Journal of Environmental Radioactivity 167 (2017) 254–268

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Improving the quantity, quality and transparency of data used to derive radionuclide transfer parameters for animal products. 2. Cow milk

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ARTICLE INFO

Article history:
Received 26 September 2016
Received in revised form
28 October 2016
Accepted 28 October 2016
Available online 21 December 2016

Keywords:
Cow milk
Transfer parameters
Radionuclides
Animal products
Modelling
Risk assessment

ABSTRACT

Under the International Atomic Energy Agency (IAEA) MODARIA (Modelling and Data for Radiological Impact Assessments) Programme, there has been an initiative to improve the derivation, provenance and transparency of transfer parameter values for radionuclides from feed to animal products that are for human consumption. A description of the revised MODARIA 2016 cow milk dataset is described in this paper. As previously reported for the MODARIA goat milk dataset, quality control has led to the discounting of some references used in IAEA's Technical Report Series (TRS) report 472 (IAEA, 2010). The number of Concentration Ratio (CR) values has been considerably increased by (i) the inclusion of more literature from agricultural studies which particularly enhanced the stable isotope data of both CR and F_m and (ii) by estimating dry matter intake from assumed liveweight. In TRS 472, the data for cow milk were 714 transfer coefficient (F_m) values and 254 CR values describing 31 elements and 26 elements respectively. In the MODARIA 2016 cow milk dataset, F_m and CR values are now reported for 43 elements based upon 825 data values for F_m and 824 for CR. The MODARIA 2016 cow milk dataset F_m values are within an order of magnitude of those reported in TRS 472. Slightly bigger changes are seen in the CR values, but the increase in size of the dataset creates greater confidence in them. Data gaps that still remain are identified for elements with isotopes relevant to radiation protection.

1. Introduction

The transfer of radionuclides from the environment to animal products is quantified for modelling using two transfer parameters published in the most recent international compilations by the International Atomic Energy Agency (IAEA) in its Technical Report Series (TRS) report 472 and Tecdoc 1616 (IAEA, 2009, 2010). The transfer coefficient, defined as the ratio of the fresh weight activity concentration in milk or meat against the daily dietary radionuclide intake at equilibrium. It has been widely adopted as the basis for quantifying transfer to milk (F_m, d l^{-1}) and meat and eggs (F_f, d kg^{-1}) for all radionuclides (Howard et al., 2009a,b). An alternative, the concentration ratio (CR), is defined as the equilibrium ratio between the radionuclide activity concentration in the animal food product (Bq kg^{-1} fresh weight) divided by the average radionuclide activity concentration in the feedstuff ingested (Bq kg^{-1} dry weight). Values for CR were first provided in TRS 472 and are now commonly reported for animal products.

The animal product tables are currently being revised in the IAEA MODARIA (Modelling and Data for Radiological Impact Assessments) programme. An initial motivation for the revision of the cow milk values was that recently improved information could be used for the IAEA's revision of the SRS 19 on Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment (IAEA, 2001).

The revision process is outlined in Howard et al. (2016a) where the first revised MODARIA 2015 goat milk dataset has been described. In this paper, we provide the revised transfer parameters from the MODARIA 2016 cow milk dataset.

2. Revision of the cow milk data set

TRS 472 and Tecdoc 1616 are accepted by the research community as a robust source of information; the underpinning data from these sources were used as a starting point for the revision. Data from new sources were added, but all data items were

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http://dx.doi.org/10.1016/j.jenvrad.2016.10.018

assessed for their appropriateness and some of the original values rejected. The general approach to revision of the cow milk dataset followed that previously described for goat milk in Howard et al. (2016a). A targeted literature review for cow milk was undertaken to incorporate recently reported F_m and CR values. Reference sources providing data which have been added to the dataset are from Green et al. (1995), Gustafson (2000), Herwig et al. (2011), Juniper et al. (2006), Karunakara et al. (2013), Kinkaid et al. (2003), Kume (1989), Phipps et al. (2008), Rosas et al. (1999), Sheppard et al. (2010), Smith et al. (1991), Štrok and Smodiš (2012), Turtiainen et al. (2014), Ujwal et al. (2011) and Vidovic et al. (2003).

2.1. Inclusion of stable element data

Previously Ng et al. (1977) have used "unassociated milk and feed" element or radioisotope concentrations" to derive F_m values, but the source data are difficult to identify and probably utilise single values. Stable element concentrations in milk and various animal feed types are often reported in publications related to agricultural production of livestock compiled by research councils or government departments. We used such "Agricultural review" data to identify stable element concentrations for both milk and feed as well as peer reviewed sources. A single CR and Fm value for an element was only included in the MODARIA dataset when we identified more than 5 independent measurements of stable element concentrations in cow milk. The corresponding value used for the feed included stable element concentrations reported for either fresh pasture grass, forage or mixed herbage. The source publications used were Church (1980), NRC (2001, 2005), MAFF (1990) and Underwood (1977). As the reference sources for the milk element concentrations are from multiple sources they are given as footnotes in Tables 2b, 3b, 5b and 6b.

The maximum tolerable level of an element (or mineral) is defined by NRC (2005) as the dietary level that, when fed for a defined period of time, will not impair animal health or performance. Only data from animals with stable element dietary intakes below maximum tolerable levels have been used to derive transfer parameters (NRC, 2005; EC, 1991).

When using source publications which provide F_m data for a range of different heavy metal concentration in feed (e.g. Kume, 1989) we have only included the control data and treatments which are below the maximum mineral tolerance levels.

2.2. Dry matter intake (DMI)

In the first food chain handbook, TRS 364, a guidance value for daily dry matter intake (DMI) for dairy cows was given of 16.1 kg d^{-1} (range 10–25 kg d^{-1}). However, it was recommended that location specific information should be used where possible. In TRS 472, no guidance was provided, but the associated TECDOC 1616 (IAEA, 2009) quoted 16 kg d^{-1} and this value was used in TRS 472 to derive cow milk transfer coefficient (F_m) values from CR values for some stable elements.

There is a positive linear relationship between the body mass of an animal and the DMI (e.g. Clauss et al., 2013). In the MODARIA 2016 cow milk dataset, where a source publication does not provide information on the daily DMI of the dairy cattle (either directly or providing data that allows it to be estimated) then assumptions have been made to estimate the liveweight of the dairy cows (Table 1). This has then enabled an estimation to be made of the daily DMI. The approach we have used is simple and does not reflect the considerable variation in daily DMI of dairy cattle. However, variation in the daily DMI is unlikely to change derived F_m values by more than a factor of 2–3.

The liveweight and milk yields of cattle has increased over recent decades through improvements in animal breeding (AHDB, 2012). Therefore, where no liveweight, milk yield or daily DMI was given in the source reference the year of publication (or year of experiment) was used to allocate the daily DMI on the basis of Table 1. The data in Table 1 covers six decades over which data were reported and is most relevant for intensive agricultural conditions in developed countries. Where the source publication provided only the liveweight, the milk yield and then the daily DMI was estimated from the year of publication. Similarly, where the source publication provided only the milk yield, the liveweight and then DMI was estimated from the year of publication. To calculate an F_m value from the derived stable CR value, we used an 'average' dairy cow daily DMI value of 21.5 kg.

Details of experimental details for some source Russian language publications were given in Fesenko et al. (2007a) which showed that milk yield was generally much lower than 10 L d^{-1} and liveweight was often <500 kg. Therefore, we have assumed a lower daily DMI for dairy cows of 10 kg d^{-1} for Russian language sources, where the source publication did not include a daily DMI value. The data in Table 1, based on Western Europe agricultural conditions, are probably not relevant for these animals in lower productivity systems.

2.3. New records in the MODARIA 2016 cow milk dataset

The adoption of the above assumptions have enabled the conversion of previously reported F_m values to provide equivalent CR values and has considerably increased the CR data. In TRS 472, CR values were reported for 26 elements, four of which were based on agricultural stable element data (calcium; nickel; phosphorus; and uranium). In the MODARIA 2016 cow milk dataset additional agricultural stable element data (CR and F_m values) are included for cadmium; copper; iron; iodine; potassium; magnesium; manganese; molybdenum; nickel; lead; and zinc.

A major source of revised or new values was a review of Russian literature for milk by Fesenko et al. (2007a). In TRS 472, data from Russian publications in the 1970–1980's were used which had been translated into English by United States Atomic Energy Commission (USAEC). The Fesenko review paper (2007b) provided an improved summary of relevant data from Russian language studies in the USSR and FSU and was used to replace or revise all such studies previously included in the dataset used for TRS 472. Some duplicate data from the USSR was identified and removed from the dataset including 47 F_m values for strontium. The impact was much less for caesium and iodine where three and four values respectively were removed from the dataset.

