radionuclides during sewage treatment, and to present these as SRFs for use in Environment Agency assessment models.

The project was carried out in three stages, by:

- prioritising the radionuclides to include in the study;
- developing an experimental methodology;
- analysing and interpreting the results.

2   Assessment methods

The project methodology is outlined in this chapter and further details are given in Appendices 2 to 6.

2.1   Prioritisation

The prioritisation process aimed to identify a subset of key radionuclides for assessment from a list of 85 of the main radionuclides discharged to sewer in England and Wales (see Appendix 2 for further details).

Radionuclides were assessed on the basis of:

- the number of sites discharging a particular radionuclide and the potential cumulative input to sewage works across England and Wales (based on the authorisation limit) − for short-lived radionuclides, the loss due to decay during transport through the sewer was accounted for in this calculation;
- relative radiotoxicity based on ingestion, inhalation and external exposure dose coefficients;
- availability of information with critical review of its reliability;
- practicality of assessment (availability of suitable tracer isotopes and methods of determination).

Where different isotopes of the same chemical element were considered, these were combined to provide a value specific to each element (rather than a radioisotope-specific one).

This process prioritised the following radio-elements for further study:

- Bromine (Br)
- Calcium (Ca)
- Cobalt (Co)
- Copper (Cu)
- Gallium (Ga)
- Indium (In)
- Iodine (I)
- Iron (Fe)

- Lanthanum (La)
- Manganese (Mn)
- Nickel (Ni)
- Phosphorous (P)
- Rhenium (Re)*
- Strontium (Sr)
- Sulphur (S)
- Thorium (Th)

- Uranium (U)
- Vanadium (V)
- Yttrium (Y)

*Re was used as an analogue for Tc

2.2   Experimental set up

The aims of the experiments were to replicate as closely as possible the physical, chemical and biological processes that occur during sewage treatment (see Appendix 3) and to assess the partitioning of a tracer substance which mimicked the chemical behaviour of the radio-elements described above. To achieve this, specialist waste water laboratories at WRc were used, which had a supply of domestic sewage from a local housing estate and experimental systems designed to replicate primary (physical settling) and secondary (aerobic biological digestion) stages of sewage treatment.

The experiments were set up to assess solid-solution partitioning during sewage transport through the sewer, primary treatment through gravitational settlement and secondary treatment using activated (aerated sludge beds), as illustrated in Table 2.1.

Table 2.1: Summary of the experiments

Sewage treatment			Experiments carried out in this study	
Sewage stage	Time	Process occurring	Experiment	Sampling time
Transport of raw sewage to works	One to five hours	Homogenisation	Equilibration with crude sewage up to four hours	Four hours
Primary settlement of crude sewage	Two to eight hours	Settlement of solids leaving primary effluent	Settling tubes used to assess loss via settlement up to 24 hours	Zero, one, two, five and 24 hours
Secondary treatment in activated sludge beds	Around 15 hours for dissolved fraction and 10 days for particulate	Microbial assimilation and removal of particulate and dissolved carbon in effluent	Porous pot used to mimic activated sludge beds over a typical sludge retention time of 10 days	Zero, four and eight hours; one, two, three, six, eight and 10 days.
Tertiary treatment of final effluent	Around one hour	Remove pathogens and/or trace elements/nutrients	Not assessed	
Anaerobic digestion of primary and secondary sludge	Up to 12-20 days	Remove pathogens in sewage solids to be applied to land		
Dewatering of sludge	Less than 24 hours	Remove water to produce final dewatered solids		

The experimental set up is described in more detail in Appendix 4.

2.3 Sample treatment and analysis

To determine dissolved tracer concentrations, samples were filtered using glass fibre membranes and the filtrate analysed. The total tracer fraction (in the dissolved and particulate phase) was determined by acidifying (to pH 1 with nitric acid) a bulk sample of unfiltered effluent for 24 hours, after which it was filtered and the filtrate analysed.

All samples were analysed by high resolution inductively coupled plasma mass spectrometry (HR ICP-MS) using a VG Axiom instrument at the University of Plymouth. High Resolution ICP-MS has the advantage of achieving trace (ppb-ppm) and ultra-trace (ppq-ppb) elemental analysis and can be used to distinguish between different isotopes of the same element. However, it could not be used for Ca-40 analysis because of interference from the presence of Ar-40 in the ICP-MS carrier gas. Given the high concentrations of Ca present, it was possible to measure the element using flame atomic adsorption spectrometry.

Other variables, such as the concentration of suspended solids and pH of the sewage, were monitored at the WRc facility.

2.4   Determination of the sludge retention factor

The Environment Agency assessment method considers the partitioning of radio-elements across the complete sewage treatment process and this partitioning is represented as a single aggregate factor. The partitioning process is complex, but dominated by two distinct processes at different points in the treatment cycle (see Appendix 3). In order to meet the needs of the assessment method, this study calculated a single sludge retention factor (SRF) which combined the partitioning from the two main processes: the primary settlement retention factor (PSRF) and aerated solid retention factor (ASRF) from the secondary biological treatment. This is illustrated in Figure 2.1 and described further in Appendix 6.
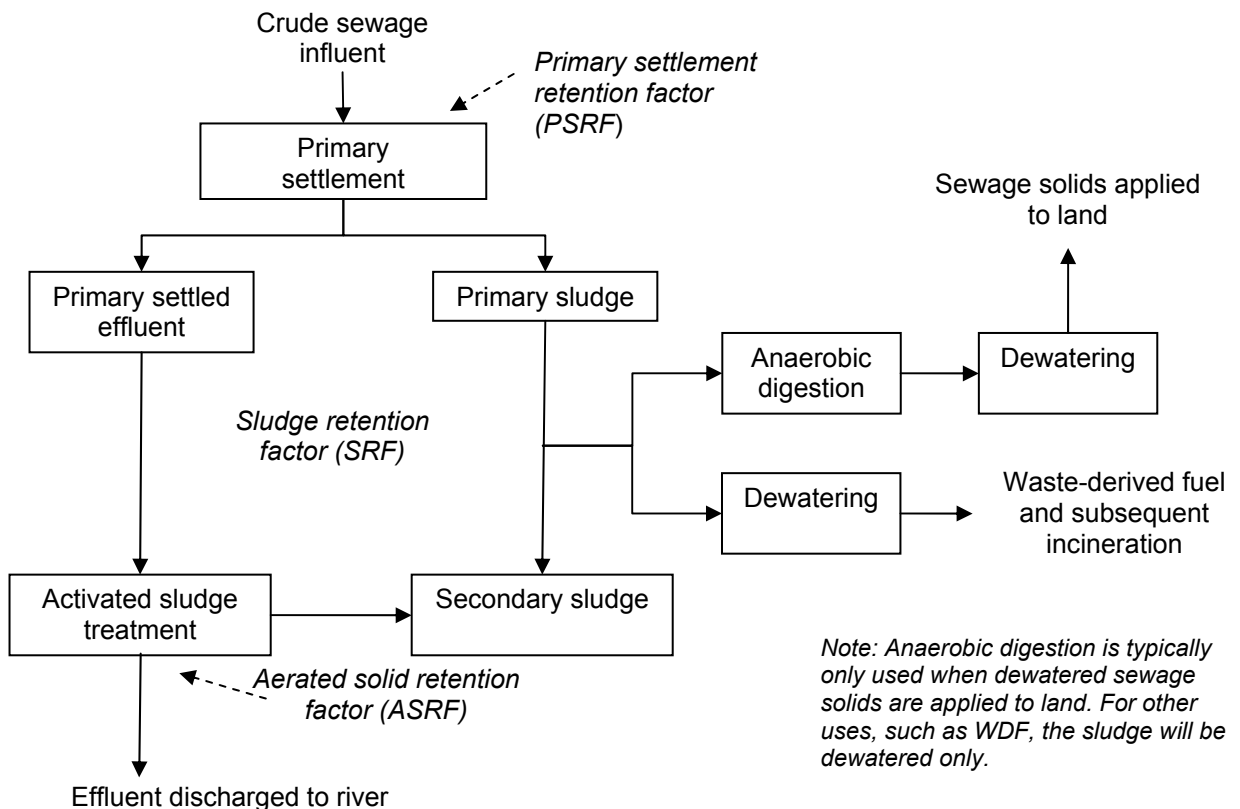
Figure 2.1: Diagram of the sewage treatment process

3 Sludge retention factors

Sludge Retention Factors calculated in this study are presented in this chapter and compared to those currently used in the Environment Agency Initial Assessment Tool. Further details are given in Appendix 6.

3.1 Sludge retention factors

Sludge Retention Factors calculated in this study are summarised in Table 3.1.

Table 3.1: SRF values calculated in this study, reported in the literature and/or currently used by the Environment Agency

| Radio-element | Sludge retention factors (SRFs) | | |
	Calculated in this study	Reported in the literature	Used by the Environment Agency
Br	0.01	Not available	0.1[a]
Ca	0.4	Not available	0.1[a]
Co	0.9	0.31-0.8	0.8[b]
Cu	1.0	0.7-0.89	Not included
Fe	0.9	0.9	0.9[b]
Ga	1.0	Not available	0.9[c]
I	0.8	0.01-0.4	0.2[b]
In	0.9	Not available	0.9[c]
La	0.7	Not available	0.1[c]
Mn	1.0	0.5-0.7	0.5[b]
Ni	0.9	0.51	Not included
P	0.8	Not determined	0.8[d]
Re (Tc)	0.01	Not available	0.1 (Tc)[a]
S	0.01	0.08-0.1	0.1[b]
Sr	0.4	0.1	0.1[b]
Th	0.9	Not available	0.9[a]
U	0.01	Not available	0.1[a]
V	0.9	Not available	0.9[c]
Y	1.0	Not available	0.1[c]

[a] Derived from partition coefficient values for organic soil.
[b] Based on values given in NRPB-W32 derived from Ham *et al*. (2003).
[c] Based on expert opinion.
[d] Cardiff assessment.

3.2 Comparison with previous studies

Sludge Retention Factors values calculated in this study were compared to those currently used in the Environment Agency assessment tool (Table 3.1).

Very good agreement was obtained between SRFs determined here and those in the Environment Agency tool for Br, Co, Fe, Ga, In, P, Re (Tc), S, Th, U and V. However, for others the agreement was less good, particularly for Ca, I, La, Mn, Sr and Y. These elements may require special consideration, as explained below.

