A Review of Transfer Parameters of I, Cs and Pu (1) -Feed Transfer Coefficients in Cow's Milk-

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Abstract. Internal dose from the ingestion of contaminated agricultural products is an important part of the biospheric radiation dose assessment for nuclear waste management. Cow's milk is consumed by infants, and they are relatively more sensitive to radiation than adults; thus we focused on the radionuclide transfer coefficient from feed to milk (C_{ft_milk}). Many countries use C_{ft_milk} values compiled in the IAEA Technical Report Series in mathematical models. Most of the original data sources for the values were from literature published in the 1950's to 1970's and they are getting difficult to access nowadays, or they were reported after the Chernobyl accident in Russian and Ukrainian languages and they are not accessible to a wide audience. It is necessary to confirm the original data for transparency of the dose assessment results and also to update the data sets by adding newly observed data. In this paper, therefore, we carried out a literature survey by tracing back references listed in the IAEA reports especially for Cs, I and Pu, and we compared those data with recently published data obtained by research institutes in many countries. Our comparison results showed that C_{ft_milk} values of I and Cs were similar among the data sources; however, Pu values differed by four orders of magnitude among the sources, which could cause large uncertainties in assessment results.

KEYWORDS: transfer coefficient; feed-to-milk transfer; environmental dose assessment

1 INTRODUCTION

To assess environmental radiation dose from artificial radionuclides released from nuclear waste disposal sites, internal dose by ingestion of contaminated agricultural products plays an important role. It has been reported that radioiodine in cow's milk was the biggest contributor to internal dose in the early stages of the Chernobyl nuclear accident [1-3]. Cow's milk is a major item in the diet of infants and young children; it is well known that their thyroids are relatively more sensitive to radioiodine exposure compared to adults [4, 5]. As well, not only radioiodine, but also other radionuclides in milk need to be considered because infants and young children are more sensitive to all types of radiation emitted by the radionuclides compared to adults [6]. Therefore, one of the important environmental transfer parameters for discharged radionuclides is the feed transfer coefficient to milk, C_{ft_milk} (d/L), which is defined as follows [7, 8].

$$C_{ft_milk} = A_{milk} / A_{feed.}$$

Here A_{milk} is activity concentration in milk (Bq/L) and A_{feed} is the daily intake of radionuclide (Bq/d).

Many countries use C_{ft_milk} values compiled in the IAEA parameter reports [7, 8] for their dose assessment mathematical models. Most of the original data sources for the values were from literature published in the 1950's to 1970's and they are getting difficult to access nowadays, or they were reported after the Chernobyl accident in Russian and Ukrainian languages and they are not accessible to a wide audience. To keep the transparency of the dose assessment results, it is necessary to clarify the accuracy of the C_{ft_milk} values by checking the original data sources. It is also important to compare these values with recently obtained C_{ft_milk} values to make sure that the old values are equally useful still now.

In this paper, therefore, we carried out a literature survey by tracing back sources listed in the IAEA TRS 364 [7] and SRS 19 [9] for iodine (I), caesium (Cs) and plutonium (Pu), and we compared those C_{ft_milk} values with recently published data obtained by research institutes around the world. We also used the most recent IAEA publication of parameter values [8] for comparison.

2 MATERIALS AND METHODS

Recommended C_{ft_milk} values in IAEA TRS 364 [7] and SRS 19 [9] were mostly collated from review papers and reports. Based on references used in these IAEA reports, we checked further to see whether we could find the original data sets used to calculate C_{ft_milk} of I, Cs and Pu. If we could access the original data sets, we stored those values in MS Excel sheets. Then, for comparison with these values, we also checked summarized reports in Japanese prepared by the Atomic Energy Society of Japan (AESJ) [10] and by the Japan Nuclear Cycle Development Institute and the Federation of Electric Power Companies (JNC & FEPC) [11] for radiation dose assessment in waste management because these reports included C_{ft_milk} values and their sources. The sources of the C_{ft_milk} values in these summarized Japanese reports were various publications and included Japanese literature values.

For the new data survey, we used Google scholar and J-stage (the largest scientific publication search engine in Japan) to collect papers and laboratory reports. The selected key words for the literature survey were, "Transfer coefficient or Concentration ratio", "Milk", and "Animal or Cow", and we searched the keywords in both English and Japanese.