Herwig et al. (2011) reported a number of stable data for cow milk and feed using ICPMS (Inductively coupled plasma mass spectrometry) measurements. Sheppard also reported cow milk F_m and CR values for 34 elements using ICPMS analysis except for chlorine which was measured by anion chromatography. Data for an element with more than seven measurable values in Sheppard et al. (2010) were included in the dataset namely: gold; barium; cerium; chlorine; cobalt; caesium; copper; iron; gallium; hafnium; magnesium; manganese; molybdenum; sodium; niobium; lead; rubidium; selenium; strontium; thorium; titanium; and zinc. However, iodine and zirconium were then excluded as the reported F_m and CR values differed by 3 orders of magnitude from other values in the dataset. When the number of reported values for an element in Sheppard et al. (2010) was <7 the element was excluded from the main dataset (arsenic; bismuth; calcium; chromium; lanthanum; neodymium; rhenium; tantalum; tellurium; and uranium). However, as transfer parameter values for some of these

Table 1
Estimated daily DMI using live-weight (based on Agriculture and Horticulture Development Board (AHDB, 2012, 2015)).

Publication period	Liveweight (kg)	Estimated daily milk yield (L d^{-1})	DMI as % of liveweight	Estimated daily DMI (kg d^{-1})
1960–1969	500	10.0	3.50	17.5
1970–1979	500	12.5	3.60	17.9
1980–1989	600	15.0	3.40	20.3
1990–1999	600	20.0	3.58	21.5
2000–2009	600	25.0	3.67	21.8
2010 onwards	700	30.0	3.50	24.5

elements have not previously been reported, they are given for some elements as “less than” values later in this paper.

2.4. Data excluded from TRS 472 dataset

Discrepancies and outliers for some data previously included in the dataset used for TRS 472 were examined and source references rechecked. There were too many excluded data to discuss individually but all excluded references for each element are listed in Tables 2–5. Some information on the reasons for exclusion and examples of data removed are given below. During the quality control, data input errors for F_m values for Squire et al. (1958) and Voigt et al. (1994) for iodine and Smith et al. (1991) for cadmium were corrected.

2.4.1. Exclusions

- The CR value given in TRS 472 for uranium was based on a single measured stable uranium milk concentration so it failed the acceptance criteria of n = 5.
- Some data values were duplicated e.g. A calcium value from Comar et al. (1961) and caesium values from Bonka et al. (1988), Piva et al. (1988) and Voigt et al. (1988).
- The only F_m value in the dataset for beryllium, based on Mullen et al. (1972), was a “less than” measurement for milk. Other “less than” data were excluded from Schüttlekopf and Kiefer (1981) (radium); Sharma et al. (1982) (lead) and Voigt et al. (1988) (cobalt and manganese).
- Data were excluded for ^{203}Hg administered directly into the rumen (Crout et al., 2004), and ^{131}I administered into the jugular (Premachandra and Turner, 1961).
- The data from Hansen and Andersson (1994) for caesium used bulked dry milk from factories in Denmark to estimate F_m values.
- Reported values for technetium were not included in TRS 472 for reasons outlined in Howard et al. (2009a). No new information for Tc has been identified to justify inclusion.
- Some F_m values in TRS 472 exceed 5.0E-2 d kg^{-1} which implies that 5% of the daily intake was secreted in each litre of milk. If a dairy cow was producing more than 20 L of milk this value is clearly not possible. Therefore, any such values were re-examined, taking into account the milk production, and excluded where appropriate. The values from Sheppard et al. (2010) for gold; bismuth and rhenium were excluded from the dataset as the F_m values exceeded 5.0E-2 d kg^{-1} (as did the value for iodine which was already excluded). From Hansen and Andersson (1994), a Swedish farm caesium value was excluded as the reported F_m value was 6.8E-2.
- The values for iodine, niobium, and zirconium from Sheppard et al. (2010) were excluded from the dataset even though n = 12 for all 3 elements. Sheppard et al. (2010) suggested that the high iodine value of 4.3E-2 was due to residual iodine in the barns and milking equipment because of iodine teat dips used before the study was carried out. The reason for the discrepancy for zirconium and niobium has not been identified and has been previously highlighted by Johnson et al. (1988). Further data are needed to clarify the extent of transfer to milk of these two elements. The reported values given in this paper as well as those reported by Sheppard et al. (2010) should be carefully considered before application in assessments.
- The value recommended in TRS 472 for plutonium was nearly an order of magnitude higher than that in TRS 364 and was two orders of magnitude higher than that for americium. The plutonium value in TRS 472 was based on then recent experimental work at Chernobyl and a recent critical evaluation (Howard et al., 2007) because there was considerable variation in the few data available. In the MODARIA 2016 cow milk dataset two values were excluded: (i) from Sansom (1964) as the authors state that there was probable faecal contamination and (ii) from Stanley et al. (1974) as one value for a 19 d feeding trial of ^{238}Pu was considered to be preliminary and the other, from a single administration of ^{238}Pu, did not give adequate data to derive a value. Furthermore, there was conflicting information from an associated paper by Daley (1977). A value from Green et al. (1995) was added, but the authors cautioned that the plutonium and americium data was close to the detection limits for grass and milk, so the values should be treated with caution. The discrepancy between the values for Pu and Am in the MODARIA 2016 cow milk dataset remains.

3. Cow milk parameter values in the MODARIA 2016 cow milk dataset

For all elements an arithmetic mean (AM) and standard deviation (ASD) has been calculated. Where $N \geq 3$ a geometric mean (GM) and standard deviation (GSD) has also been calculated for F_m. For CR in TRS 472, only AM was reported, so direct comparisons with the GM values in MODARIA 2016 cow milk data are not possible. The Cs and I F_m values differ slightly from those in TRS 472. The caesium F_m value in TRS 472 should have been 4.7E-3 (not 4.6E-3) because 3 values were incorrectly omitted from the calculation. Similarly, the iodine F_m value in TRS 472 should have been 6.5E-3 (not 5.4E-3) as 5 values were incorrectly omitted.

As many values are available for caesium, iodine, and strontium,

Table 2a
Summary transfer coefficient data (F_m) for caesium (Cs), iodine (I) and strontium (Sr) from TRS 472 and MODARIA 2016 cow milk dataset; GM: geometric mean; GSD: geometric standard deviation AM: arithmetic mean; ASD: arithmetic standard deviation; Min: mimimum; Max: maximum; N: number of records.

Element	TRS 472					MODARIA 2016						
	GM	GSD	Min	Max	N	AM	ASD	GM	GSD	Min	Max	N
Cs	4.6E-3	2.0	6.0E-4	6.8E-2	288	6.7E-3	7.8E-3	4.9E-3	2.1	6.0E-4	5.7E-2	289
I	5.4E-3	2.4	4.0E-4	2.5E-2	104	9.2E-3	8.3E-3	6.0E-3	2.7	4.0E-4	4.4E-2	105
Sr	1.3E-3	1.7	3.4E-4	4.3E-3	154	1.5E-3	8.3E-4	1.3E-3	2.1	1.5E-5	4.3E-3	118

Table 2b
Sources for summary transfer coefficient (F_m) data for caesium (Cs), iodine (I) and strontium (Sr) in MODARIA 2016 cow milk dataset showing relationship to TRS 472.