For Ca, La, and Y, there is no data to underpin the values used in the Initial Assessment tool; however, the partitioning behaviour measured in this study is

consistent across the three experiments and the values derived here are therefore likely to be more realistic than those currently used.

In this study Mn had a high uptake in solids, particularly during secondary treatment. This is higher than the values quoted in the literature, but is relatively consistent between experiments.

The SRF for I determined here is significantly higher than that given in the literature. Although the value of 0.2 used in the Initial Assessment tool is probably at the lower end of the likely range, the value of 0.8 determined here may be unrealistic as there was a large range in results from the different experiments undertaken in this study. Iodide (I^-) was used as a tracer, but only small amounts of I^- will adsorb onto the sludge under typical conditions. This is because the sludge is organic rich and is negatively charged. However, porous pots are fully oxygenated environments which means that I^- could be readily oxidised to iodate, $IO3^-$. Iodate has a strong tendency to complex with Ca to form $CaIO_3^+$ (similarly with Mg). Bearing in mind that there is about 25,000 times more Ca than I in the pots, the formation of these complexes is highly likely. $CaIO_3^+$ is positively charged and will, therefore, be attracted to the negatively charged particle surface. Nonetheless, porous pots replicate the highly oxygenated conditions of activated sludge treatment, so it is unclear why there was such a difference. This may have been an artefact of sample treatment, but it was not possible to assess this further.

4   Conclusions

This study adapted and applied established techniques to assess the solid-dissolved partitioning of a number of elements, to provide essential data for the Environment Agency's Initial Assessment Tool. The conclusions of the study are that:

- It is possible to replicate the physical, chemical and biological processes of sewage treatment under carefully controlled laboratory conditions. Preliminary partitioning and settlement experiments can be easily established with minimal specialist equipment. However, replicating the activated sludge process is labour-intensive and here, the experimental set up required several weeks to stabilise before it could be used. The system requires a daily supply of freshly collected primary sewage effluent and the stability of the bacterial culture needs to be carefully monitored and controlled.

- Use of stable element tracers and analysis via HR ICP-MS is consistent with Environment Agency best practicable means (BPM) guidance, but the complexity of the sample matrix and the high organic loading means that instruments may need to be frequently cleaned and recalibrated.

- The study shows that the SRFs for Br, Co, Fe, Ga, In, P, Re (Tc), S, Th, U and V currently used in the Environment Agency Initial Assessment Tool are entirely consistent with our findings. However, those of Ca, La, Mn, Sr and particularly Y may be underestimated. The currently used SRF value for iodine is probably at the lower end of its range of behaviour, but this assumption is probably conservative as exposure pathways are likely to be dominated by effluent discharged to surface waters rather than solids applied to land.

- Assessing the influence of chemical form of the radionuclide discharged to sewer was beyond the remit of this project. However, it is probable that where the radionuclide exists as a complex chemical compound, its behaviour is likely to be different to that of ionic forms.

5 Recommendations

It is recommended that some values used in the Environment Agency's assessment tool are updated to accommodate the conclusions of this study (Table 5.1). It is also clear that other values may not be accurate and may need careful review and potentially further experimental work.

For short-lived radionuclides, increasing the SRF will mean that a greater proportion of the radioactivity will be estimated to be lost via decay before off-site exposure routes are realised. Any modification to the Initial Assessment Tool should therefore consider the sensitivity of dose predictions to changes in the SRF and compare these to the importance of other parameters used in the model. However, a user of the Initial Assessment Tool cannot modify the SRF, so it is therefore not possible to easily assess the radiological implications to critical groups through changes in the SRF. Nonetheless, an alternative tool based on the same formulation could be developed to allow the modification and testing of these parameters. Such a tool could be used to assess the sensitivity of dose results via terrestrial or aquatic pathways, and to prioritise radionuclides for study.

Table 5.1: Recommended SRFs

Radio-element	Sludge retention factors (SRFs)		
	Calculated in this study	Used by the Environment Agency	Recommended revision
Br	0.01	0.1	No change
Ca	0.4	0.1	0.4
Co	0.9	0.8	0.9
Fe	0.9	0.9	No change
Ga	1.0	0.9	No change
I	0.8	0.2	0.8
In	0.9	0.9	No change
La	0.7	0.1	0.7
Mn	1.0	0.5	1.0
P	0.8	0.8	No change
Re (Tc)	0.01	0.1 (Tc)	No change
S	0.01	0.1	No change
Sr	0.4	0.1	0.4
Th	0.9	0.9	No change
U	0.01	0.1	No change
V	0.9	0.9	No change
Y	1.0	0.1	1.0

Site-based monitoring can generate highly useful data (see Punt *et al*., 2007), and this should be explored as a possible means of validating assumptions in the model, along with simpler laboratory methods.

References

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Appendix 1: Literature review of sludge retention factors

An important part of this study was to evaluate the data available on the behaviour and partitioning of radionuclides during sewage treatment.

A review of partitioning factors was previously undertaken on behalf of the Environment Agency by Ham *et al*. (2003). This review and other studies were critically assessed here to determine the extent and quality of data available in published sources, to feed into the selection process to determine which radionuclides to include in this project. Subsequent to this project, further field data became available (Punt *et al*., 2007).

The chemical behaviour of radionuclides during sewage treatment is more likely to be element- rather than isotope-specific. Data on the partitioning of stable isotope counterparts was therefore included, where available.

A1.1 Partitioning data for radionuclides during sewage treatment

A review of partitioning data conducted by Ham *et al*. in 2003 forms the basis for many of the values adopted within the Environment Agency assessment tool.

The Ham *et al*. (2003) review used information derived from field studies assessing radionuclide and stable partitioning, but in several places had to rely on expert judgement to suggest SRFs. With any such review, inconsistencies and uncertainty in data can arise due to the sampling, treatment and analysis regime used, the interpretation of results and the type of sewage works being studied.

A number of studies have also shown that the chemical form of a radionuclide can have a significant effect on its behaviour during sewage treatment. This has been demonstrated for iodine-131 and further data are given in the Section A1.1.3.

A1.1.1 Tritium

Experimental data for H-3 behaviour during sewage treatment is presented in Palfrey *et al*. (2001). This data was derived from porous pot experiments which were used to mimic sewage treatment processes during secondary treatment using activated sludge beds.

A number of tritiated compounds were assessed using the porous pots to take account of the various organically bound forms in which H-3 may be found within sewage. The percentage partitioned to sludge ranged from six to 22 per cent (average 12 per cent) with the exception of tritiated water (HTO), where the percent removal to sludge was found to be reduced to just 0.1 per cent.

In their review, Ham *et al*. (2003) concluded that, in the absence of information on the chemical form in which H-3 occurred, a default SRF for organically bound H-3 of 15 per cent should be used. No information was provided on the exact reasoning behind the selection of 15 per cent and it is therefore difficult to ascertain whether or not this is a realistic assumption. The work undertaken by Palfrey *et al*. (2001) was also restricted to the assessment of radionuclide behaviour during secondary treatment and does not

include the potential for activity transfer to solids during primary settlement. The results given here are therefore not comparable to SRFs used in the Environment Agency tool.

A1.1.2 Carbon-14

Carbon-14, in a range of different chemical forms, was also assessed in the study by Palfrey *et al*. (2001). Similar results were obtained as for H-3, resulting in a range of six to 22 per cent (average 12 per cent) partitioning to sewage. Ham *et al*. (2003) again assumed, on the basis of results presented by Palfrey *et al*. (2001) that 15 per cent of C-14 would partition to sludge during sewage treatment.

Similar comments to those presented for H-3 above apply to the selection of a partitioning fraction for C-14.

A1.1.3 Iodine-125 and Iodine-131

A detailed review of iodine behaviour during sewage treatment is provided by Punt *et al*. (2007). This is described below.

Erlandsson *et al*. (1989) determined that in a treatment plant in Lune in southern Sweden, the concentration of I-131 in digested sludge was approximately 2,000 times that in treated liquid sewage, but that less than 14 per cent of the activity arriving at the plant left in the sludge. They also noted that the mean residence time for water in the works was one to two days, whilst that of sludge was three to four weeks. Implicit in their assessment is the loss of I-131 through decay, and the value of 14 per cent transfer to sludge should not be used as indicative of the SRF.

At the Oak Ridge sewer, Tennessee, Stetar *et al*. (1993) estimated that within crude sewage, about three per cent of I-131 activity was associated with solids; and that the overall removal efficiency was three to five per cent, with 55 per cent of iodine-131 received at the sewage works being discharged with the treated effluent. Although it is not discussed explicitly within the paper, the results indicate that around 40 per cent of the I-131 may have been retained in the sewer and/or lost through decay.

Barci-Funel *et al*. (1993) noted that sludge contained up to 55 Bq kg^{-1} (dw) I-131 associated with administrations of 0.02 to 7.4 GBq, and estimated that approximately one per cent of that administered was transferred to the final dewatered solids in a sewage treatment plant in Nice, France. Again, this value accounts for loss of activity through decay during sludge treatment.

Dickson (1994) reports a study associated with discharges from the Amersham (now GE Healthcare) White Lion Road Laboratories and associated Maple Lodge sewage treatment works. Samples were collected from the sewage system and treatment works on one day and the analysis included I-125 and I-131. Unfortunately, due to large limits of detection (up to 120,000 Bq l^{-1}) no results for iodine-131 were generated. However, a concentration factor was estimated between raw and digested (and presumably dewatered) sludge of at least six for I-125 (which has a half-life of approximately 60 days).

Dalmasso *et al*. (1997) monitored I-131 in sewage solids from two sewage treatment plants in Nice and St Laurent, which produced 45 and 11 tonnes of dewatered sewage solids on a daily basis respectively. The Nice works received sewage from a hospital where administrations ranged from 0.1 to 10 GBq, with two to three administrations per day. Over a 2.5 month period, the activity concentration in sewage solids varied from three to 320 Bg kg^{-1} (dw). Urine from patients who received a dose greater than 0.4 GBq was stored for seven weeks prior to discharge to sewer, and hence it is difficult to

compare these results to other studies. Nonetheless, the authors estimated that about two per cent of the iodine administered to patients in the hospital was transferred to the final solids. At the St Laurent site, activities in sludge varied from five to 110 Bq kg^{-1} (dw) and peaked one month following a single administration of 0.2 GBq.