3 RESULTS AND DISCUSSION

3.1 Survey of literature values used in IAEA TRS 364 and SRS 19

We classified references into four categories as follows.

[C1] Original experimental data or C_{ft_milk} data reported with original data sources (tracer or global fallout)

[C2] C_{ft_milk} data calculated by metabolic models of farm animals

[C3] C_{ft_milk} data reported without sources (data not identified)

[C4] Summarized C_{ft_milk} data (review results)

The references used in IAEA TRS 364 and SRS 19 for C_{ft_milk} values of I, Cs and Pu were from categories C2 and C4, respectively. For C4, we further checked the sources of the data. Table 1 shows the reference code we used.

The results for Cs are shown in Figure 1. After following the above-described steps, we could identify two original data sources: one was Coughtrey [12] (category C1), and the other was Ng et al. [13] (category C2). The latter was used more frequently and was an institutional report prepared by Ng et al. for the U.S. DOE. This report cited experimental C_{ft_milk} data available at that time, and they were mostly obtained in the 1950's and 1960's with some data in the 1970's.

Figure 1: Traced-back references in IAEA reports for feed-to-cow's milk transfer coefficient for Cs. Table 1 shows the reference code we used.



Cs	Ι	Pu	Code used	Authors (year) and reference information (report number, article title, etc.)
•	•	•	Baes (1984)	Baes, C. F. et al. (1984) Oak Ridge Natl. Lab. ORNL-5786.
	•		Comar (1963)	Comar, C.L (1963) Federation proceedings 22, 1402-1409.
•	•	•	Coughtrey (1983)	Coughtrey, P.J., et al. (1983) Radionuclide distribution and transport in terrestrial and aquatic ecosystems, vol. 1, Taylor and Francis.
•	•	٠	Coughtrey (1990)	Coughtrey, P.J. (1990) EUR series 12608, Commission of the European Communities.
	•		Garner (1972)	Garner, R.J. and Comar, C.L. (1972) CRC Crit. Rev. Environ. Contr. 2, 337-385.
	•		Hoffman (1978)	Hoffman, F.O. (1978) Health Phys. 35, 413-416.
٠	٠	٠	Hoffman (1980)	Hoffman, F.O., et al. (1980) Oak Ridge Natl. Lab. ORNL/TM-7386.
	•		Hoffman (1988)	Hoffman, F.O., et al. (1988) J. Environ. Radioactiv. 8, 53-71.
	•		Handl (1989)	Handl, J., Pfau, A. (1989) Sci. Total Environ. 85, 245-252.
•	٠	٠	IAEA (1982)	IAEA (1982) IAEA Safety Series No.57.
•	٠	٠	IAEA (1987)	IAEA (1987) IAEA-TECDOC-401.
•	•	•	IAEA (1994)	IAEA (1994) IAEA Technical Reports Series No. 364.
	•		IAEA (1998)	IAEA (1998) IAEA-TECDOC-1000.
•	٠	٠	IAEA (2001)	IAEA (2001) IAEA Safety Reports Series No. 19.
•	•	٠	IAEA (2010)	IAEA (2010) IAEA Technical Reports Series No. 472.
	•		Lengemann (1964)	Lengemann F. W., Comar C.L (1964) Health Phys. 10, 55-59.
	•		Lengemann (1969)	Lengemann F. W. (1969) Health Phys. 17., 565-569.
•	•	•	NCRP (1996)	National Council on Radiation Protection and Measurement (NCRP) (1996) Report No.123.
•	•	•	Ng (1977)	Ng, Y.C. et al. (1977) Lawrence Livermore Lab. UCRL-51939.
	•		Ng(1979)	Ng, Y. C., C. S. Colsher, S. E. Thompson (1979), IAEA-SM-237/54.
	٠		Weiss (1975)	Weiss, B.H., et al. (1975) NUREG-75/021.
	•		Weiss (1977)	Weiss, B.H., Keller J.H. (1977) Paper #184 International Radiological Protection Association Annual Meeting.