Element	Authors from TRS 472	Added/Removed
Cs	Assimakopoulos et al. (1994), Averin et al. (2002), Belli et al. (1989, 1993), Beresford et al. (2000a), Bertilsson et al. (1988), Bonka et al. (1988), Bonka (1989), Bradley and Wilkins (1989), Buldakov and Moskalev (1967), Clooth and Aumaan (1990), Daburon et al. (1989), De Meijer et al. (1990), Fabbri et al. (1994), Fulker and Grice (1989), Gastberger et al. (2001), Gattavecchia et al. (1988), Green et al. (1994), Haas et al. (1995), Handl and Pfau (1987, 1988), Hansen and Andersson (1994), Harris (1962), Hawthorne (1967), Hazzard et al. (1969), Heeschen (1987), Heine and Wiechen (1980), Horyna (1990), Ilyin and Moskalev (1957), Issamov et al. (1998), Johnson et al. (1968), Kahn et al. (1965a), Kalmykov and Mikhailov (2001), Karlen et al. (1991, 1995), Kirchner (1989), Korneyev and Sirotkin (1970, 1982), Koster (1989), Kudryavtsev and Sirotkin (1991), Lacourly et al. (1971), Lengemann et al. (1968), Lettner et al. (2007), Mikhailov et al. (1984a), Mitchell et al. (1989), Moiseev et al. (1967), Monte (1990), Musatovová and Vavrova (1991), Nedbaevskaya et al. (1991), Pearce et al. (1989), Pelletier and Voileque (1971), Petersen (1993), Piva et al. (1988, 1989), Potter et al. (1969), Pshikhodsky et al. (2001), Richter and Eder (1986), Sam et al. (1980), Sansom (1966), Sirotkin and Sokolova (1999), Sirotkin and Sarapultsev (1973), Sirotkin (1991), Sirotkin et al. (1970), Sobolev et al. (1992), Sokolova et al. (1999), Spezzano and Giacomelli (1991), Steinwender et al. (1988), Stewart et al. (1965), Sumerling et al. (1984a,b), Unsworth et al. (1989), Van Den Hoek (1980), Van Den Hoek and Kirchmann (1970), Vankerkom et al. (1988), Voigt et al. (1988, 1989a,b, 1996), Voors and Van Weers (1989, 1991), Vreman et al. (1989), Ward et al. (1965, 1966, 1967, 1989), Wegener and Nygard (1964), Wiechen (1972)	Added: Karunakara et al. (2013), Popplewell and Ham (1989), Rosén et al. (2012), Sheppard et al. (2010), Sirotkin et al. (1978b), Sumerling et al. (1984b), Ujwal et al. (2011) Removed: Bonka (1989), De Meijer et al. (1990), Haas et al. (1995), Hansen and Andersson (1994), Nedbaevskaya et al. (1991), Sirotkin (1991), Sirotkin and Sarapultsev (1973), Voigt et al. (1988)
I[a]	Assimakopoulos et al. (1988), Bertilsson et al. (1988), Bonka et al. (1988), Bustad et al. (1963), Clooth and Aumaan (1990), Daburon et al. (1989), De Meijer et al. (1990), Dreicer et al. (1988), Fulker et al. (1995), Garner and Jones (1960), Garner and Sansom (1959), Handl and Pfau (1987, 1989), Hauschild and Aumann (1989), Heeschen (1987), Horyna (1990), Kirchner (1989), Korneyev and Sirotkin (1970, 1982), Lengemann (1964, 1969), Lengemann and Swanson (1957), Lengemann and Comar (1964), Miller et al. (1972), Monte (1990), Potter et al. (1969, 1971a), Premachandra and Turner (1961), Richter and Eder (1986), Robens and Aumann (1988), Sam et al. (1980), Sasser and Hawley (1966), Sirotkin (1991), Sirotkin et al. (1972), Spezzano and Giacomelli (1991), Squire et al. (1958, 1961), Steenberg (1959), Straub and Fooks (1963), Vandecasteele et al. (2000), Vankerkom et al. (1988), Voigt et al. (1988, 1989a,b, 1994), Voigt and Kiefer (2007), Weiss et al. (1974), Wilkins (1989), Wilkins and Stewart (1987), Wilkins et al. (1988)	Added: Agricultural review; Harris (1962), Parache et al. (2011), Petersen (1993) Removed: De Meijer et al. (1990), Korneyev and Sirotkin (1982)
Sr	Annenkov (1961, 1964, 1969), Averin et al. (1992, 2001), Beresford et al. (2000a,b), Buldakov and Moskalev (1968), Burov (1974), Comar and Wasserman (1956), Comar et al. (1961, 1966), Diadiuchin (1973), Fabbri et al. (1994), Garner and Sansom (1959), Garner et al. (1960), Gastberger et al. (2001), Green et al. (1994), Hardy and Rivera (1968), Harris (1962), Heine and Wiechen (1979), Kahn et al. (1965b), Kalmykov and Mikhailov (2001), Korneyev et al. (1973, 1989), Lettner et al. (2007), Mikhailov et al. (1984a), Musatovová and Vavrova (1991), Panchenko et al. (1974), Pelletier and Voileque (1971), Popplewell and Ham (1989), Pshikhodsky et al. (2001), Sirotkin (1973, 1977, 1991), Squire et al. (1958, 1961), Sumerling et al. (1984a,b), Van Den Hoek and Kirchmann (1970), Ward and Johnson (1983)	Added: Annenkov (1967), Averin et al. (2002), Comar et al. (1966), Herwig et al. (2011), Sheppard et al. (2010) Removed: Korneyev et al. (1973), Sirotkin (1977, 1991)

Source references used for stable milk concentrations which were used to derive transfer coefficients (F_m) values from agricultural review data.
[a] Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), NDC (1992), NRC (2001), Underwood (1977).

these transfer parameter values are described separately in Tables 2a and 3a for F_m and CR respectively; associated data sources are presented in Tables 2b and 3b. Further statistical analysis are presented below in Table 4.

The modification of the TRS 472 dataset with the addition and deletion of values for caesium, strontium and iodine has marginally improved the characteristics of the dataset without markedly changing any of the associated statistics. The sample size (N) of F_m has stayed approximately the same, whilst for CR the sample size has more than doubled for all three isotopes (Table 4). Consequently, the difference in values of the means is much smaller for the F_m data (~1–2%) whilst the CR shows slightly larger differences; iodine shows the largest change in mean value of just over 10%. However, the statistical confidence in the values has increased for the CR data whilst they remain similar for the F_m as shown by the coefficients of variation (CV). For roughly the same sample, the CV for F_m (between 10 and 20%) is smaller than that for CR (between 15 and 50%). More CR data would be needed to reduce the CV further.

The values for the 95th percentile, commonly used for a precautionary risk assessment, do not show much variation, indeed some values remain the same. The similarity should be expected as the datasets contain many of the same values. There is no consistent trend in the change, but the CR 95th percentile values have generally increased as a consequence of having larger samples.

The shapes of the distributions have changed marginally, but again with no major distortions in terms of skewness, kurtosis (flatness/steepness) or movement. Across all the data, greater confidence has been gained in the CR values of the MODARIA 2016 cow milk dataset, but those in TRS 472 have not been compromised. Use of the MODARIA 2016 values can be seen as a minor improvement for caesium, strontium and iodine.

The F_m and CR values for other elements are given in Tables 5a and 6a. The relevant reference sources are listed in Tables 5b and 6b which includes information regarding modifications to the

Table 3a
Summary Concentration Ratio data (CR) from for caesium (Cs), iodine (I) and strontium (Sr) from TRS 472 and MODARIA 2016 cow milk dataset; GM: geometric mean; GSD: geometric standard deviation AM: arithmetic mean; ASD: arithmetic standard deviation; Min: mimimum; Max: maximum; N: number of records.

Element	TRS 472					MODARIA 2016						
	CR	SD	Min	Max	N	AM	ASD	GM	GSD	Min	Max	N
Cs	1.1E-1	1.2E-1	3.6E-3	6.9E-1	119	1.1E-1	1.1E-1	8.4E-2	2.1	3.6E-3	9.0E-1	289
I	3.0E-1	2.8E-1	3.0E-3	7.9E-1	44	2.0E-1	2.3E-1	1.1E-1	3.1	3.0E-3	1.1E-1	105
Sr	2.3E-2	2.2E-2	5.0E-3	1.4E-1	43	2.1E-2	1.5E-2	1.7E-2	1.9	5.6E-3	1.4E-1	118

Table 3b
Sources for summary Concentration Ratio (CR) data for caesium (Cs), iodine (I) and strontium (Sr) in MODARIA 2016 showing relationship to TRS 472.