Fenner and Martin (1997) also describe the varying behaviour of ionic and organically complexed forms of I-131 discharged to sewer and received at the Ann Arbor treatment works at Michigan. They found that 17 per cent of I-131 in the organically complexed form metaiodobenzyguanidine (mIBG), but only one per cent of NaI, was found in the primary sludge. Fenner and Martin (1997) concluded that the behaviour of different chemical forms of radioiodine was significantly different and attributed this to the greater positive charge of the mIBG form and hence affinity for binding to biosolids. They noted that organic material can accumulate within the sewer system, particularly under low flow conditions, and that mIBG forms of radioiodine may therefore be retained within the sewer system.

Puhakainen (1998) monitored I-131 at a wastewater plant in Finland, which received discharges from a hospital in Helsinki with daily administrations of 3.7 to 7.4 GBq iodine-131. Activity concentrations in the crude sewage typically reached 48 Bq l^{-1} (representing a flux of around five GBq per day) and peaked on the day or the day after administration. Activity concentrations in the final effluent ranged from four to 32 Bq l^{-1} (representing a flux of 0.4 to three GBq per day). Most of the iodine was assessed to have reached the works within three to four days of administration. Overall, 80 per cent of the activity received at the works was estimated to be discharged with the treated effluent. Following an administration of 18.5 GBq to three patients over two days, I-131 activity concentrations in primary sludge (before digestion) reached 8,700 Bq kg^{-1} (dw).

In addition to the Worcester Park results, Titley et al. (2000) provide monitoring data from two other large urban sewage treatment works (the Beckton works in greater London and the Knostrop works in Leeds). At the Beckton site, I-131 activities in crude sewage at the time of sampling were about one Bq l^{-1}, with approximately two-thirds of the activity associated with solids in the sewage. Activity concentrations in the final effluent were less than one Bq l^{-1}. Analysis of primary and secondary sludge indicated that the majority of activity in the sample was associated with solid material. At the Knostrop site, I-131 activities ranged from five to 25 Bq l^{-1} in the primary settlement tanks, eight to 28 Bq l^{-1} in the secondary treatment tanks and three to 31 Bq l^{-1} in the final effluent.

Akinmboni et al. (2005) describe a study to assess the impact of I-131 discharges to sewer in the Ringsend works, which provides waste water treatment for the City of Dublin. Sampling was timed to coincide with I-131 administrations in two Dublin hospitals and activity concentrations of influent, effluent and sewage sludge cake were assessed. The authors found that activity concentrations of I-131 in influent ranged from less than one Bq l^{-1} to 21 Bq l^{-1}, while activity concentrations in effluent ranged from below one Bq l^{-1} to 5.8 Bq l^{-1}. Concentrations in influent samples peaked between 10 and 24 hours following administration and in effluent samples, between 32 and 47 hours following administration. I-131 activity concentrations in dewatered sewage solids ranged from 119 Bq kg^{-1} to 605 Bq kg^{-1}.

Nakamura et al. (2005) present the results of a study to assess I-123 activity concentration in sewage at the Kurume works in Japan, where they found that less than five per cent of the I-123 administered was detected in the final effluent. The lower value compared to that of I-131 may relate to the shorter half-life of I-123 (13.2 hours, approximately 15 times more rapid than that of I-131) and correspondingly greater loss due to decay during transit through the sewage system.

Following a detailed investigation of I-131 behaviour in a sewage works in London, Punt *et al*. (2007) concluded that of the activity received about 40 per cent became associated with sludge while 60 per cent was discharged with treated effluent to rivers. However, the study found that about 90 per cent of the activity initially transferred to sludge during primary and secondary treatment would be lost by decay before final dewatered sewage solids were produced. They also noted that a proportion of activity may be retained in the sewer, potentially associated with larger solids that settle out during low flow conditions. Overall, the authors concluded that of the I-131 activity administered to a patient, about two per cent would remain in final sewage solids after dewatering and about 30 per cent would be discharged to rivers.

Ham *et al* (2003) assumed 20 per cent partitioning to sludge on the basis of their review of the references described above. This is a factor of two lower than that determined by Punt *et al*. (2007).

A1.1.4 Sulphur-35

Little data on the partitioning of S-35 during sewage treatment is available. One study reported a value of eight per cent retention in sludge; however, this was derived on the basis of the amount retained in sludge against an assumed discharge in the liquid effluent. No S-35 could be measured in the liquid effluent and the derived value did not take into account any potential volatisation of sulphur to atmosphere. The chemical form of S-35 was considered important in determining the amount retained in the sludge fraction. However, little information was available to allow further assessment of the importance of chemical form. A default value of 10 per cent partitioning to sludge was therefore assumed by Ham *et al*. (2003), although this was based on limited information, and the conclusions of the review were consistent.

A1.1.4 Sodium-24

Values of seven to 13 per cent sludge retention of Na-24 were identified in the review by Ham *et al*. (2003), based on two studies. In the first, a range of seven to 13 per cent was reported from measurements at a sewage treatment works and, in the second, a value of 10 per cent was obtained from a pilot plant study. The date (ca. 1950) of the first study must be considered and the results are likely to be less applicable to a modern sewage treatment works. The values were considered to be consistent with the assumed behaviour of sodium during sewage treatment, in that alkali metals such as sodium are unlikely to undergo complexation with organic matter and would therefore remain mostly in the liquid phase. This view was supported by expert judgement and a value of 10 per cent retention recommended as the default value by Ham *et al*. (2003).

A1.1.5 Chlorine-36

It was assumed in the review of Ham *et al*. (2003) that chlorine-36 would be present in the form of chloride and would not undergo complexation reactions. No data specific to Cl-36 were available and it was therefore assumed that the majority of Cl-36 would remain in the liquid phase and that 10 per cent would be partitioned to sludge. This value was selected by Ham *et al*. (2003) on the assumption that, since complexation was unlikely to occur, the partitioning would be similar to that of Na-24. However, no data were presented to substantiate this assumption.

A1.1.6 Manganese-54, chromium-51, iron-59, cobalt-57, cobalt-58, selenium-75, rubidium-84, strontium-89 and lead-210

Partitioning data for Mn-54, Cr-51, Fe-59, Co-57, Co-58, Se-75, Rb-84, Sr-89 and Pb-210 were derived by Ham *et al*. (2003) from published measurements for metals from a

pilot plant study (Gans *et al*., 1988). Further data is also provided in a later study by Karvelas *et al*. (2003).

Although default values are given for a wide range of radionuclides by Ham *et al*. (2003), not all are based on a number of published data; some are defined on the basis of a single published data point and therefore, their validity could be questioned. In the case of cobalt, measured data on partitioning to sludge were obtained from four different sources. Three of these values were in reasonably close agreement (31, 50 and 55 per cent retention). However, the authors selected the fourth and most conservative value (80 per cent) as a default for their study. This is in contradiction to the selection of other default partition values, for example H-3 and C-14, where a value within the range of data reported was selected. The value chosen for Mn-54 is comparable to values reported later by Karvelas *et al*. (2003) of above 70 per cent. However, the value of 90 per cent for Fe-59 and Pb-210 is outside of the range (47 to 63 per cent) suggested by Karvelas *et al*. (2003).

A1.1.7 Other radionuclides

Ham *et al.* (2003) investigated the partitioning of P-32, P-33 and Ca-45. In the case of P-32 and P-33, limited data on radionuclide or stable element partitioning was available and the data which was presented was contradictory. Therefore, no default value was recommended in the Ham *et al.* (2003) review. Similarly, data for Ca-45 was limited and contradictory, and was therefore not considered robust enough to recommend a default value for partitioning to sludge.

Data on Cs-134 and Cs-137 were also presented in Ham *et al*. (2003), with partitioning values of 39 per cent and 50 per cent respectively for Cs-134 and Cs-137 determined by Erlandsson *et al.* (1990). However, no default values were given. Karvelas *et al*. (2003) provided data for cadmium, copper, zinc and nickel. In each case, the measured partition factors were in the range of 47 to 63 per cent.

A1.2   Summary of data availability

A significant amount of data is available on radionuclide partitioning within the terrestrial or aquatic environment; however, because of the environmental specificity of Kds, this data requires careful extrapolation to enable its use in determining the partitioning of radionuclides between sludge and effluent during sewage treatment.

Although a number of studies and reviews provide estimated or recommended SRF values, these are often unsupported or unsubstantiated by strong data. For instance, measurements given in the literature do not always clearly indicate where samples have been collected or how they have been prepared or analysed, whether the influence of decay of short-lived radionuclides has been considered and the chemical form of the radionuclide discharged to sewer.

Data summarised by Ham *et al*. (2003) and subsequent studies (Karvelas *et al*., 2003 and Punt *et al*., 2007) are given in Table A1.1.

Table A1.1: Review of SRFs reported previously

Radionuclide (or element)	Ham *et al.* (2003) SRF*	SRF range†	Number of studies considered	Number of data estimates	SRF (Karvelas *et al*., 2003)	SRF (Punt *et al*., 2007)
Be	0.56					
C-14	0.15	0.06-0.22	2	3		
Cd	0.42				0.47-0.63	
Cl-36	0.1		0	0		
Co-57/Co-58	0.8	0.31-0.8	4	3		
Cr-51	0.9	0.43-0.9	4	3	0.47-0.63	
Cs	0.12					
Cu	0.89				>0.7	
Fe-59	0.9		1	1	0.47-0.63	
H-3 (HTO)	0.001		2	3		
H-3 (OBT)	0.15	0.06-0.22	2	3		
I-125/I-131	0.2	0.01-0.23	11	6		0.4+
Mn-54	0.5		1	2	>0.7	
Na-22	0.1	0.07-0.13	2	0		
Ni	0.51				0.47-0.63	
Pb-210	0.9		3	3	0.47-0.63	
Rb-84	0.1		1	1		
Ru-103	0.1	0.03-0.06	1	2		
S-35	0.1	0.08-0.1	1	2		
Se-79	0.8		1	1		
Sr-89	0.1		1	2		
Zn	0.61				0.47-0.63	

* Derived by Ham *et al*. (2003) on the basis of available data.
† Where more than one data point is available.
+Based on fraction transferred to sludge and not considering loss due to decay in subsequent treatment.

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Appendix 2: Nuclide selection processes

Over 80 radionuclides are discharged to sewer in England and Wales (Table A2.1). Limited resources were available to assess all of these, therefore a prioritisation process was used to identify a subset for consideration.

