Radioactivity in Crude Drugs Imported from Asian Countries

Shojiro Kimura,[a] Rumi Yamaoki,*,[a] Yuka Matsumoto,[a] Noriko Fujita,[a] and Kesamitsu Shimizu[b]

[a]*Osaka University of Pharmaceutical Sciences, 4–20–1 Nasahara, Takatsuki, Osaka 569–1094, Japan and* [b]*Kanebo Healthcare Research Laboratory, 3–1 Kanebo, Takaoka, Toyama 933–0856, Japan*

(Received June 24, 2002; Accepted September 2, 2002)

We measured radioactivity levels of ^{137}Cs and ^{40}K in crude drugs imported from Asian countries by γ-ray spectrometry using Ge detector — MCA. Concentrations of ^{137}Cs in imported crude drugs were very low and close to the limits of detection (about 0.3 Bq/kg dry weight). Furthermore, the type of radioactivity absorbed differed among species. The concentration of ^{137}Cs was high only in ASIASARI RADIX, whereas ^{40}K concentrations were high in herb drugs such as PLANTAGINIS HERBA.

Key words —— crude drug, cesium-137, potassium-40, radioactivity, East Asia

INTRODUCTION

Since the 1986 Chernobyl nuclear accident, radioactive contamination of dietary crops has been studied by many European investigators. Mushrooms were found to be highly contaminated with radiocesium.[1–3)]

The Japanese Ministry of Health, Labor and Welfare has established a provisional limit of 370 Bq/kg (the sum of ^{134}Cs and ^{137}Cs) for foods imported from European countries into Japan. In 1987, concentrations of radioactivity in some foods such as herbs or mushrooms imported from European countries exceeded the provisional limit. Because the radioactive concentrations of some herbs and mushrooms remained above 50 Bq/kg, the radioactivity levels of these foodstuffs are still being monitored.[4)] However, radioactive concentrations in herbs and crude drugs imported for medicinal purposes are not monitored. The international demand for medicinal herbs or crude drugs has been increasing, leading to a need to guarantee their quality, safety and efficacy. The WHO guidelines for the quality of crude drugs describes the levels of contamination by radioactive substances determined by qualification testing.[5)] Curini *et al*. found that leaves and flowers of some medicinal plants from Eastern Europe were exposed to fall-out immediately after the 1986 Chernobyl accident, and that the ^{137}Cs concentration in *Pini silvestris gemma* increased with elapsed time because of the turnover mechanism from soil to plants.[6)] Chereji *et al*. reported that the radiocesium concentration in a perennial medicinal plant of Transylvania, *Lichen Islandicus*, was 1577 Bq/kg dry weight in 1994.[7)]

Most crude drugs used in Japan are imported from East Asian countries. We therefore examined ^{137}Cs concentrations in imported and domestic crude drugs that are frequently used in Japan. We also investigated the ^{40}K content of the natural radionuclide for contrast.

MATERIALS AND METHODS

Drug Sources —— Imported crude drugs (15 species, 58 samples); roots and rhizomes: ASIASARI RADIX (saishin), GLYCYRRHIZAE RADIX (kanzo), PAEONIAE RADIX (shakuyaku), REHMANNIAE RADIX (jio), ALISMATIS RHIZOMA (takusha), RHEI RHIZOMA (daio), ZINGIBERIS RHIZOMA (shokyo), MOUTAN CORTEX (botanpi); herbs, flowers and barks: EPHEDRAE HERBA (mao), MENTHAE HERBA (hakka), PERILLAE HERBA (soyo), PLANTAGINIS HERBA (shazenso), MAGNOLIAE FLOS (shin-i), CINNAMOMI CORTEX (keihi); sclerotium: PORIA (bukuryo); earthworm: LUMBRICUS (jiryu). Domestic crude drugs (3 species, 5 samples); roots: ANGELICAE RADIX (touki), GINSENG RADIX (ninjin); herbs: PLANTAGINIS HERBA. Generally, the producer collects, washes, dries below 60°C the crude medicinal potions of wild or cultivated species. These dried sources are then shipped as crude drugs to the market place. All samples were obtained from wholesalers of crude drugs in Japan between 1991 and 2001.

Sample Preparation —— Cut sources, and dried whole specimens were crushed and pressed into 1

*To whom correspondence should be addressed: Osaka University of Pharmaceutical Sciences, 4–20–1 Nasahara, Takatsuki, Osaka 569–1094, Japan. Tel: +81-726-90-1043; Fax: +81-726-90-1042; E-mail: yamaoki@gly.oups.ac.jp

or 2 l Marinelli beakers depending on the amount of the samples.