	Authors from TRS 472	Added/Removed
Cs	Authors: Assimakopoulos et al. (1994), Belli et al. (1989, 1993), Beresford et al. (2000a), Bertilsson et al. (1988), Bonka (1989), Bradley and Wilkins (1989), Clooth and Aumaan (1990), Daburon et al. (1989), Fabbri et al. (1994), Fulker and Grice (1989), Gastberger et al. (2001), Gattavecchia et al. (1988), Green et al. (1994), Hazzard et al. (1969), Heeschen (1987), Heine and Wiechen (1980), Horyna (1990), Johnson et al. (1968), Kahn et al. (1965a,b), Koster (1989), Lacourly et al. (1971), Lettner et al. (2007), Mitchell et al. (1989), Musatovová and Vavrova (1991), Piva et al. (1988, 1989), Potter et al. (1969), Sam et al. (1980), Spezzano and Giacomelli (1991), Steinwender et al. (1988), Stewart et al. (1965), Van Den Hoek (1980), Vankerkom et al. (1988), Voigt et al. (1996), Voors and Van Weers (1989, 1991), Vreman et al. (1989), Ward et al. (1989), Wegener and Nygard (1964), Wiechen (1972)	Authors Added: Averin et al. (2002), Buldakov and Moskalev (1968), Handl and Pfau (1987, 1988), Hansen and Andersson (1994), Harris (1962), Hawthorne (1967), Ilyin and Moskalev (1957), Issamov et al. (1998), Kalmykov and Mikhailov (2001), Karlen et al. (1991, 1995), Karunakara et al. (2013), Kirchner (1989), Korneyev and Sirotkin (1982), Korneyev et al. (1989), Kudryavtsev and Sirotkin (1991), Lengemann et al. (1968), Mikhailov et al. (1984a), Moiseev et al. (1967), Monte (1990), Pearce et al. (1989), Pelletier and Voileque (1971), Petersen (1993), Popplewell and Ham (1989), Pshikhodsky et al. (2001), Richter and Eder (1986), Rosén et al. (2012), Sansom (1966), Sheppard et al. (2010), Sirotkin and Sokolova (1999), Sirotkin et al. (1970, 1978b), Sobolev et al. (1992), Sokolova et al. (1999), Sumerling et al. (1984a,b), Ujwal et al. (2011), Unsworth et al. (1989), Van Den Hoek and Kirchmann (1970), Vasiliev et al. (1995), Voigt et al. (1988, 1989a,b), Ward et al. (1965, 1966, 1967)
I[a]	Authors: Bertilsson et al. (1988), Bonka et al. (1988), Clooth and Aumaan (1990), Daburon et al. (1989), Dreicer et al. (1988), Hauschild and Aumann (1989), Heeschen (1987), Horyna (1990), Potter et al. (1969), Robens and Aumann (1988), Sam et al. (1980), Spezzano and Giacomelli (1991), Steenberg (1959), Straub and Fooks (1963), Vankerkom et al. (1988), Voigt and Kiefer (2007), Voigt et al. (1994), Weiss et al. (1974), Wilkins (1989), Wilkins and Stewart (1987)	Authors Added: Agricultural review; Assimakopoulos et al. (1988), Bustad et al. (1963), Fulker et al. (1995), Garner and Sansom (1959), Garner and Jones (1960), Handl and Pfau (1987, 1989), Harris (1962), Kirchner (1989), Korneyev and Sirotkin (1970), Lengemann and Comar (1964), Lengemann and Swanson (1957), Lengemann (1964, 1969), Monte (1990), Parache et al. (2011), Petersen (1993), Potter et al. (1971a), Richter and Eder (1986), Sirotkin (1991), Sirotkin et al. (1972), Spezzano and Giacomelli (1991), Squire et al. (1958, 1961), Vankerkom et al. (1988), Voigt et al. (1988, 1989a,b), Wilkins et al. (1988)
Sr	Authors: Beresford et al. (2000a), Comar et al. (1961), Fabbri et al. (1994), Gastberger et al. (2001), Green et al. (1994), Hardy and Rivera (1968), Heine and Wiechen (1979), Kahn et al. (1965b), Lettner et al. (2007), Musatovová and Vavrova (1991), Popplewell and Ham (1989), Sumerling et al. (1984a), Ward and Johnson (1983)	Added: Annenkov (1961, 1964, 1967, 1969), Averin et al. (1992, 2001, 2002), Beresford et al. (2000b), Buldakov and Moskalev (1968), Burov (1974), Comar and Wasserman (1956), Comar et al. (1966), Diadiuchin (1973), Garner and Sansom (1959), Harris (1962), Herwig et al. (2011), Kalmykov and Mikhailov (2001), Korneyev et al. (1989), Mikhailov et al. (1984a), Panchenko et al. (1974), Pelletier and Voileque (1971), Pshikhodsky et al. (2001), Sheppard et al. (2010), Sirotkin and Sarapultsev (1973), Sirotkin (1973), Squire et al. (1958, 1961), Sumerling et al. (1984b), Van Den Hoek and Kirchmann (1970)

Source references used for stable milk concentrations which were used to derive concentration ratio (CR) values from agricultural review data.
[a] Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), NDC (1992), NRC (2001), Underwood (1977).

Table 4
Characteristics of the statistical distributions of the datasets in MODARIA 2016 cow milk dataset for transfer coefficients (F_m) and concentration ratios (CR) values of caesium (Cs), iodine (I) and strontium (Sr). N – sample size, GM - geometric mean, CV – coefficient of variation (percent), 5th P–5th percentile, 95th P–95th percentile, Skewness-measure of symmetry, Kurtosis – measurement of peakiness or flatness.

		N	GM	CV (%)	5th P	95th P	Skewness	Kurtosis
Cs	F_m	289	4.9E-03	14	1.6E-03	1.8E-02	0.49	1.61
	CR	288	8.5E-02	30	2.9E-02	2.7E-01	−0.02	2.37
I	F_m	105	6.0E-03	19	1.1E-03	2.4E-02	−0.32	−0.26
	CR	105	1.1E-01	53	2.0E-02	7.2E-01	−0.24	0.15
Sr	F_m	118	1.3E-03	10	4.5E-04	2.9E-03	−1.48	5.26
	CR	118	1.7E-02	16	7.2E-03	4.2E-02	−1.24	7.73

Russian language references now used in the MODARA 2016 cow milk dataset based on the revision by Fesenko et al. (2007b).

3.1. F_m

The MODARIA 2016 cow milk dataset of F_m values provides a total of 43 F_m values compared with 31 in TRS 472, with values reported for an additional 13 elements (aluminium; arsenic; chlorine; copper; gallium; hafnium; mercury; potassium; magnesium; nitrogen; rubidium; thorium; and titanium) with one element (beryllium) excluded.

3.2. CR

The MODARIA 2016 cow milk dataset provides CR values for 43 elements compared with the 26 reported in TRS 472. The additional CR values reported are for: aluminium, americium; arsenic; copper; gallium; hafnium; mercury; potassium; magnesium; nitrogen; plutonium; radium; rubidium; ruthenium; thorium; titanium and tungsten. The CR values reported in TRS 472 were based on limited data with one value for some elements taken from agricultural stable element reviews. Using the assumptions described above, the number of data underpinning the CR values has increased from 254 data values to 824. Despite the changes, the resulting CR AM

Table 5a
Summary transfer coefficient data (F_m) for elements, excluding Cs, I and Sr, from TRS 472 and MODARIA 2016 cow milk dataset; GM: geometric mean; GSD: geometric standard deviation AM[a]: arithmetic mean; ASD: arithmetic standard deviation; Min: mimimum; Max: maximum; N: number of records.