Table A2.1: Radionuclides discharged to sewer in England and Wales

Radionuclide					
Tritium	H-3	Strontium-89	Sr-89	Europium-154	Eu-154
OBT	H-3	Strontium-90	Sr-90	Europium-155	Eu-155
Carbon-14	C-14	Yttrium-90	Y-90	Samarium-153	Sm-153
Fluorine-18	F-18	Niobium-95	Nb-95	Erbium-169	Eb-169
Sodium-22	Na-22	Zirconium-95	Zr-95	Gold-198	Au-198
Sodium-24	Na-24	Technetium-99	Tc-99	Thallium-201	Tl-201
Phosphorus-32	P-32	Technetium-99m	Tc-99m	Lead-210	Pb-210
Phosphorus-33	P-33	Ruthenium-103	Ru-103	Polonium-210	Po-210
Sulphur-35	S-35	Ruthenium-106	Ru-106	Radium-226	Ra-226
Chlorine-36	Cl-36	Indium-111	In-111	Thorium-230	Th-230
Calcium-45	Ca-45	Indium-113m	In-113m	Thorium-232	Th-232
Calcium-47	Ca-47	Iodine-123	I-123	Thorium-234	Th-234
Vanadium-48	V-48	Iodine-125	I-125	Uranium-234	U-234
Chromium-51	Cr-51	Iodine-129	I-129	Uranium-235	U-235
Manganese-52	Mn-52	Iodine-131	I-131	Uranium-238	U-238
Manganese-54	Mn-54	Iodine-132	I-132	Neptunium-237	Np-237
Manganese-56	Mn-56	Iodine-133	I-133	Plutonium-238	Pu-238
Iron-55	Fe-55	Iodine-134	I-134	Plutonium-239	Pu-239
Iron-59	Fe-59	Iodine-135	I-135	Plutonium-240	Pu-240
Cobalt-56	Co-56	Antimony-125	Sb-125	Plutonium-241	Pu-241
Cobalt-57	Co-57	Caesium-134	Cs-134	Plutonium-242	Pu-242
Cobalt-58	Co-58	Caesium-136	Cs-136	Americium-241	Am-241
Cobalt-60	Co-60	Caesium-137	Cs-137	Americium-242	Am-242
Nickel-63	Ni-63	Barium-140	Ba-140	Americium-243	Am-243
Zinc-65	Zn-65	Lanthanum-140	La-140	Curium-242	Cm-242
Gallium-67	Ga-67	Cerium-141	Ce-141	Curium-243	Cm-243
Selenium-75	Se-75	Cerium-144	Ce-144	Curium-244	Cm-244
Bromine-82	Br-82	Promethium-147	Pm-147		
Rubidium-84	Rb-84	Europium-152	Eu-152		

The key factor in selecting radionuclides for assessment is the measure of radiological risk they pose. At a generic level, the probability of exposure is driven by the typical amount of activity discharged by any one site and the number of sites involved. Exposure of sewer workers and members of the public will also depend on the rate of radioactive decay. For instance, for radionuclides that decay rapidly (in the order of minutes to hours), a significant proportion of the activity is likely to be lost before the sewage reaches the plant. An assessment of risk must also account for the radiological dose per unit exposure. Depending upon the radionuclide, this can range by up to four orders of magnitude for ingestion and six orders of magnitude for inhalation.

Any prioritisation process should also take into account the breadth and validity of data, and the practicality and availability of tracer materials and analytical methods.

A2.1 Relative radiological risk

Permit data supplied by the Environment Agency was collated to determine the total possible discharge to sewer across England and Wales, based on the authorisation limits specified. For short-lived radionuclides, this value was decay-corrected to account for the loss of activity during transport through the sewer, to give a possible annual load to sewage works across England and Wales.

Based on this, four categories were determined:

- no specific recorded discharge
- below $1E+07$ Bq yr^{-1}
- from $1E+08$ Bq yr^{-1} to $1E+11$ Bq yr^{-1}
- above $>1E+11$ Bq yr^{-1}.

Despite the high levels of radioactivity discharged to sewer, it is clear that some radionuclides are likely to be more toxic via either internal or external exposure than others. Based on dose per unit ingestion and inhalation, and external exposure factors, the radionuclides discharged to sewer in England and Wales were grouped according to a radiotoxicity ranking of:

- very high
- high
- moderate
- slight.

These two qualitative measures were combined to provide a measure of relative radiological risk, the results of which are presented in Table A2.2.

Table A2.2: Relative radiological risk

Relative risk	Radionuclide
Very high	Y-90
High	Ca-45; Mn-56; Fe-59; Co-60; Sr-89; Sr-90; I-125; I-131; U-238; Th-232.
Moderate	Na-22; Na-24; P-32; P-33; S-35; Cl-36;; Co-57; Co-58; Ni-63; Zn-65; Ga-67; Se-75; Br-82; Tc-99; Cs-137; La-140; Pb-210; Ra-226; Th-234; U-234; Pu-241; Am-241.
Slight	H-3 (OBT & HTO); C-14; F-18; Ca-47; V-48; Cr-51; Mn-52; Mn-54; Fe-55; Co-56; Rb-84; Nb-95; Zr-95; Tc-99m; Ru-103; Ru-106; In-111; In-113m; I-123; Sb-125; I-129; I-132; I-133; I-134; Cs-134; I-135; Cs-136; Ba-140; Ce-141; Ce-144; Pm-147; Eu-152; Eu-155; Sm-153; Eu-154; Er-169; Au-198; TI-201; Po-210; Th-230; U-235; Np-237; Pu-238; Pu-239; Pu-240; Pu-242; Am-242; Am-243; Cm-242; Cm-243; Cm-244.

This assessment was based on permit data; it did not account for radionuclides that might be released to sewer outside of the RSA 93 authorisation process via

inadvertent, accidental or other means or for potential future changes in site discharge schedules.

A2.2 Assessment of existing data

The literature review given in Appendix 1 shows that many radionuclides have limited or no available data to substantiate values used in the modelling tool.

The final distribution of activity between effluent and dewatered solids will vary for different radioisotopes of the same element because of different proportions lost through decay. However, the underlying biochemical behaviour and degree to which these become associated with solids in the sewer or during treatment should be applicable to all radioisotopes of the same element. Where data for one radioisotope is available, we have assumed that it is applicable to all radioisotopes of the same element. The results of this assessment are given in Table A2.3.

Table A2.3: Data available

Existing data	Radionuclide
None	F-18; Cl-36; Ca-45; Ca-47; V-48; Ga-67; Br-82; Y-90; Nb-95; Zr-95; In-111; In-113m; Sb-125; Ba-140; La-140; Ce-141; Ce-144; Pm-147; Eu-152; Sm-153; Eu-155; Eu-154; Er-169; Au-198; Tl-201; Po-210; Ra-226; Th-230; Th-232; Th-234; U-234; U-235; U-238; Np-237; Pu-238; Pu-239; Pu-240; Pu-241; Pu-242; Am-241;Am-242; Am-243; Cm-242; Cm-243; Cm-244.
Limited	Na-22; Na-24; S-35; Cr-51; Mn-52; Mn-54; Mn-56; Fe-55; Fe-59; Ni-63; Zn-65; Se-75; Rb-84; Sr-89; Sr-90; Tc-99; Tc-99m; Ru-103; Ru-106; Cs-134; Cs-136; Cs-137.
Moderate	H-3 (OBT & HTO); C-14; P-32; P-33; Co-56; Co-57; Co-58; Co-60; Pb-210.
Multiple studies	I-123; I-125; I-129; I-132; I-133; I-134; I-131; I-135.

A2.3 Prioritisation process

After discussion with the Environment Agency, a final list was drawn up prioritising elements based on their relative radiological risk and availability of data. The results are given in Table A2.4. Due to the consistency in biochemical behaviour of different radioisotopes of the same element, the results are presented per element and not per radioisotope.

Table A2.4: Final prioritised list

Priority	Radio-Element
Very high	Cl; Y; Sr; Ga; U
High	Th; P; S; Ca; Ni; Br; Tc; La
Medium	Na; Zn; Am; Fe; F; V; Sm; Tl; Pb; Po; Ra; Pu; Cm; In; Co
Low	Se; Ba ;Cs ;Eu; H ;Ce; Mn; C; Cr; Nb; Zr; Sb; Pm
Very low	Rb; Ru; I; Er; Au; Np

A2.4 Final selection for study

In the initial stage of the project, the feasibility of using enhanced stable isotope tracers and analysis by HR ICP-MS was compared to the use of radioisotopes and associated radiometric analysis; this is outlined in more detail in Appendix 4. Enhanced stable isotopes were judged to offer a more practical and cost effective approach in line with Environment Agency guidance to minimise, wherever possible, the use of radioactive substances within laboratory experiments (BPM).

Within the very high priority and high priority categories, all elements could be studied with the exception of chlorine. The naturally high concentrations of chlorine in sewage meant that any non-radioactive tracer would not be distinguishable from the background. Chlorine was therefore excluded from the study. P concentrations and differences in speciation would also make the detection of a tracer difficult. However, naturally occurring P present in the samples could be assessed to determine the solid-solution behaviour.

A multi-element solution containing a mixture with a natural abundance of isotopes for the elements S, Co, Y, Th, U, I, Re and Ga, and isotopically modified abundance for the elements, Ca, Ni, Fe, Sr, In, La and Br, was used to mimic the behaviour of the prioritised radio-elements. It had been planned to use isotopically modified V, but failure to deliver by the supplier meant it was not possible to add a V tracer. Instead, an isotope of V, naturally occurring in the sample, was analysed to determine its behaviour.

The behaviour of naturally occurring Zn was also measured for interference correction and that of Fe and Mn, to understand better the overall chemical process during sewage treatment. In addition, the behaviours of naturally occurring elements in the upper band of the medium priority ranking (F, In and Co) were analysed. Naturally occurring copper was also analysed in the samples; although no discharges of radioisotopes of copper have been identified, it is a known toxic metal and has supporting information. Iodine was included for comparison with literature values.

Appendix 3: Sewage treatment processes

Sewage treatment is a complex procedure involving physical, chemical and biological processes to remove or reduce the following components of crude sewage:

- non-biodegradable solids that may enter the sewers via drains or storm water routes, including debris such as sanitary material and grit from road run-off;

- biodegradable organic rich sewage solids;

- dissolved organic carbon;

- nutrients;

- pathogens.

Sewage treatment processes for a total of 208 plants were reviewed as part of this project. Approximately 70 per cent of works were found to carry out primary treatment and 85 per cent, secondary treatment. The remaining plants included sludge processing plants and sites that failed to supply any data.