Radioactivity Measurements —— We measured radioactivity levels in PLANTAGINIS HERBA and LUMBRICUS using a Ge(Li) detector (Ortec, 8001-1521w) and in the others using a pure Ge detector (Ortec, GEM-15190-P). The detectors were connected to a multichannel analyzer (Camberra, Series 95). Counting times were 80000–270000 sec. The ^{137}Cs concentration was calculated by deducting the contribution of the ^{214}Bi (0.6655 MeV) photopeak from the γ-ray photo-peak of ^{137m}Ba (0.662 MeV). If no ^{137m}Ba peak was detected, the concentration was not calculated. The ^{40}K concentration was calculated by deducting the photo-peak of the background from that of the γ-ray (1.46 MeV). The stable potassium contents in crude drugs was calculated from the ^{40}K values as the natural isotopic ratio of ^{40}K; 0.0117%. We compared the results with those of standard materials (Laboratorie de Metrologie des Rayonnements Iomsants) to validate analytical quality, and established 2.0 (2σ) as the detection limit. Under these conditions, the detection limits of ^{137}Cs were about 0.3 Bq for the Ge and 0.6 Bq for the Ge(Li) detectors. Relative counting errors for the measurements were better than 10%, but worse than 20% when the radioactive concentration was at the limits of detection.

RESULTS AND DISCUSSION

Table 1 shows ^{137}Cs and ^{40}K concentrations, stable potassium contents and ^{137}Cs/^{40}K ratios in imported and domestic crude drugs. The 52% ^{137}Cs concentrations in crude samples were below the limits of detection. The values of twenty root and rhizome samples, eight herb, flower and bark samples, and two earthworms were slightly above 0.3 Bq/kg dry weight. The ^{137}Cs concentrations differed among species, but relevance to the source region or year was not clear. We previously investigated the radioactive concentrations in imported and domestic HOUTTUYNIAE HERBA (jyuyaku, herb drug). Both the ^{137}Cs and ^{40}K concentrations in domestic HOUTTUYNIAE H. samples (mean values, 2.9 and 1639 Bq/kg dry weight) tended to be higher than those in imported samples from China and Korea (mean values, 0.7 and 1309 Bq/kg dry weight).[8] The present study found that the concentration of ^{137}Cs in the domestic PLANTAGINIS HERBA was slightly higher than that in the imports. Among the root drugs, the ^{137}Cs concentrations of Japanese ANGELICAE RADIX and GINSENG RADIX were below 0.3 Bq/kg dry weight, and were the same as the imported root drugs such as GLYCYRRHIZAE RADIX, PAEONIAE RADIX and REHMANNIAE RADIX except ASIASARI RADIX. The ^{137}Cs concentrations of ASIASARI R. samples were higher than in any other species. The ^{137}Cs concentration was highest in ASIASARI R. imported from North Korea in 1993 (17 Bq/kg dry weight). This radioactivity level is the same as the mean ^{137}Cs concentration in an edible mushroom (*Lentinula edodes*, 15.0–22.7 Bq/kg dry weight) cultivated in Japan.[9,10] In contrast, the ^{137}Cs concentrations of Chinese ASIASARI R. obtained in the same year were bellow 20% of the highest value. In comparison with the 1993 ASIASARI R. samples, the variation in ^{137}Cs values among the same species obtained between 2000 and 2001 was smaller. China performed atmosphere nuclear weapons testing from the 1960s to the 1980s. Furthermore, radioactive contamination expanded worldwide after the Chernobyl accident. However, even the highest ^{137}Cs concentration in ASIASARI R. among these crude samples was bellow 5% of the provisional limit (370 Bq/kg) for foods imported from European countries into Japan. This result indicated that crude drugs in East Asia were not highly contaminated by fallout from these events.

The concentration range of ^{40}K and stable potassium in crude drugs were 6–1115 Bq/kg and 0.02–3.6% of the dry weight. The ^{40}K concentrations among species varied more widely compared with those of ^{137}Cs. The value of potassium was the lowest in PORIA, in the sclerotium of *Poria cocos* Wolf, and a low stable potassium content of 0.057% of the dry weight has also been determined by atomic absorption.[11] Conversely, the contents of potassium in herb drugs such as MENTHAE HERBA, PERILLAE HERBA and PLANTAGINIS H. were high. The concentrations of ^{40}K in domestic and imported PLANTAGINIS H. were similar, and the stable potassium contents were 2.0–3.6% of the dry weight. These values are same as those of stable potassium in cabbage (0.20% of the wet weight, water content 92.7%).[12] Among the root drugs, ASIASARI R. and GLYCYRRHIZAE R., the variations among individual samples were larger, and the differences of values between samples were more than double. Crude drugs are sometimes traded through brokers, so the origin may be obscure. Thus, the variation in ^{40}K content between ASIASARI R.