TRS 472						MODARIA 2016						
Element	GM	GSD	Min	Max	N	AM	ASD	GM	GSD	Min	Max	N
Al	n/a					3.3E-4						1
Am	4.2E-7				1	1.0E-5	1.7E-5	1.6E-6	12.6	3.0E-7	3.0E-5	3
As	n/a					1.8E-4	1.2E-4	1.4E-4	2.2	4.3E-5	3.5E-4	8
Ba	1.6E-4	2.7	3.8E-5	7.3E-4	15	2.7E-4	2.3E-4	1.8E-4	2.7	3.8E-5	7.3E-4	17
Be	8.3E-7				1							
Ca	1.0E-2	1.7	4.0E-3	2.5E-2	15	1.1E-2	4.7E-3	9.9E-3	1.6	4.0E-3	2.1E-2	15
Cd	1.9E-4	15	1.8E-6	8.4E-3	8	2.2E-3	3.0E-3	2.6E-4	16.2	1.8E-6	7.9E-3	13
C	2.0E-5	5.8	2.0E-6	1.3E-4	6	4.2E-5	4.5E-5	1.5E-5	6.7	1.0E-6	1.3E-4	8
Cl	n/a					2.4E-2	1.7E-2	1.8E-2	2.9	5.4E-3	3.9E-2	3
Co	1.1E-4	2.0	6.0E-5	3.0E-4	4	1.9E-3	3.0E-3	3.2E-4	9.2	2.2E-5	1.0E-2	16
Cr	4.3E-4	26.0	1.0E-5	4.3E-3	3	1.3E-3	1.1E-3	3.4E-4	21.0	1.0E-5	2.1E-3	3
Cu	n/a					3.0E-4	2.3E-4	1.8E-4	3.4	1.8E-5	8.0E-4	16
Fe	3.5E-5	2.0	1.0E-5	9.7E-5	7	8.8E-3	1.4E-4	3.7E-5	3.8	4.9E-6	4.5E-4	13
Ga	n/a					6.5E-3						1
Hf	n/a					3.3E-3						1
Hg	n/a					2.8E-3	4.9E-3	8.3E-4	5.6	1.0E-4	1.2E-2	5
K	n/a					4.4E-3				2.9E-3	5.9E-3	2
Mg	n/a					2.6E-3	9.1E-4	2.6E-3	1.4	1.7E-3	3.5E-3	3
Mn	4.1E-5	4.9	7.0E-6	3.3E-4	4	3.9E-5	8.0E-5	1.3E-5	4.9	5.2E-7	3.3E-4	16
Mo	1.1E-3	2.3	4.3E-4	5.2E-3	7	1.8E-3	1.8E-3	1.2E-3	2.6	2.8E-4	5.9E-3	13
N	n/a					1.2E-2						1
Na	1.3E-2	2.0	5.0E-3	5.0E-2	7	1.9E-2	1.3E-2	1.6E-2	1.7	9.8E-3	5.0E-2	9
Nb	4.1E-7				1	4.1E-7						1
Ni	9.5E-4		6.5E-4	1.3E-3	2	4.7E-3	5.3E-3	2.5E-3	3.8	6.1E-4	1.4E-2	5
P	2.0E-2					7.1E-3	6.7E-4	4.1E-3	3.7	1.1E-3	1.5E-2	4
Pb	1.9E-4	1.0	7.3E-6	1.2E-3	15	4.2E-4	4.2E-4	2.3E-4	3.7	1.1E-5	1.7E-3	22
Po	2.1E-4	1.8	8.9E-5	3.0E-4	4	2.5E-4	8.9E-4	2.4E-4	1.6	1.2E-4	3.0E-4	4
Pu	1.0E-5					1.7E-4	2.8E-4	3.6E-5	9.8	7.5E-6	5.0E-4	3
Ra	3.8E-4	2.3	9.0E-5	1.4E-3	11	6.9E-4	5.2E-4	4.6E-4	2.5	1.1E-4	1.8E-3	15
Rb	n/a					6.5E-3						1
Ru	9.4E-6	8.5	6.7E-7	1.4E-4	6	3.6E-5	5.4E-5	9.4E-6	8.5	6.7E-7	1.4E-4	6
S	7.9E-3				1	8.4E-3				7.9E-3	8.9E-3	2
Sb	3.8E-5	2.5	2.0E-5	1.1E-4	3	5.2E-5	5.1E-5	3.8E-5	2.5	2.0E-5	1.1E-4	3
Se	4.0E-3	2.1	1.5E-3	1.6E-2	12	4.8E-3	3.2E-3	3.8E-3	2.1	6.7E-4	1.3E-2	27
Te	3.4E-4	2.4	7.8E-5	1.0E-3	11	4.2E-4	2.6E-4	3.2E-4	2.3	7.8E-5	1.0E-3	11
Th	n/a					3.7E-3	4.6E-3	1.4E-3	7.8	1.5E-4	8.9E-3	3
Ti	n/a					1.6E-2						1
U	1.8E-3	3.5	5.0E-4	6.1E-3	3	3.1E-3	1.7E-3	2.5E-3	2.2	5.0E-4	6.1E-3	7
W	1.9E-4	3.1	3.4E-5	6.8E-4	7	3.3E-4	2.1E-4	2.6E-4	2.4	5.0E-5	6.8E-4	7
Zn	2.7E-3	3.9	1.3E-4	9.0E-3	8	3.6E-3	1.8E-3	2.8E-3	2.5	1.3E-4	6.9E-3	18
Zr	3.6E-6	4.3	5.5E-7	1.7E-5	6	7.1E-6	6.9E-6	3.6E-6	4.3	5.5E-5	1.7E-5	6

[a] AM only when N = 1 or 2.

values for these elements are within an order of magnitude of those reported in TRS 472, except for cobalt.

3.3. *Other reported data which are not included in the dataset*

In some cases the application of the selection criteria adopted for the MODARIA 2016 cow milk dataset has led to the exclusion of transfer parameter values which may constitute the only values available for some elements. However, such data may be valuable in providing upper limits for the transfer to cow milk so less than values which are the only available data for the element are reported separately in Table 7.

4. Discussion

Preceding papers discussing the values for animal products in TRS 472 (Howard et al., 2009a,b; Fesenko et al., 2007a) have already outlined some of the key factors that influence elemental transfer of radionuclides to cow milk and will not be discussed in detail here, but are briefly summarised below.

Some factors are likely to have a greater effect for some groups of elements. For instance, where homeostatic control is involved (such as calcium, iron, and copper) there might be a disruption of the linear relationship between the concentration in feed and that of milk. Although for many radionuclides the activity concentrations in milk rapidly reach a quasi-equilibrium compared with those in the diet, the assumption of equilibrium between radionuclide activity concentrations in milk and the diet is unlikely to be valid for radionuclides with long biological half-lives such as plutonium, radium, cerium and tungsten (Fesenko et al., 2007b).

Other factors affecting transfer include: (i) the concentration of an element within the animal, as elements previously deposited in animal tissues can be remobilised and transferred to milk; (ii) the interaction of stable elements in the diet (for instance the effect of calcium intake on strontium); (iii) the bioavailability of the dietary source which may be influenced by herbage type, chemical form and soil (or sediment) ingestion; (iv) the differences in daily DMI; (v) the milk yield and stage of lactation; (vi) the age/body weight; (vii) the animal's physiological status and (viii) breed.

The water intake for dairy cows is approximately 70 L d^{-1}. Most of the studies considered here do not provide information on water consumption or the elemental composition of the water consumed. The omission of uptake through drinking water may be more important in the MODARIA 2016 cow milk dataset than TRS 472 due to the inclusion of a larger number of stable element values. Furthermore, analysis of a number of natural radionuclides in vegetation, soil and water near uranium mining and milling facilities in New Mexico showed that the water activity concentrations in control and contaminated areas used for cattle grazing were relatively high compared with feed and may therefore contribute substantially to daily intake (Lapham et al., 1989). This could explain the relatively high transfer values for uranium in Tables 5a and 6a which are based on field measurements. The contribution to intake of water consumed by cows was reported in Herwig et al. (2011). Of the seven elements in the paper which are included in the MODARIA 2016 cow milk dataset, nickel in drinking water contributed 51% and 79% for the two farms studied whereas for the other elements (cobalt; copper; manganese; phosphorus; lead and strontium) the contribution was <11%. In the revision of the goat milk dataset (Howard et al., 2016a) possible nickel cross contamination due to copper-nickel milk vats was a reason for exclusion of some data.

Table 5b
Sources used to generate summary transfer coefficient data (F_m) for elements, excluding Cs, I and Sr, from TRS 472 and MODARIA 2016 cow milk dataset.