Of those for which data were available on secondary treatment, approximately 51 per cent used activated sludge treatment, 29 per cent employed biological filters, and four per cent employed sequential batch reactors. Sixteen percent reported that they used secondary biological processes, but did not classify whether this was by biological filter, activated sludge or some other process. From this data, it can be surmised that activated sludge is the most common secondary treatment process.

The review also revealed that more sludge is produced as a result of primary treatment (63 per cent) compared to secondary treatment. However, there is a large range associated with this (50-80 per cent), partly as a result of the different secondary treatment processes that can be employed. For example, activated sludge plants appear to produce a greater proportion of sludge (50 per cent) compared with biological filters (20 to 40 per cent).

The overall process for a site using primary treatment with activated sludge bed secondary treatment − the most common arrangement in England and Wales − is summarised in Figure A3.1 and briefly described below.

Inlet works

(Pumping, grit screenings
and removal)
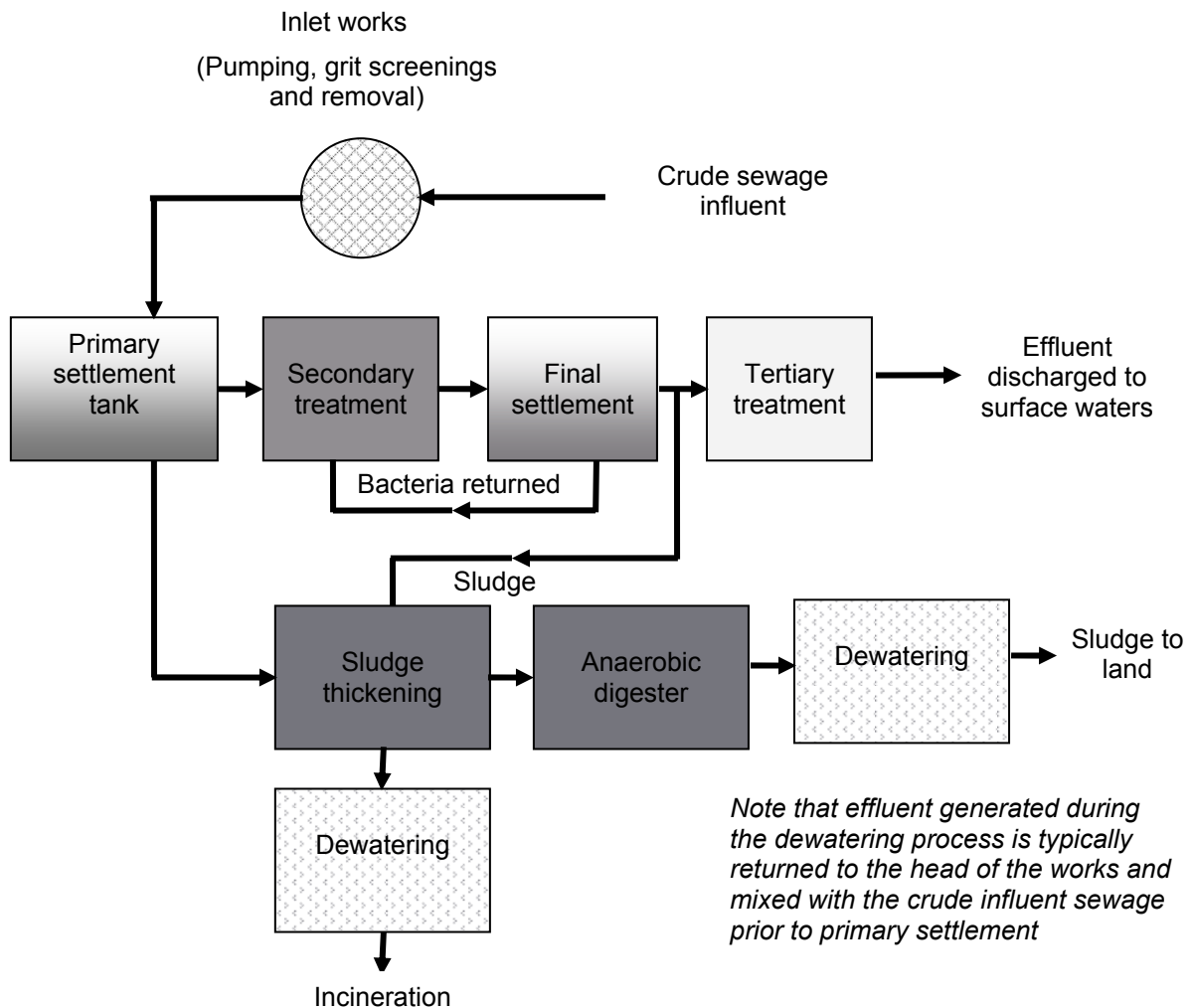
Figure A3.1: Diagram of the sewage treatment process

A3.1 Screening

This process physically screens crude sewage as it enters the head of the sewage works. It removes larger non-biodegradable solids that may have entered the sewer system, along with grit from road run-off. The material collected is normally disposed of to landfill. Any radioactivity discharged to sewer is unlikely to become associated with this material. Therefore, it is not considered further in this assessment.

A3.1 Primary treatment (gravitational settlement)

In this process, crude sewage flows through a series of settlement tanks. Residence times in the tanks are typically two to seven hours depending on the flow rate of crude sewage (primarily dependent upon the rain water input into the sewer system).

As the crude sewage flows through the tanks, up to 50 per cent of solids not removed during the screening process settle out, where they collect at the bottom of the tank and are subsequently pumped out to holding tanks. These solids are termed primary

sludge. The remaining liquid or 'primary effluent', now with a lower suspended solid load, is then transferred to the activated sludge plants.

Although primary treatment is essentially a physical process, the removal of solids to which a proportion of radioactivity may have been adsorbed during transport through the sewer is an important process, influencing how activity may be portioned between solid and dissolved phases.

A3.2 Secondary treatment (activated sludge beds)

Secondary treatment using activated sludge beds is primarily a biological process designed to remove dissolved and particulate carbon from the primary effluent.

Primary effluent passes through the activated sludge beds with a typical residence time of 15 hours. The beds consist of holding tanks which are highly aerated with high (3,000 mg l^{-1}) concentrations of suspended solids, primarily consisting of microbial biomass. Concentrations are maintained by a feedback system that recycles solids from the outflows back into the head of the secondary system. This recycling means the typical residence time of a solid particle in the activated sludge beds is 10 days.

Within the activated sludge beds, the high concentrations of microbial biomass under the highly aerated conditions metabolises dissolved and particulate forms of carbon, converting them into further microbial biomass. To maintain a relatively constant mass of microbial material, some of this material is removed. The secondary sludge is then combined with the primary sludge derived during the initial settlement stage and mixed prior to further treatment.

After it leaves the activated sludge beds, the secondary effluent passes through a series of settlement tanks where further solids settle out. The remaining effluent may then be discharged directly into surface waters or undergo further tertiary treatment. The settled solids are combined with those produced earlier in the treatment process. As with primary settlement, this is an important process in the segregation of activity associated with solids compared to that remaining in the dissolved phase, which is likely to pass rapidly through this stage of treatment.

A3.3 Tertiary effluent treatment

Many sites employ a range of tertiary treatment methods to reduce the nutrient load or the suspended material load prior to discharge of the treated effluent into river or sea. This process generally does not produce any solid material which may act as a pathway for radiological exposure of terrestrial critical groups, although it may reduce the activity concentration of effluent discharged.

A3.4 Anaerobic sludge treatment

After primary and secondary sludge are produced, the material is typically homogenised in holding tanks where some thickening and dewatering may occur (any liquors produced being returned to the head of the plant).

At sites where sewage solids are applied to farmland, the sludge undergoes anaerobic digestion for a minimum period of 12 days. This biological process generates heat which kills off the pathogen load of the material. If the microbial load is not sufficiently reduced during the 12-day period, the sludge may undergo longer periods of anaerobic digestion, or may be stored on site to allow natural die-off prior to dewatering.

This process may lead to loss of activity in the sludge being treated, either via decay during treatment or via the production of methane and carbon dioxide gas (relevant to tritium and carbon-14). It is possible that other radio-elements such as iodine may volatilise during this process, but no data is, to date, available to determine this. This process has therefore not been assessed further in this study.

Where sludge is not applied to land − for example, where it is used for the production of WDF − it will not be treated by anaerobic digestion, but will simply be dewatered.

A3.5 Sludge dewatering

Dewatering typically uses presses to remove liquid, to produce final dewatered solids that may subsequently be applied to land or used as WDF. Any liquors produced during this process are normally returned to the head of the sewage treatment plant.

There is clearly a possibility that some of the activity in sludge which is still in the dissolved phase may be removed during the dewatering phase. Although not specifically addressed in this study, the experimental process used here is designed to account for this eventuality.

Appendix 4: Experimental design

As discussed in the previous chapter, the main processes likely to segregate activity between effluents discharged to surface waters and those retained on solids (which might be applied to land) arise during primary and secondary treatment.

The experimental methods used here aimed to determine:

- whether the use of a chemical spike might affect the biological processes that occur during sewage treatment;

- whether all of the spike introduced could be accounted for, with no significant loss to, or contamination by, experimental equipment;

- what the dissolved-solid partitioning might be during transport through the sewer and primary and secondary sewage treatments.

Addressing these processes and the use of appropriate tracers is discussed below.

A4.1 Choice of experimental tracer

The use of both radio-tracers and stable isotope tracers were considered in this study. Stable isotopes were chosen for the following reasons:

- their use complied with the Environment Agency BPM guidance to, where possible, avoid the use of radioactive substances in laboratory studies;

- they avoided the need for RSA 93 authorisation for the holding and disposal of radioactive materials by the laboratory hosting the experimental systems;

- isotopic abundances of stable isotope tracers could be differentiated from naturally occurring isotopes through the use of HR ICP-MS analysis;

- costs of stable isotope tracers were comparable or cheaper to those of radioisotope equivalents;

- measured concentrations in samples did not need to be corrected for loss of activity due to decay;

- stable tracers were available for the very high and high priority radio-elements identified in Appendix 2.

The use of stable tracers in partitioning studies has in the past been subject to criticism, because concentrations unrepresentative of natural conditions had to be used due to the sensitivity of analytical methods. However, the use of an ultra-sensitive HR ICP-MS detector and isotopically enhanced tracers here circumvented this problem.

As discussed in Appendix 2, stable element isotopic tracers and naturally occurring stable isotopes were used to assess the behaviour of 19 elements. These are described in Table A4.1.