Table 1. ^{137}Cs and ^{40}K Concentrations and Stable Potassium Contents in Imported and Domestic Crude Drugs

Source Species	Number	Origin		Year	^{137}Cs (Bq/kg dry wt)	^{40}K (Bq/kg dry wt)	K (% dry wt)	$^{137}Cs/^{40}K$
Roots and Rhizomes (below ground)								
ASIASARI RADIX	11	NORTH KOREA		1993	17	287	0.9	0.061
Asiasarum sieboldii F. Maekawa,				2000/5	6.4	282	0.9	0.023
A. heterotropoides F. M. *var.*				2000/10	7.8	270	0.9	0.029
mandshuricum F. M.				2001/1	10	313	1.0	0.033
		KOREA		1993	7.1	332	1.1	0.021
				2000	8.1	278	0.9	0.029
		CHINA	Liaoning	1993	3.5	146	0.5	0.024
			Heilungkiang	1993	2.1	266	0.9	0.008
				2000	5.9	254	0.8	0.023
				2000	9.5	307	1.0	0.031
			Kirin	2001/1	7.2	287	0.9	0.025
				(Mean	7.7	275	0.9	0.028)
GLYCYRRHIZAE RADIX	16	CHINA	Inner Mongolian	1996	< 0.3	213	0.7	
Glycyrrhiza uralensis Fisher				1999	0.3	86	0.3	0.003
				1999	< 0.3	209	0.7	
				1999	0.3	74	0.2	0.004
				1999	< 0.3	55	0.2	
			Hopeh	1999	0.3	91	0.3	0.003
				1999	< 0.3	114	0.4	
			Ningsia Hui	1999	< 0.3	126	0.4	
				1999	0.3	129	0.4	0.001
				1999	< 0.3	107	0.3	
				1999	< 0.3	132	0.4	
			Shensi	1999	0.3	251	0.8	0.001
				1999	< 0.3	208	0.7	
				1999	< 0.3	136	0.4	
				1999	< 0.3	208	0.7	
			Kansu	1999	< 0.3	135	0.4	
				(Mean	0.3	142	0.5	0.003)
PAEONIAE RADIX	5	CHINA	Anhwei	1999	—	161	0.5	
Paeonia lactiflora Pallas		NORTH KOREA		2000	—	162	0.5	
				2001	0.4	161	0.5	0.003
				2001	—	174	0.6	
				2001	—	170	0.5	
				(Mean	0.4	166	0.5	0.003)
REHMANNIAE RADIX	1	CHINA	Honan	1987	< 0.3	542	1.8	
Rehmannia glutinosa Liboschitz								
ANGELICAE RADIX	1	JAPAN		2001	—	533	1.7	
Angelica acutiloba Kitagawa								
GINSENG RADIX	1	JAPAN		1999	0.3	449	1.5	0.001
Panax ginseng C. A. Meyer								
ALISMATIS RHIZOMA	2	CHINA	Szechwan	2000	< 0.3	270	0.9	
Alisma orientale Juzepczuk			Kwangsi Chuang	2000	—	290	0.9	

— : not detected.