Element	Sources from TRS 472	Added/removed
Al		Gustafson (2000)
Am	Sutton et al. (1978)	Green et al. (1995)
As		Crout et al. (2004), Rosas et al. (1999)
Ba	Garner et al. (1960), Harris (1962), Johnson et al. (1988), Potter et al. (1969), Potter et al. (1971a), Sirotkin (1973), Sirotkin and Sarapultsev (1973), Sirotkin et al. (1978b), Sirotkin (1987), Squire et al. (1958), Ward and Johnson (1983)	Sheppard et al. (2010), Sirotkin (1991) Modification to Russian language publications authors: Sirotkin (1973) should be Sirotkin (1973) and Sirotkin and Sarapultsev (1973)
Be	Mullen et al. (1972)	Removed: Mullen et al. (1972)
Ca[a]	Comar and Wasserman (1956), Comar et al. (1961, 1966), Garner et al. (1960), Korneyev et al. (1989), Samson and Garner (1966), Sirotkin (1973), Sirotkin et al. (1978b), Ward and Johnson (1983)	Agricultural review; Gustafson (2000)
Cd[b]	Van Bruwaene et al. (1982a), Crout et al. (2004), Fitzgerald et al. (1985), Nelmes et al. (1974), Sharma et al. (1982), Smith et al. (1991), Vreman et al. (1986)	Agricultural review; Vidovic et al. (2003)
Ce	Sam et al. (1980), Sirotkin et al. (1970, 1978b), Sirotkin and Sarapultsev (1973)	Sheppard et al. (2010), Sirotkin (1991)
Cl		Gustafson (2000), Levchuk et al. (2008), Sheppard et al. (2010)
Co	Van Bruwaene et al. (1984), Sam et al. (1980), Sirotkin et al. (1978b), Sirotkin (1991)	Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010) Modification to Russian language publications authors: Sirotkin (1991) should be Sirotkin (1987)
Cr	Van Bruwaene et al. (1984), Fitzgerald et al. (1985), Ward and Johnson (1983)	
Cu[c]		Agricultural review; Fitzgerald et al. (1985), Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010), Smith et al. (1991), Ward and Johnson (1983)
Fe[d]	Van Bruwaene et al. (1984), Sam et al. (1980), Sirotkin et al. (1978a,b, 1980), Sirotkin (1991), Smith et al. (1991), Ward and Johnson (1983)	Agricultural review; Gustafson (2000), Kume (1989), Sheppard et al. (2010) Modification to Russian language publications authors: Sirotkin (1991) should be Sirotkin (1978)
Ga		Sheppard et al. (2010)
Hf		Sheppard et al. (2010)
Hg		Mullen et al. (1975), Neathery et al. (1974), Vreman et al. (1986)
K[e]		Agricultural review; Gustafson (2000)
Mg[f]		Agricultural review; Gustafson (2000), Sheppard et al. (2010)
Mn[g]	Van Bruwaene et al. (1984), Sam et al. (1980), Voigt et al. (1988), Wilson and Ward (1967), Ward and Johnson (1983)	Agricultural review; Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010)
Mo[h]	Hart et al. (1967), Johnson et al. (1988), Potter et al. (1969), Sirotkin (1991), Ward and Johnson (1983)	Agricultural review; Kume (1989), Sheppard et al. (2010)
N		Gustafson (2000)
Na	Burov and Antakova (1984), De Bortoli et al. (1966), Shilov and Kulakova (1984), Voigt et al. (1988), Ward and Johnson (1983)	Gustafson (2000), Sheppard et al. (2010)
Nb	Johnson et al. (1988)	
Ni[i]	Fitzgerald et al. (1985), Ward and Johnson (1983)	Agricultural review; Herwig et al. (2011)
P[j]	Animal nutrition literature	Gustafson (2000), Herwig et al. (2011)
Pb[k]	Bovay (1971), Djuric and Novak (1971), Fitzgerald et al. (1985), Kerin and Kerin (1971), Mikhailov et al. (1984b, 1990), Nelmes et al. (1974), Potter et al. (1971b), Sharma and Street (1980), Sirotkin (1991), Stanley et al. (1971), Vreman et al. (1986)	Agricultural review; Herwig et al. (2011), Sheppard et al. (2010), Turtiainen et al. (2014) Removed: Bovay (1971), Djuric and Novak (1971), Potter et al. (1971b), Sharma and Street (1980), Stanley et al. (1971)
Po	Johnson and Watters (1972), Moroz and Parfenov (1972), Schüttelkopf and Kiefer (1981), Watters and McIntroy (1969)	Sirotkin (1991) Removed Moroz and Parfenov (1972)
Pu	Howard et al. (2007)	Sirotkin (1991), Green et al. (1995)
Ra	Haas et al. (1995), Kirchman et al. (1972), Samson and Garner (1966), Schüttelkopf and Kiefer (1981)	Štrok and Smodiš (2012), Turtiainen et al. (2014); Removed Haas et al. (1995), Schüttelkopf and Kiefer (1981)
Rb		Sheppard et al. (2010)
Ru	Sirotkin et al. (1970, 1978b), Sirotkin (1973, 1991), Squire et al. (1958)	Modification to Russian language publications authors: Sirotkin (1973) should be Sirotkin and Sarapultsev (1973)
S	Johnson and Watters (1972)	Gustafson (2000)
Sb	Van Bruwaene et al. (1982b), Sam et al. (1980), Sirotkin (1991)	
Se	Aspila (1991), Conrad and Moxon (1979), Johnson and Watters (1972), Perry et al. (1977), Sam et al. (1980)	Gustafson (2000), Juniper et al. (2006), Kume (1989), Phipps et al. (2008), Sheppard et al. (2010)
Te	Handl and Pfau (1987), Johnson et al. (1988), Korneyev and Sirotkin (1970), Mullen and Stanley (1974), Potter et al. (1969, 1971a,b), Squire et al. (1958)	
Th		Sheppard et al. (2010), Štrok and Smodiš (2012)
Ti		Sheppard et al. (2010)
U	Martyushov et al. (1984), Prister (1967), Sirotkin (1987)	Štrok and Smodiš (2012)

Table 5b (*continued*)

Element	Sources from TRS 472	Added/removed
W	Mullen et al. (1976), Potter et al. (1971a), Sirotkin et al. (1978a)	
Zn[l]	Fitzgerald et al. (1985), Kirchgessner et al. (1994), Miller (1969), Miller et al. (1989), Neathery et al. (1973), Sam et al. (1980), Sirotkin (1987), Smith et al. (1991), Ward and Johnson (1983)	Agricultural review; Gustafson (2000), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010), Smith et al. (1991)
Zr	Johnson et al. (1988), Sirotkin et al. (1970, 1978b), Sirotkin (1973, 1991)	

Source references used for stable milk concentrations which were used to derive transfer coefficients (F_m) values from agricultural review data.

[a] ARC (1980), NDC (1992), NRC (2001), Rodriguez et al. (1999).

[b] Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2005), Underwood (1977).

[c] Anderson (1992), Benemariya et al. (1993), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2001), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).

[d] Anderson (1992), Flynn (1992), Hurley (1997), Lopez et al. (1985), NDC (1992), NRC (2001), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).

[e] ARC (1980), Lopez et al. (1985), NDC (1992), NRC (2001), Rodriguez et al. (1999).

[f] ARC (1980), Lopez et al. (1985), NRC (2001, 2005), NDC (1992), Rodriguez et al. (1999).

[g] Anderson (1992), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2001, 2005), Underwood (1977).

[h] Anderson (1992), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2005).

[i] Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985).

[j] ARC (1980), Lopez et al. (1985), NDC (1992), NRC (2001, 2005).

[k] Hurley (1997), Lopez et al. (1985), NRC (2005), Underwood (1977), Zmudzki et al. (1992).

[l] Anderson (1992), ARC (1980), Benemariya et al. (1993), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NDC (1992), NRC (2001, 2005), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).

Table 6a

Summary Concentration Ratio data (CR) for elements, excluding Cs, I and Sr, from TRS 472 and MODARIA 2016 cow milk dataset; GM: geometric mean; GSD: geometric standard deviation AM[a]: arithmetic mean; ASD: arithmetic standard deviation; Min: mimimum; Max: maximum; N: number of records.

Element	TRS 472					MODARIA 2016						
	CR	SD	Min	Max	N	AM	ASD	GM	GSD	Min	Max	N
Al	n/a					5.7E-3						1
Am	n/a					2.1E-4	3.6E-4	7.7E-6	1.4	6.2E-6	6.2E-4	3
As	n/a					3.7E-3	2.6E-3	2.8E-3	2.2	9.0E-4	7.4E-3	8
Ba	1.3E-2	1.6E-3	1.2E-2	1.5E-2	3	7.7E-3	1.5E-2	3.2E-3	3.7	3.8E-4	6.2E-2	17
Ca	2.5E-1					1.7E-1	8.1E-2	1.5E-1	1.7	5.2E-2	3.6E-1	15
Cd	4.3E-2	7.4E-2	2.7E-5	1.3E-1	3	4.5E-2	6.2E-2	5.1E-3	17	2.7E-5	1.6E-1	13
Ce	3.2E-3				1	7.3E-4	1.1E-3	1.9E-4	8.4	1.0E-5	3.2E-3	8
Cl	6.9E-2				1	2.1E-1	2.1E-1	1.5E-1	2.6	7.4E-2	4.5E-1	3
Co	2.5E-3				1	4.7E-2	7.3E-2	6.1E-3	11.3	4.5E-4	2.4E-1	16
Cr	4.0E-2		3.7E-2	4.3E-2	2	2.7E-2	2.3E-2	6.1E-3	25.5	1.5E-2	4.3E-2	3
Cu	n/a					6.1E-3	4.7E-3	3.8E-3	3.2	5.3E-4	1.6E-2	16
Fe	1.2E-3	2.4E-4	1.0E-3	1.5E-3	3	1.8E-3	3.0E-3	5.9E-4	4.4	1.0E-4	9.7E-3	13
Ga	n/a					1.3E-1						1
Hf	n/a					6.6E-2						1
Hg	n/a					5.1E-2	8.8E-2	2.6E-2	5.8	1.8E-3	2.1E-1	5
K	n/a					8.2E-2				6.2E-2	1.0E-1	2
Mg	n/a					5.3E-2	2.1E-2	5.0E-2	1.5	3.5E-2	7.6E-2	3
Mn	4.5E-3		8.6E-4	8.2E-3	2	8.9E-4	2.0E-3	2.7E-4	4.7	1.9E-5	8.2E-3	16
Mo	2.8E-2	1.3E-2	1.9E-2	4.3E-2	3	3.1E-2	2.8E-2	2.2E-2	2.6	3.9E-3	1.0E-1	13
N	n/a					2.0E-1						
Na	3.7E-1		2.3E-1	5.0E-1	2	2.7E-1	1.6E-1	2.3E-1	1.9	9.8E-2	5.0E-1	9
Nb	1.0E-5				1	9.0E-6						1
Ni	8.2E-2					1.4E-1	1.6E-1	6.2E-2	4.7	1.3E-2	4.0E-1	5
P	3.1E-1					1.5E-1	1.3E-1	1.0E-1	2.7	4.1E-2	3.1E-1	4
Pb	2.4E-3	1.3E-3	9.9E-4	4.3E-3	7	9.1E-3	1.0E-2	5.0E-3	3.4	4.1E-4	4.0E-2	22
Po	2.4E-3				1	4.0E-3	1.5E-3	3.8E-3	1.6	2.4E-3	5.4E-3	4
Pu	n/a					1.8E-3	2.8E-3	4.3E-4	9.6	5.8E-5	5.0E-3	3
Ra	n/a					1.3E-2	1.1E-2	8.9E-3	2.6	1.9E-3	4.0E-2	15
Rb	n/a					1.3 E-1						1
Ru	n/a					3.6E-4	5.3E-4	1.0E-4	7.4	1.0E-5	1.4E-3	6
S	1.4E-1				1	1.5E-1				1.4E-1	1.5E-1	2
Sb	2.7E-3				1	1.1E-3	1.4E-3	5.9E-4	3.9	2.0E-4	2.7E-3	3
Se	5.7E-2	4.5E-2	2.6E-2	1.5E-1	7	9.6E-2	6.4E-2	6.3E-2	2.3	1.3E-2	2.3E-1	27
Te	8.0E-3		4.8E-3	1.1E-2	2	7.2E-3	3.2E-3	6.1E-3	2.0	1.4E-3	1.1E-2	11
Th	n/a					3.7E-2	4.5E-2	1.8E-2	5.4	3.1E-3	8.9E-2	3
Ti	n/a					3.2E-1						1
U	5.0E-3					3.1E-2	1.7E-2	2.5E-2	2.2	5.0E-3	6.1E-2	7
W	n/a					5.9E-3	3.9E-3	4.3E-3	2.9	5.0E-4	1.2E-2	7
Zn	7.5E-2	1.6E-2	5.5E-2	9.5E-2	6	6.9E-2	2.6E-2	6.1E-2	1.5	2.9E-2	1.2E-1	18
Zr	1.4E-5				1	7.2E-5	6.8E-5	4.1E-5	3.6	1.0E-5	1.7E-4	6