Table A4.1: Isotopic tracers used in this study

Type of tracer	Stable element isotopes
Natural isotopic abundance elements	Co-59; **Ga-69**; I-127; **Re-185 (Tc)**; **S-32**; **Th-232**; **U-238**; **Y-89**
Isotopically modified elements	**Br-79**; Fe-54; In-113; **Ni-58**; **Sr-84**; **La-139**
Isotopes naturally occurring in sample	**P-31**, V-51, Mn-55, Cu-63, Zn-66

Isotopes in bold are the very high and high priority ones identified in Appendix 2.

A4.2 Preliminary experiments

A series of preliminary experiments were conducted to ensure that the introduction of the tracer solution to the sample did not inhibit the biological aspects of sewage treatment, with no excessive loss of tracer to the equipment during sample processing or possible sources of contamination.

A4.2.1 Microbial inhibition

The addition of a cocktail of stable isotopes could inhibit the bacteria responsible for carbon assimilation during secondary treatment. A respiration inhibition test was therefore undertaken. The respiration rate of the bacterial culture was measured using a Strathtox respirometer, which measures the oxygen uptake rate as the bacteria uses the oxygen; if there is an inhibition effect, the respiration rate is decreased. The rate was measured simultaneously in six samples, which had varied amounts of spiking solution added.

The addition of the spiking solution had little or no effect on the respiration rate of the samples. A small effect measured at 10 times the normal spiking concentration was probably more an effect of the change in pH and not the addition of the metal isotopes. It was therefore concluded that the introduction of the spike would not affect the biological processes occurring in sewage treatment.

A4.2.2 Loss of tracer to equipment and potential contamination

These checks were used to evaluate the efficiency and accuracy of the analytical methods used, and also to demonstrate that sewage samples could be filtered without undue losses of determinands by adsorption to the filter media.

A number of samples were spiked and subsequently filtered and analysed. They showed no significant loss to or contamination by experimental equipment.

A4.2.3 Preliminary investigation of tracer partitioning

Crude sewage samples were spiked and allowed to equilibrate for four hours, during which period they were gently stirred. Dissolved and total concentrations were then determined to assess potential uptake to solids during transport through the sewer.

A4.3 Primary treatment

Primary settlement is an initial phase of sewage treatment where particulate material is allowed to settle out under gravity (Appendix 3). This has the effect of removing particulate bound radionuclides from the resultant primary effluent.

To replicate this process, 12 litre samples of crude sewage were spiked with 4 ml l[-1] of the spike solution and allowed to equilibrate for four hours whilst being continuously stirred. Sub-samples (four litres) were then placed into three settlement columns.

A 200 ml sample of supernatant (equivalent to the primary effluent) was taken from each column at time intervals of zero, one, two, five and 24 hours to determine how the dissolved and total concentrations of tracer, and the amount of suspended solids, varied with different settlement periods.

A4.4 Secondary treatment

Aerobic digestion of sludge in activated sludge beds is a biologically mediated process where high concentrations of microbes are responsible for the digestion and assimilation of dissolved and particulate organic material (Appendix 3).

To replicate this biological process, a standard 'Blue Book' system was used. The method is a practical means of simulating the aeration basin of the activated sludge process, which treats more than 60 per cent of all waste water in the UK[1]. This method uses high concentrations (3,000 mg l[-1]) of microbial cultures grown in aerated primary sewage effluent, contained within a permeable membrane which allows secondary treated effluent to drain out while retaining the microbial solids (greater than 20 µm in size). These containers are termed porous pots and are illustrated in Figure A4.1.

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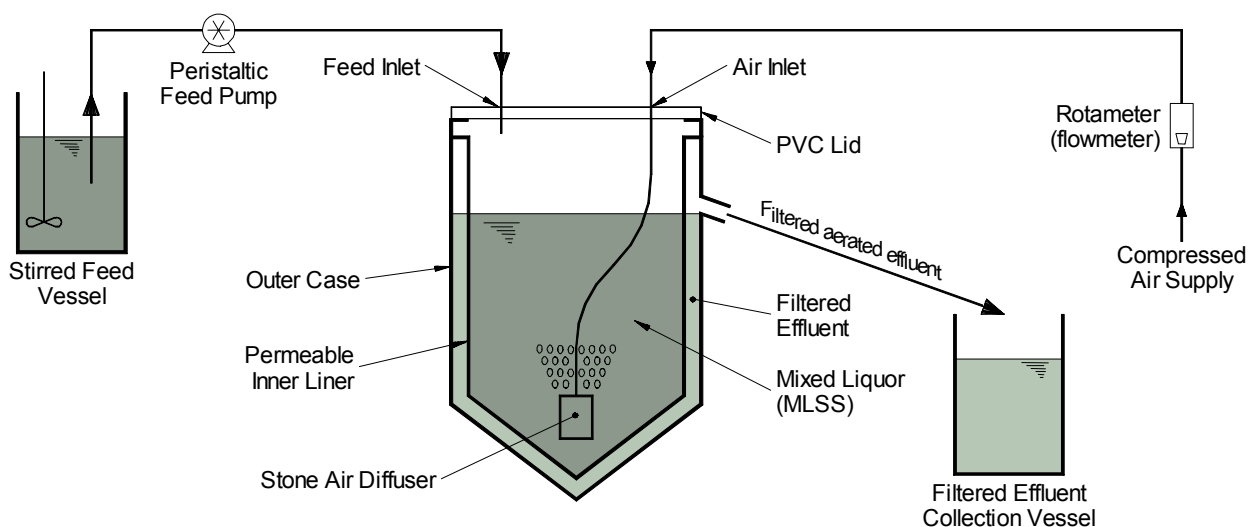
[1] A full description of the porous pot apparatus and methods can be found in the standard "Blue Book" method described in Section E of *Methods for assessing the treatability of chemicals and industrial waste waters and their toxicity to sewage treatment processes* (HMSO, 1982).

Figure A4.1: Diagram of the porous pots

The influent waste water was held in continuously mixed plastic vessels which were refilled on a daily basis. The waste water was fed to the porous pots using a peristaltic pump, controlled using a timer. Treated effluent was collected in separate plastic vessels.

Four porous pots were established by the addition of three litres of activated sludge from a continuously operated pilot scale activated sludge plant fed with domestic sewage. Each pot was provided with an automatic feed of unspiked settled domestic sewage for seven weeks to allow the microbial culture to stabilise. Following this stabilisation process, the feedstock was changed to that which had been spiked. The spiked settled sewage, which was changed daily, was prepared 24 hours before it was fed to the porous pots to allow the tracer to equilibrate with the sewage.

Primary sewage effluent was added to the pots at a rate of 4.7 litres per day by peristaltic pump, to give a hydraulic retention time of 14 hours typical of a standard activated sludge bed. Compressed air and a fine bubble stone diffuser were used to provide aeration and good mixing within the pots.

The final effluent vessels were weighed and emptied daily to determine the flow rate. Each day, 300 ml of the microbial liquor was removed form each porous pot to prevent overflowing due to bacterial mass growth. The influent and mixed liquor of each porous pot was sampled after zero hours, four hours, eight hours, one day, two days, three days, six days, eight days and 10 days to determine the dissolved and total tracer concentrations, and the amount of suspended solids.

Appendix 5: Analytical techniques

Sewage effluent samples were analysed by high resolution inductively coupled plasma mass spectrometry (HR ICP-MS) using a VG Axiom instrument (Thermo Elemental, Winsford, UK). The instrument was tuned at least daily, using a multi-element solution of Mg, Co, In, Pb and U at a concentration of one ng g^{-1}, for optimal sensitivity, stability and peak shape. A mass calibration was also performed daily.

Sewage effluent is a highly complex sample matrix with a high total dissolved solid content. Therefore, samples from the preliminary tests were used to determine the optimal data acquisition parameters, dilution factor and instrumental resolution for each element. A major problem was matrix deposition on the skimmer cone, through which the ion beam passes into the mass spectrometer. This necessitated turning the instrument off, cleaning both the sampler and skimmer cones, retuning and running a fresh set of calibration standards after a maximum of 32 samples. In addition, a calibration standard and a blank were required after every eight samples. It was found that a five-fold dilution of the raw sewage effluent offered the best compromise between minimising matrix effects and keeping the analyte concentration sufficiently high to give a reasonable ion signal for trace elements.

There are a number of polyatomic interferences which are believed to be formed in the interface region of the ICP-MS, arising for the majority of the elements determined. The formation of potential polyatomic interferences, such as $^{16}O_2^+$ on $^{32}S^+$ and $^{35}Cl^{16}O^+$ on $^{51}V^+$, depends partly on the nature of the sample matrix. The resolution necessary to resolve these interferents was determined by analysis of the preliminary test samples.

The elements were also split into two groups for analysis. The first group comprised P, S, Mn, Co, Ga, Sr, Y, Cd, In, Re, Th, and U, whilst the second group was V, Fe, Ni, Cu, Zn, Br, I and La. Niobium and Iridium, which were not detected in the samples, were added to all standards and samples at 10 ng g^{-1} to account for instrumental drift. Multi-element calibration standards were prepared gravimetrically in two per cent HNO_3 from individual elemental BDH Aristar grade (Poole, Dorset, UK) ICP-MS stock solutions, of either 1,000 or 10,000 μg g^{-1} depending on the element. Two per cent HNO_3 blanks were prepared from the same material as the standards. Subsequently, all standards and samples were blank subtracted before further data processing. Linear five-point calibration curves for all analytes were constructed from the blank subtracted, internal standard normalised, integrated counts per second by the least mean squares method. Each calibration curve was graphically plotted for visual inspection. R^2 values were typically 0.999 or better. Experiments were also conducted to determine the effect of the major ion component of sewage effluent on the recovery of trace analytes. A subsample of a calibration standard was spiked with inorganic Na, Mg, S, Cl and Ca at 20, 10, 20, 0.2 and 30 μg g^{-1} respectively. No significant difference in the concentrations of V, Mn, Fe, Co, Ni, Cu, Ga, Br, Sr, Y, In, I, La, Re, Th, and U was observed between the spiked and unspiked calibration standards.

The calcium concentration was determined by flame atomic absorption spectrometry, using a Varian (Yarnton, Oxford, UK) A50. The observed wavelength was 239.9 nm, with a lamp current of 10 mA and a slit width of 0.2 nm. For this analysis, a seven-point calibration curve was obtained, with an R^2 value of greater than 0.997.