Table 1: List of references for feed-to-cow's milk transfer data for Cs, I and Pu, and reference code used

The results for I are shown in Figure 2; for many cases, the original literature was classified into C1 or C2. Some papers could be accessed for reading and thus we could confirm that the data originated from experimental data; however, unfortunately, we could not read some of the original literature as shown by the yellow shading in Figure 2, because they were published as books or laboratory reports and are not available digitally. Because review papers and reports in the C4 summarized data category were provided as results of data surveys at times which we could not identify, we needed further original data collation to confirm accuracy of these data.

Figure 2: Traced-back references in IAEA reports for feed-to-cow's milk transfer coefficient for I. Table 1 shows the reference code we used.



The C_{ft_milk} values of Cs, I and Pu in major publications which are widely cited in dose assessment (shown in Table 1) are plotted in Figure 3 for comparison. Clearly the values for Cs and I were similar across the literature probably because the original data sources were similar. However, interestingly, data for Pu varied by three orders of magnitude, which was much larger than the variations for Cs and I, probably because of the relatively short time period for radiotracer experiments compared to global fallout observations, and also because relationships among chemical forms of Pu are complex. In addition, the difficulty in making Pu measurements has led to only a few Pu C_{ft_milk} values being reported.



Figure 3: Comparison of C_{ft_milk} values for I, Cs and Pu.

The amounts of farm animal products consumed per person in Japan are smaller than those consumed per person in European countries [14], several-fold differences in parameter values have little effect on the dose assessment results in Japan; however, the large variation of C_{ft_milk} for Pu could cause large uncertainty of the assessment results. Further detailed studies are needed to provide more reliable data for Pu transfer to milk.

3.2 Survey of recently published literature values

The original data sources used in the IAEA reports to supply recommended values were obtained about 50 to 70 years ago. Numbers of data used for obtaining the latest parameter values in IAEA TRS 472 were 104 for I and 288 for Cs, but no original data sources for Pu were listed. These values are still valuable, however, it would be better to provide updates by adding recently observed C_{ft_milk} values. Howard et al. [15] reported TRS 472 updates by adding or excluding data; C_{ft_milk} values for Cs, I and Pu in TRS 472 were 4.6×10^{-3} , 5.4×10^{-3} and 1.0×10^{-5} d/L, respectively, and those in ref [15] were 4.9×10^{-3} , 6.0×10^{-3} , and 3.6×10^{-5} d/L (N=3), respectively.

In the list of data used in ref. [15], data observed in Japan were not included; we found several reports on C_{ft_milk} of radiocaesium and ¹³¹I observed after the Fukushima Daiichi Nuclear Power Plant accident, but no data were found for Pu. The recently published IAEA Tecdoc 1927 [16] provided details of the C_{ft_milk} data in Japan and the results were (2.0-2.3)×10⁻³ d/L for Cs, and 3.6×10⁻³ d/L for I. These values were similar to those reported by Howard et al. [15].

Figure 4 compares the C_{ft_milk} results for Cs published in literature reported after 2010 as well as unused values in TRS 472 [17-25]. We found that the values varied within two orders of magnitude. From the figure, we clearly saw that some of the C_{ft_milk} data reported by Karunakara et al. [24] were slightly higher than other values; the former values were from locally bred cows (cows of local farmers). According to ref [24], it was assumed that probably because of their higher soil ingestion, lower body mass and lower milk yield compared to dairy farm cows, C_{ft_milk} values for the locally bred cows were about one order of magnitude higher than the TRS 472 values [8], although dairy farm cows in India [23] had similar results to the TRS 472 values. For more realistic dose assessment, data observed in the specific regions need to be considered.



Figure 4: Comparison of feed transfer coefficient ($C_{ft milk}$, d/L) values for Cs published after 2010.

4 CONCLUSIONS

In this paper, we carried out tracing back of C_{ft_milk} data for Cs, I and Pu used in IAEA to confirm the original data. The original data were mostly based on tracer experiments or global fallout observation results (designated as category C1) and calculated data using metabolic models (designated as category C2); some of the original data sources were difficult to obtain. The C_{ft_milk} values for Cs and I were similar in the literature, but those for Pu varied by three orders of magnitude. We survey recent data and that allowed us to add some more data from Japan and India for Cs; data for I still remained scarce and no further data addition for Pu was possible. Although the C_{ft_milk} values of Pu were much smaller than those of Cs and I, to reduce uncertainty for dose assessment from Pu, further studies are necessary.

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