[a] AM only when N = 1 or 2.

Table 6b
Sources used to generate Concentration Ratio (CR) for elements, excluding Cs, I and Sr, from TRS 472 and MODARIA 2016 cow milk dataset.

Element	Sources from TRS 472	Added/removed
Al		Gustafson (2000)
Am		Sutton et al. (1978), Green et al. (1995)
As		Crout et al. (2004), Rosas et al. (1999)
Ba	Johnson et al. (1988), Potter et al. (1969), Ward and Johnson (1983)	Garner et al. (1960), Harris (1962), Potter et al. (1971a), Sheppard et al. (2010), Sirotkin (1973, 1987, 1991), Sirotkin and Sarapultsev (1973), Squire et al. (1958)
Ca[a]	Stable element review	Agricultural review; Comar and Wasserman (1956), Comar et al. (1961, 1966), Garner et al. (1960), Gustafson (2000), Korneyev et al. (1989), Samson and Garner (1966), Sirotkin (1973), Sirotkin et al. (1978b), Ward and Johnson (1983)
Cd[b]	Crout et al. (2004), Fitzgerald et al. (1985), Smith et al. (1991)	Agricultural review; Van Bruwaene et al. (1982a), Nelmes et al. (1974), Sharma et al. (1982), Vidovic et al. (2003), Vreman et al. (1986)
Ce	Sam et al. (1980)	Sirotkin et al. (1970, 1978b), Sirotkin (1973, 1991), Sirotkin and Sarapultsev (1973), Sheppard et al. (2010)
Cl	Levchuk et al. (2008)	Gustafson (2000), Sheppard et al. (2010)
Co	Sam et al. (1980)	Van Bruwaene et al. (1984), Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010), Sirotkin (1987), Sirotkin et al. (1978a)
Cr	Van Bruwaene et al. (1984), Fitzgerald et al. (1985)	Ward and Johnson (1983)
Cu[c]		Agricultural review; Fitzgerald et al. (1985), Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010), Smith et al. (1991), Ward and Johnson (1983)
Fe[d]	Sam et al. (1980), Smith et al. (1991), Ward and Johnson (1983)	Agricultural review; Van Bruwaene et al. (1984), Gustafson (2000), Kume (1989), Sheppard et al. (2010), Sirotkin et al. (1978a,b, 1980)
Ga		Sheppard et al. (2010)
Hf		Sheppard et al. (2010)
Hg		Mullen et al. (1975), Neathery et al. (1974), Vreman et al. (1986)
K[e]		Agricultural review; Gustafson (2000)
Mg[f]		Agricultural review; Gustafson (2000), Sheppard et al. (2010)
Mn[g]	Sam et al. (1980), Ward and Johnson (1983)	Agricultural review; Van Bruwaene et al. (1984), Gustafson (2000), Herwig et al. (2011), Kinkaid et al. (2003), Kume (1989), Sheppard et al. (2010), Wilson and Ward (1967)
Mo[h]	Johnson et al. (1988), Potter et al. (1969), Ward and Johnson (1983)	Agricultural review; Hart et al. (1967), Kume (1989), Sheppard et al. (2010), Sirotkin (1991)
N		Gustafson (2000)
Na	De Bortoli et al. (1966), Ward and Johnson (1983)	Burov and Antakova (1984), Gustafson (2000), Sheppard et al. (2010), Shilov and Kulakova (1984), Voigt et al. (1988)
Nb	Johnson et al. (1988)	
Ni[i]	Stable element review	Agricultural review; Fitzgerald et al. (1985), Herwig et al. (2011), Ward and Johnson (1983)
P[j]	Stable element review	Gustafson (2000), Herwig et al. (2011)
Pb[k]	Bovay (1971), Djuric and Novak (1971), Fitzgerald et al. (1985), Kerin and Kerin (1971), Sharma and Street (1980)	Agricultural review; Herwig et al. (2011), Mikhailov et al. (1984b, 1990), Nelmes et al. (1974), Sheppard et al. (2010), Shilov and Kulakova (1984), Sirotkin (1991), Turtiainen et al. (2014), Vreman et al. (1986) Data removed Bovay (1971), Djuric and Novak (1971), Sharma and Street (1980)
Po	Schüttelkopf and Kiefer (1981)	Johnson and Watters (1972), Sirotkin (1991), Watters and McIntroy (1969)
Pu		Green et al. (1995), Howard et al. (2007), Sirotkin (1991)
Ra		Kirchman et al. (1972), Samson and Garner (1966), Štrok and Smodiš (2012), Turtiainen et al. (2014)
Rb		Sheppard et al. (2010)
Ru		Sirotkin and Sarapultsev (1973), Sirotkin (1991), Sirotkin et al. (1970, 1978b), Squire et al. (1958)
S	Langlands and Sutherland (1973)	Gustafson (2000), Johnson and Watters (1972) Data removed Langlands and Sutherland (1973)
Sb	Sam et al. (1980)	Van Bruwaene et al. (1982b), Sirotkin (1991)
Se	Aspila (1991), Conrad and Moxon (1979), Sam et al. (1980)	Gustafson (2000), Johnson and Watters (1972), Juniper et al. (2006), Kume (1989), Perry et al. (1977), Phipps et al. (2008), Sheppard et al. (2010)
Te	Johnson et al. (1988), Potter et al. (1969)	Handl and Pfau (1987), Korneyev and Sirotkin (1970), Mullen and Stanley (1974), Potter et al. (1971a), Squire et al. (1958)
Th		Sheppard et al. (2010), Štrok and Smodiš (2012)
Ti		Sheppard et al. (2010)
U[l]	Stable element review	Martyushov et al. (1984), Prister (1967), Sirotkin (1987), Štrok and Smodiš (2012)
W		Mullen et al. (1976), Potter et al. (1971a), Sirotkin et al. (1978b)
Zn[m]	Fitzgerald et al. (1985), Miller (1969), Miller et al. (1989), Sam et al. (1980), Smith et al. (1991), Ward and Johnson (1983)	Agricultural review; Gustafson (2000), Kinkaid et al. (2003), Kirchgessner et al. (1994), Kume (1989), Neathery et al. (1973), Sheppard et al. (2010), Sirotkin (1987)
Zr	Johnson et al. (1988)	Sirotkin and Sarapultsev (1973), Sirotkin (1991), Sirotkin et al. (1970, 1978b)