Appendix 6: Assessment of sludge retention factors

To account for the different processes within sewage treatment, two components to the sludge retention factor (SRF) were considered: the primary settlement retention factor (PSRF) and aerated sludge retention factor (ASRF).

The PSRF will vary depending upon the settlement time and the ASRF will also vary as tracer concentrations in influent feedstock to the experimental system, the microbial mass in the porous pot and the effluent reach equilibrium.

Primary settlement retention factor values are presented as a function of settlement time, while ASRF values are given based on when the experimental system reached equilibrium. For sewage works that only receive episodic inputs of radioactivity, such as medically used radionuclides that may only be administered to patients one day per week, the system may not be in equilibrium. However, assessing non-equilibrium ASRF values was beyond the scope of this study.

A6.1 Derivation of the sludge retention factors

The PSRF is defined in Equation 1, where Q is the total concentration of the element in the liquid and solid phases and t is time:

$$PSRF = 1 - \frac{[Q]_{t=t}}{[Q]_{t=0}} \qquad \text{(Eq. 1)}$$

If Q is the total amount of any chemical entering the treatment works, then the amount lost to the solid phase during primary settlement, R, is shown in Equation 2, whilst the total amount remaining in the effluent after settlement, E, is shown in Equation 3:

$$R = [Q] \times PSRF \qquad \text{(Eq. 2)}$$

$$E = [Q] \times (1 - PSRF) \qquad \text{(Eq. 3)}$$

The ASRF is defined in Equation 4:

$$ASRF = \frac{[Q]_{total} - [Q]_{disolved}}{[Q]_{total}} \qquad \text{(Eq. 4)}$$

Thus, the amount of Q in the solid phase, S, after aerated sludge treatment is given by Equation 5 whilst the amount of Q in the effluent for discharge, D, is given by Equation 6:

$$S = [Q] \times (1 - PSRF) \times ASRF \qquad \text{(Eq. 5)}$$

$$D = [Q] \times (1 - PSRF) \times (1 - ASRF) \qquad \text{(Eq. 6)}$$

Therefore, the total amount, T, of element Q in the solid phase after both primary settlement and aerated sludge treatment is given by combining Equation 2 and Equation 5 to give Equation 7 and 8:

$$T = ([Q] \times PSRF) + ([Q] \times (1 - PSRF) \times ASRF) \qquad \text{(Eq. 7)}$$

$$T = [Q](PSRF + ASRF - (PSRF \times ASRF)) \qquad \text{(Eq. 8)}$$

The overall sludge retention SRF factor is given by Equation 9:

$$SRF = PSRF + ASRF \times (PSRF \times ASRF) \qquad \text{(Eq. 9)}$$

A6.2 PSRF, ASRF and SRF values

A6.2.1 Preliminary partitioning experiment

Potential partitioning between solid and dissolved phases during transport through the sewer was assessed (Table A6.1).

Table A6.1: Preliminary partitioning experiment

Radio-element	Preliminary partitioning SRF
Br	0.00
Ca	0.04
Co	0.17
Cu	0.79
Fe	0.63
Ga	0.71
I	0.00
In	0.61
La	0.36
Mn	0.50
Ni	0.10
P	0.49
Re (Tc)	0.06
S	0.02
Sr	0.04
Th	0.39
U	0.00
V	0.27
Y	0.84

A6.2.2 Primary settlement retention factors

The sewage influent residence time in the settlement process is typically between two and eight hours. Therefore, the PSRF for each element was taken as the mean value of the individual experimental PSRFs obtained after two and five hours of settlement (Table A6.2).

Table A6.2: Primary settlement retention factors

| Radio-element | PSRF as a function of settlement time | | | | | | | | | |
| | Replicate 1 | | | | | Replicate 2 | | | | |
	0 h	1 h	2 h	5 h	24 h	0 h	1 h	2 h	5 h	24 h
Br	0	0.22	0.26	0.34	0.25	0	0.03	0.28	0.37	0.40
Ca	0	0.02	0.07	0.01	0.07	0	0.08	0.02	0.05	0.03
Co	0	0.11	0.09	0.16	0.13	0	0.26	0.29	0.28	0.28
Cu	0	0.45	0.52	0.50	0.51	0	0.36	0.53	0.51	0.52
Fe	0	0.42	0.61	0.69	0.69	0	0.51	0.63	0.76	1.00
Ga	0	0.38	0.42	0.48	0.54	0	0.37	0.43	0.50	0.61
I	0	0.25	0.28	0.33	0.05	0	0.05	0.25	0.29	0.32
In	0	0.38	0.51	0.65	0.77	0	0.47	0.57	0.66	0.75
La	0	0.35	0.27	0.43	0.30	0	0.31	0.48	0.59	0.57
Mn	0	0.29	0.35	0.44	0.47	0	0.44	0.47	0.46	0.43
Ni	0	0.09	0.05	0.06	0.03	0	0	0	0	0
P	0	0.24	0.26	0.35	0.28	0	0.37	0.44	0.46	0.40
Re	0	0.05	0	0.05	0.05	0	0	0.02	0	0
S	0	0.07	0.00	0.06	0.02	0	0.14	0.20	0.14	0.12
Sr	0	0.09	0.11	0.18	0.28	0	0.14	0.26	0.17	0.15
Th	0	0.19	0.32	0.39	0.44	0	0.09	0.19	0.14	0.20
U	0	0	0	0	0.05	0	0	0.06	0.04	0.38
V	0	0.36	0.46	0.52	0.58	0	0.25	0.39	0.46	0.52
Y	0	0.53	0.78	0.82	0.75	0	0.69	0.72	0.80	0.78

For many elements the settlement period over one hour has little affect on the PSRF, although there is a general, but small increase in the PSRF with settlement time. Only two replicates of the settlement experiment were undertaken so it is not possible to assess the standard deviation. However, across the radionuclides assessed the agreement between the two studies was good with a correlation coefficient in excess of 0.7.

A6.2.3 Secondary sludge retention factors

The ASRF for each element was calculated from the data obtained in porous pot experiments from samples taken at three, six, eight and 10 days (Table A6.3). For each element, the ASRF was calculated for each sampling time, and the mean of these values obtained, giving an ASRF at each sampling time from Porous Pots 2 to 4. Subsequently, the mean of these individual porous pot ASRFs was taken to give the overall ASRF for each element.

Table A6.3: Secondary sludge retention factors

Element	Time (h)	SRFs Pot 1	Pot 2	Pot 3	Mean	SD	Mean +/- SD (72-240 hours)
V	0	0.63	0.86	0.92	0.81	0.15	0.80 +/- 0.05
	4	0.83	0.85	0.87	0.85	0.02	
	8	0.78	0.77	0.95	0.83	0.10	
	24	1.00	0.82	0.82			

Element	Time (h)	SRFs Pot 1	Pot 2	Pot 3	Mean	SD	Mean +/- SD (72-240 hours)
	72	0.87	0.84	0.88	0.86	0.82	
	144	0.77	0.81	0.81	0.80	0.03	
	192	0.78	0.75	0.80	0.78	0.02	
	240	0.80	0.70	0.83	0.77	0.07	
Mn	0	0.92	0.91	0.88	0.90	0.02	
	4	0.96	0.92	0.80	0.89	0.08	
	8	0.93	0.90	0.83	0.89	0.05	
	24	1.00	0.72	0.69	0.71	0.02	
	72	0.88	0.91	0.90	0.90	0.02	
	144	0.96	0.97	0.96	0.96	0.01	
	192	1.00	0.99	1.00	1.00	0.00	
	240	0.95	0.95	0.97	0.96	0.01	0.95 +/- 0.04
Fe	0	0.65	0.86	0.86	0.79	0.12	
	4	0.92	0.85	0.78	0.85	0.07	
	8	0.75	0.60	0.85	0.73	0.12	
	24	1.00	0.76	0.65	0.71	0.08	
	72	0.81	0.78	0.83	0.81	0.03	
	144	0.56	0.81	0.65	0.67	0.13	
	192	0.90	0.76	0.79	0.82	0.07	
	240	0.79	0.77	0.79	0.78	0.01	0.77 +/- 0.09
Co	0	0.99	0.80	0.99	0.93	0.11	
	4	0.80	0.67	0.72	0.73	0.06	
	8	0.74	0.66	0.81	0.74	0.08	
	24	1.00	0.64	0.66	0.65	0.01	
	72	0.91	0.91	0.93	0.92	0.01	
	144	0.89	0.86	0.90	0.88	0.02	
	192	0.91	0.88	0.91	0.90	0.02	
	240	0.92	0.88	0.93	0.91	0.02	0.90 +/- 0.02
Ni	0	0.75	0.91	0.90	0.85	0.09	
	4	0.91	0.89	0.87	0.89	0.02	
	8	0.86	0.73	0.93	0.84	0.10	
	24	1.00	0.82	0.81	0.82	0.01	
	72	0.90	0.82	0.93	0.88	0.05	
	144	0.85	0.82	0.85	0.84	0.01	
	192	0.87	0.80	0.89	0.85	0.04	
	240	0.88	0.86	0.90	0.88	0.02	0.86 +/- 0.04
Cu	0	0.91	0.98	0.98	0.96	0.04	
	4	0.97	0.98	0.98	0.98	0.01	
	8	0.96	0.96	0.99	0.97	0.02	
	24	1.00	0.96	0.97	0.97	0.01	
	72	0.99	0.96	1.00	0.98	0.02	
	144	0.97	0.96	0.99	0.97	0.01	
	192	0.93	0.95	0.99	0.95	0.03	
	240	0.92	0.90	0.94	0.92	0.02	0.96 +/- 0.03
Ga	0	1.00	0.88	1.00	0.96	0.07	
	4	1.00	0.88	1.00	0.96	0.07	
	8	1.00	0.86	1.00	0.95	0.08	
	24	1.00	0.90	1.00	0.95	0.07	
	72	1.00	0.97	1.00	0.99	0.02	
	144	1.00	0.94	0.99	0.98	0.03	
	192	1.00	0.94	0.99	0.98	0.03	
	240	1.00	0.95	0.98	0.98	0.03	0.98 +/- 0.02
In	0	0.00	0.23	0.00	0.08	0.13	0.82 +/- 0.11
	4	0.00	0.48	0.00	0.16	0.28	
	8	1.00	0.48	0.92	0.80	0.28	
	24	1.00	0.60	0.96	0.78	0.26	