Source references used for stable milk concentrations which were used to derive CR values from agricultural review data.
[a] ARC (1980). NDC (1992). NRC (2001). Rodriguez et al. (1999).
[b] Flynn (1992). Hurley (1997). Lopez et al. (1985). NRC (2005). Underwood (1977).
[c] Anderson (1992). Benemariya et al. (1993). Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2001), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).
[d] Anderson (1992), Flynn (1992), Hurley (1997), Lopez et al. (1985), NDC (1992), NRC (2001), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).
[e] ARC (1980), Lopez et al. (1985), NDC (1992), NRC (2001), Rodriguez et al. (1999).
[f] ARC (1980), Lopez et al. (1985), NRC (2001, 2005), NDC (1992), Rodriguez et al. (1999).
[g] Anderson (1992), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2001, 2005), Underwood (1977).
[h] Anderson (1992), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NRC (2005).
[i] Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985).
[j] ARC (1980), Lopez et al. (1985), NDC (1992), NRC (2001, 2005).
[k] Hurley (1997), Lopez et al. (1985), NRC (2005), Underwood (1977), Zmudzki et al. (1992).
[l] NRC (2001).
[m] Anderson (1992), ARC (1980), Benemariya et al. (1993), Dobrzanski et al. (2005a,b), Flynn (1992), Hurley (1997), Lopez et al. (1985), NDC (1992), NRC (2001, 2005), Rodriguez et al. (1999), Underwood (1977), Zmudzki et al. (1992).

4.1. The benefit of the inclusion of more stable data to derive animal transfer parameter values

ICPMS analyses are now being used to provide CR values for a large number of elements from the same samples (e.g. Sheppard et al., 2010; Herwig et al., 2011). One concern is whether the use of data for stable isotopes is appropriate for studies of radioisotopes which are from anthropogenic and naturally occurring radiosources.

In Table 8, the relative importance of radioisotope and stable data are given for each element. When there is an element with n = 1, and the derived F_m is based upon stable data, the source publication is Sheppard et al. (2010). For most elements the data is largely dominated by either radioisotope or stable data. Data from stable element analysis may be more appropriate for assessment of long-term contamination (Sheppard et al., 2010).

For 13 elements, all the derived F_m values are solely based on stable isotope data. The transfer parameter values for 14 elements (including caesium, iodine and strontium) include less than 10% stable element data. Those for 12 elements have more than 50% of the F_m and CR values based on stable isotope data. Four elements included in TRS 472 (calcium; nickel; phosphorus; uranium) were based solely on stable element review data, whereas the CR value is now based on both stable element and radioisotope data.

4.2. Elements with the greater range in values

In Table 9, the 95th percentile are shown for those elements with at least 10 values as these are of particular interest for assessments adopting a precautionary principle; the CV % is also presented. The data values for some elements are briefly discussed below.

The transfer parameter values for cadmium and cobalt are the most variable for both F_m and CR. For F_m, the other elements are in a reasonable range of 10–20%. With the sole exception of zinc, the CV % for CR are higher (as they are for casium, strontium and iodine (Table 4)), particularly those for selenium, calcium and molybdenum.

Most of the derived F_m and CR values included in the dataset for cadmium are based on stable data (Table 8). The mammary gland limits cadmium transport so the concentration of cadmium in milk is not increased by high dietary concentrations of cadmium (Sharma et al., 1979; Smith 1986; Van Bruwaene et al., 1982a; NRC, 2001). Many other factors are reported to influence cadmium absorption such as solubility of the chemical form ingested (NRC, 2001) and high dietary concentrations of calcium, chromium, magnesium and zirconium which can reduce transfer to milk (NRC, 2005).

Ruminants have a dietary requirement for cobalt, but most animal feedstuffs are low in cobalt so it is generally supplemented in

Table 7
Maximum Cow milk transfer coefficient (F_m) and concentration ratio (CR) values based on less than values.

Element	GM F_m	GM CR	N	Reference
Be	<8.3E-7	<1.5E-5	4	Mullen et al. (1972)
Bi	<3.4E-1	<6.8E0	2	Sheppard et al. (2010)
La	<5.0E-5	<1.0E-3	4	Sheppard et al. (2010)
Nd	<3.1E-5	<6.3E-4	4	Sheppard et al. (2010)
Re	<7.2E-3	<1.5E-1	6	Sheppard et al. (2010)
Ta	<4.9E-4	<9.8E-4	1	Sheppard et al. (2010)
Tl	<4.4E-4	<9.2E-3	5	Sheppard et al. (2010)

Table 9
Compilation of 95 percentiles (P) and CV – coefficient of variation (percent) values for elements (other than casium, strontiumor iodine) where N ≥ 10 in the MODARIA 2016 cow milk dataset for transfer coefficient (F_m) and concentration ratio (CR) values.

Element	N	F_m		CR	
		95th P	CV%	95th P	CV%
Ba	17	6.4E-4	11	2.4E-2	23
Ca	15	1.9E-2	10	2.9E-1	27
Cd	13	7.2E-3	33	1.5E-1	56
Co	16	1.1E-2	30	1.7E-1	47
Cu	16	6.5E-4	12	1.3E-2	21
Fe	13	3.9E-4	13	8.1E-3	20
Mn	16	1.1E-4	13	2.5E-3	19
Mo	13	5.5E-3	14	8.0E-2	25
Pb	22	1.2E-3	14	3.5E-2	23
Ra	15	1.5E-3	12	3.3E-2	20
Se	27	1.1E-2	14	2.2E-1	32
Te	11	8.0E-4	10	1.0E-2	13
Zn	18	6.1E-3	15	1.1E-1	14

Table 8
The relative contribution of radioisotope and stable data for individual elements in the MODARIA 2016 cow milk dataset.

Element	Radioisotopes (n)	Stable (n)	Stable (%)	Element	Radioisotopes (n)	Stable (n)	Stable (%)
Al	0	1	100	Na	6	3	33
Am	3	0	0	Nb	0	1	100
As	1	8	89	Ni	0	5	100
Ba	15	2	12	P	0	4	100
Ca	12	3	20	Pb	10	12	55
Cd	2	11	85	Po	4	0	0
Ce	7	1	13	Pu	3	0	0
Cl	0	3	100	Ra	15	0	0
Co	4	12	75	Rb	0	1	100
Cr	1	3	75	Ru	6	0	0
Cs	283	6	2	S	1	1	50
Cu	0	16	100	Sb	3	0	0
Fe	5	8	62	Se	2	25	93
Ga	0	1	100	Sr	113	5	4
Hf	0	1	100	Tc	2	0	0
Hg	2	3	60	Te	11	0	0
I	94	11	10	Th	2	1	33
K	0	2	100	Ti	0	1	100
Mg	0	3	100	U	7	0	0
Mn	4	12	75	W	7	0	0
Mo	4	9	69	Zn	4	14	78
N	0	1	100	Zr	6	0	0

their diets (NRC, 2005). The level of cobalt supplementation is not specified in some reference sources, which is a also a source of potential underestimation of stable element intake for other elements.

5. Summary

Despite the increased number of cow milk transfer parameter values in the MODARIA 2016 cow milk dataset, data gaps remain for elements with isotopes relevant to radiation protection, notably for natural radionuclides, and for plutonium and americium, and zirconium/niobium for which there are large discrepancies in the available data values.

When a CR value is required which is not included in the revised tables for cow milk, a value could be used for another milk products if available as there is evidence that there is no difference in CR between species (Howard et al., 2009b). Also, extrapolation methods could be used such as those outlined in Howard et al. (2016b). The use of removed or excluded data discussed above could be considered after further analysis of the possible validity of the reported value for the particular element.

This paper provides the second set of revised and improved parameter values for milk. Revision of the animal product tables will continue under MODARIA II where possible. Other mechanisms to revise and report the tables are being explored and further work will continue to establish suitable metadata and online reporting approaches for such datasets.

Acknowledgements

Much of the work described here was conducted within the EC funded STAR Network of Excellence (Fission-2010-3.5.1-269672) (http://www.starradioecology.org) and COMET framework (Fission-2012-3.4.1-604794) (http://www.comet-radioecology.org). It was also supported by the Centre for Ecology and Hydrology which is part of the Natural Environment Research Council of the UK. We would like to thank all members of the IAEA MODARIA Working Group 4 (http://www-ns.iaea.org/projects/modaria/), Steve Sheppard and Jochen Vogl who contributed to this work with helpful advice and information. We also thank Christine Balmer for compiling the enormous reference list.

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