Element	Time (h)	SRFs Pot 1	Pot 2	Pot 3	Mean	SD	Mean +/- SD (72-240 hours)
	72	0.96	0.77	0.99	0.91	0.12	
	144	0.84	0.62	0.81	0.76	0.12	
	192	0.81	0.64	0.82	0.76	0.10	
	240	0.69	0.53	0.65	0.63	0.08	
Re (Tc)	0	0.00	0.64	0.87	0.5	0.45	
	4	0.00	0.00	0.00	0.00	0.00	
	8	0.00	0.00	0.00	0.00	0.00	
	24	0.00	0.00	0.00	0.00	0.00	
	72	0.00	0.00	0.00	0.00	0.00	
	144	0.00	0.00	0.00	0.00	0.00	
	192	0.00	0.00	0.00	0.00	0.00	
	240	0.00	0.00	0.00	0.00	0.00	0.00
Ca	0	0.28	0.33	0.33	0.31	0.03	
	4	0.30	0.28	0.35	0.31	0.04	
	8	0.29	0.31	0.35	0.32	0.03	
	24	0.28	0.29	0.34	0.30	0.03	
	72	0.38	0.39	0.39	0.38	0.00	
	144	0.36	0.33	0.38	0.36	0.02	
	192	0.35	0.33	0.34	0.34	0.01	
	240	0.30	0.20	0.29	0.26	0.06	0.34 +/- 0.05
Sr	0	0.21	0.39	0.53	0.38	0.16	
	4	0.51	0.35	0.68	0.51	0.16	
	8	0.46	0.18	0.63	0.42	0.23	
	24	1.00	0.24	0.35	0.30	0.08	
	72	0.43	0.47	0.62	0.51	0.10	
	144	0.33	0.05	0.38	0.25	0.17	
	192	0.29	0.26	0.14	0.23	0.08	
	240	0.13	0.32	0.45	0.30	0.16	0.32 +/- 0.16
Y	0	0.00	0.00	0.00	0.00	0.00	
	4	0.00	0.00	0.00	0.00	0.00	
	8	0.00	0.97	0.86	0.91	0.53	
	24	1.00	1.00	0.99	0.99	0.00	
	72	1.00	1.00	1.00	0.998	0.00	
	144	1.00	1.00	1.00	0.999	0.00	
	192	1.00	1.00	1.00	0.999	0.00	
	240	1.00	1.00	1.00	0.998	0.00	1.00 +/- 0.00
La	0	0.00	0.00	0.70	0.23	0.41	
	4	0.37	0.30	0.28	0.32	0.05	
	8	0.50	0.28	0.60	0.46	0.16	
	24	1.00	0.13	0.01	0.07	0.08	
	72	0.21	0.29	0.22	0.24	0.04	
	144	0.50	0.56	0.01	0.36	0.30	
	192	0.52	0.37	0.26	0.38	0.13	
	240	0.66	0.63	0.36	0.55	0.17	0.38 +/- 0.2
P	0	0.60	0.71	0.74	0.68	0.07	
	4	0.75	0.73	0.73	0.74	0.01	
	8	0.70	0.62	0.74	0.69	0.06	
	24	1.00	0.60	0.60	0.60	0.00	
	72	0.68	0.74	0.77	0.73	0.05	
	144	0.66	0.56	0.62	0.62	0.05	
	192	0.65	0.58	0.66	0.63	0.04	
	240	0.58	0.54	0.59	0.57	0.03	0.64 +/- 0.07
S	0	0.00	0.00	0.00	0.00	0.00	0.00
	4	0.02	0.00	0.00	0.01	0.01	
	8	0.00	0.00	0.14	0.05	0.08	

Element	Time (h)	SRFs Pot 1	Pot 2	Pot 3	Mean	SD	Mean +/- SD (72-240 hours)
	24	ND	0.00	0.00	0.00	0.00	
	72	0.00	0.00	0.00	0.00	0.00	
	144	0.00	0.00	0.00	0.00	0.00	
	192	0.00	0.00	0.00	0.00	0.00	
	240	0.00	0.00	0.00	0.00	0.00	
Br	0	0.00	0.27	0.17	0.15	0.14	
	4	0.04	0.29	0.00	0.11	0.15	
	8	0.17	0.00	0.41	0.19	0.20	
	24	ND	0.00	0.00	0.00	0.00	
	72	0.00	0.00	0.03	0.01	0.02	
	144	0.00	0.00	0.00	0.00	0.00	
	192	0.00	0.00	0.00	0.00	0.00	
	240	0.00	0.00	0.00	0.00	0.00	0.00
I	0	0.43	0.70	0.68	0.61	0.15	
	4	0.73	0.72	0.66	0.71	0.04	
	8	0.68	0.55	0.51	0.58	0.09	
	24	1.00	0.66	0.43	0.54	0.16	
	72	0.60	0.62	0.90	0.71	0.17	
	144	0.62	0.66	0.87	0.72	0.14	
	192	0.65	0.72	0.88	0.75	0.12	
	240	0.72	0.71	0.88	0.77	0.10	0.74 +/- 0.12
Th	0	0.84	0.89	0.89	0.87	0.03	
	4	0.83	0.90	0.86	0.86	0.04	
	8	0.79	0.95	0.87	0.87	0.08	
	24	1.00	0.90	0.92	0.91	0.02	
	72	0.77	0.74	0.75	0.75	0.02	
	144	0.86	0.89	0.85	0.86	0.02	
	192	0.72	0.91	0.87	0.83	0.10	
	240	0.84	0.79	0.67	0.77	0.09	0.80 +/- 0.08
U	0	0.00	0.00	0.00	0.00	0.00	
	4	0.00	0.00	0.00	0.00	0.00	
	8	0.00	0.00	0.00	0.00	0.00	
	24	ND	0.00	0.00	0.00	0.00	
	72	0.00	0.00	0.00	0.00	0.00	
	144	0.00	0.00	0.00	0.00	0.00	
	192	0.00	0.00	0.00	0.00	0.00	
	240	0.00	0.00	0.00	0.00	0.0	0.00

The results show that for many of the elements the relative standard deviation (RSD) was low and the results obtained from the three porous pots over the last four sampling times are highly consistent (RSD < 10%). For some, the results are more variable, RSD 10-20% (e.g. In and Ca), while the results for Sr were highly variable (RSD ~ 50%). This variability arises from differences between the different experimental systems and the times at which samples were collected. This variability is not however fully understood.

The preliminary partitioning experiment, PSRF, ASRF and derived SRFs are compared to those used in the Environment Agency tool in Table A6.4.

A6.2.4 Preliminary partitioning experiment

The preliminary partitioning experiment assessed potential tracer uptake to solids over a four hour period when the sewage was mixed, but not in aerated conditions similar to that which occurs in the sewer.

The experiment found that Br, Ca, I, Ni, Re (Tc), S, Sr and U had little affinity for sewage solids. That of Co and V was low, while others were moderate to high. The highest affinity for sewage solid material was shown by Y.

A6.2.5 Primary sludge retention factor

Mean PSRF values are averaged over the two and five-hour settlement periods. Very little of the Ca, Ni, Re, S and U tracers were lost during settlement. This implies that under the test conditions assessed, the majority of these elements remained in the dissolved phase and were not affected by loss of solids through gravitational settling.

For the majority of other elements, between 20 and 50 per cent of the total concentration in the crude sewage was lost with the settled solids and at real works would be transferred to the primary sludge. This value was higher for In and Fe (60 and 67 per cent respectively) and nearly 80 per cent of the Y was transferred to sludge.

Table A6.4: Comparison of sludge retention factors

Radio-element	Sludge retention factors (SRFs)			Calculated in this study	Reported in the literature	Used by the Environment Agency
	Preliminary partitioning experiment	PSRF	ASRF			
Br	0.00	0.31	0.00	0.01	Not available	0.1[a]
Ca	0.04	0.04	0.34	0.4	Not available	0.1[a]
Co	0.17	0.20	0.90	0.9	0.31-0.8	0.8[b]
Cu	0.79	0.52	0.96	1.0	0.7-0.89	Not included
Fe	0.63	0.67	0.77	0.9	0.9	0.9[b]
Ga	0.71	0.46	0.98	1.0	Not available	0.9[c]
I	0.00	0.29	0.74	0.8	0.01-0.4	0.2[b]
In	0.61	0.60	0.82	0.9	Not available	0.9[c]
La	0.36	0.44	0.38	0.7	Not available	0.1[c]
Mn	0.50	0.43	0.95	1.0	0.5-0.7	0.5[b]
Ni	0.10	0.01	0.86	0.9	0.51	Not included
P	0.49	0.38	0.64	0.8	Not assessed	0.8[d]
Re (Tc)	0.06	0.00	0.00	0.01	Not available	0.1 (Tc)[a]
S	0.02	0.10	0.00	0.01	0.08-0.1	0.1[b]
Sr	0.04	0.18	0.32	0.4	0.1	0.1[b]
Th	0.39	0.26	0.80	0.9	Not available	0.9[a]
U	0.00	0.01	0.00	0.01	Not available	0.1[a]
V	0.27	0.46	0.80	0.9	Not available	0.9[c]
Y	0.84	0.78	1.00	1.0	Not available	0.1[c]

[a] Derived from partition coefficient values for organic soil.
[b] Based on values given in NRPB-W32 derived from Ham *et al*. (2003).
[c] Based on expert opinion.
[d] Cardiff assessment.

A6.2.6 Activated sludge retention factor

Mean ASRF values are averaged over three separate porous pots and the samples collected three, six and 10 days into the experiment.

There was no detectable transfer to solids within the mixed liquor in the porous pots for Br, Re, S and U. The behaviour of Re, S and U was consistent with that observed during the settlement experiment. There was also relatively low (30 to 40 per cent) transfer of Ca, La and Sr to the solids in the porous pots.

For the other elements, 60 per cent or more of the tracer became associated with the microbial solids and at a real works would be transferred to the secondary sludge. This

was particularly true for Cu, Ga, Mn and Y, where the vast majority of the tracer was transferred to the sludge. The behaviour of Y was consistent with that observed during the settlement test, indicating that it has a high affinity for particle association.

A6.2.7 Sludge retention factor

By combining the PSRF and ASRF, an SRF could be derived. This suggests that the combined processes of primary and secondary treatment will result in only small amounts of Br, Re (Tc), S, and U being transferred to sludge. In contrast, over 90 per cent of Co, Cu, Fe, Ga, In, Mn, and Y is anticipated to be transferred to sludge.

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