Table 1. Continued

Source Species	Number	Origin		Year	^{137}Cs	^{40}K	K	$^{137}Cs/^{40}K$
					(Bq/kg dry wt)		(% dry wt)	
RHEI RHIZOMA	3	CHINA	Szechwan	2001	2.8	460	1.5	0.006
Rheum palmatum Linné,				2001	4.7	362	1.2	0.013
R. tanguticum Maximowicz				2001	< 0.3	308	1.0	
				(Mean	< 2.6	377	1.2	0.009)
ZINGIBERIS RHIZOMA	2	CHINA	Yunnan	1997	< 0.3	478	1.5	
Zingiber officinale Roscoe				2000	< 0.3	433	1.4	
MOUTAN CORTEX	1	CHINA	Anhwei	2000	< 0.3	162	0.5	
Paeonia suffruticosa Andrews								
Herbs, Flowers and Barks (above ground)								
EPHEDRAE HERBA	4	CHINA	Inner Mongolian	1999	—	354	1.1	
Ephedra sinica Stapf				2001	—	352	1.1	
				2001	—	337	1.1	
				2001	—	343	1.1	
				(Mean		347	1.1)	
MENTHAE HERBA	2	CHINA	Shanghai	2000	—	664	2.1	
Mentha arvensis Linné var.			Kiangsu	2001	0.4	582	1.9	0.001
piperascens Malinvaud								
PERILLAE HERBA	1	CHINA	Kiangsu	2000	—	782	2.5	
Perilla frutescens Britton								
PLANTAGINIS HERBA	4	CHINA	Kiangsi	1991	1.3	1115	3.6	0.001
Plantago asiatica Linné		KOREA		1991	0.7	978	3.2	0.001
				1991	1.0	645	2.1	0.001
				1991	0.9	740	2.4	0.001
				(Mean	1.0	870	2.8	0.001)
	3	JAPAN	Miyazaki	1991	2.1	1047	3.4	0.002
			Miyazaki	1991	1.7	634	2.0	0.003
			Tokushima	1991	1.5	881	2.8	0.002
				(Mean	1.8	854	2.8	0.002)
MAGNOLIAE FLOS	1	CHINA	Honan	1993	< 0.3	270	0.9	
Magnolia kobus DC.								
CINNAMOMI CORTEX	1	CHINA	Kwangtong	2000	< 0.3	118	0.4	
Cinnamomum cassia Blume								
Sclerotium								
PORIA	2	CHINA	Anhwei	1995	< 0.3	25	0.1	
Poria cocos Wolf			Yunnan	2001	< 0.3	6	0.02	
Earthworm								
LUMBRICUS	2	THAILAND		1991	2.0	429	1.4	0.005
Pheretima asiatica Michaelsen				1991	1.8	499	1.6	0.003

and GLYCYRRHIZAE R. samples might be due to the sources of the wild plants.

The range of $^{137}Cs/^{40}K$ ratio in crude drug samples was < 0.001–0.061. The $^{137}Cs/^{40}K$ ratio of ASIASARI R. samples was one order of magnitude greater than those of most of other samples. The accumulation of ^{137}Cs in ASIASARI R. may be affected by the part of the plant that is medicinal and by characteristic of the growth location. ASIASARI R. is the fibrous root and rhizome of *Asiasarum sieboldii* or *A. heterotropoides*, which are both perennial plants and which grow in wet humus soil under trees. Organic soil matter increases plant uptake of cesium.[13)]

The present study found that the concentration of ^{137}Cs in imported crude drugs from Asian countries is very low. Thus, radioactive contamination in these crude drugs is not an issue. However, over 350 species of crude drugs are used in Japan, and they are sourced from plants, animals or minerals. Consequently, when wild species are sourced, species that accumulate ^{137}Cs should be carefully monitored.

REFERENCES

1) Borio, R., Chiocchini, S., Cicioni, R., Degli Esposti, P., Rongoni, A., Sabatini, P., Scampoli, P., Antonini, A. and Salvadori, P. (1991) Uptake of radiocesium by mushrooms. *Sci. Total Environ.*, **106**, 183–190.
2) Smith, M. L., Taylor, H. W. and Sharma, H. D. (1993) Comparison of the post-Chernobyl ^{137}Cs contamination of mushrooms from Eastern Europe, Sweden, and North America. *Appl. Environ. Microbiol.*, **59**, 134–139.
3) Amundsen, I., Gulden, G. and Strand, P. (1996) Accumulation and long term behaviour of radiocaesium in Norwegian fungi. *Sci. Total Environ.*, **184**, 163–171.
4) Kondo, T. (1999) Yunyushokuhin no houshanou-kensa kekka. *Isotope News*, **4**, 12–17.
5) World Health Organization (1992) *Quality control methods for medicinal plant materials*, WHO, Geneva.
6) Curini, M., Rosati, O., Borio, R., Saetta, D. M. S., Cicioni, R., Forini, N., Rongoni, A. and Dipilato, A. C. (1995) Evaluation of ^{137}Cs activity in plant drugs and in some phytoderivatives from Chernobyl accident up to present. *Pharmacol. Res.*, **32**, 69–74.
7) Chereji, I., Daraban, L., Dreve, S., Boscaneanu, S., Cosma, C. and Vari, E. (1996) Chernobyl-derived radiocesium in some pharmaceutical plants. *J. Radioanal. Nucl. Chem.*, **212**, 85–92.
8) Kimura, S. and Yamaoki, R. (1997) Distribution of ^{137}Cs in environmental soil and crude drugs. *J. Toxicol. Environ. Health*, **43**, 349–358.
9) Ban-nai, T., Muramatsu, Y. and Yoshida, S. (1997) Concentrations of ^{137}Cs and ^{40}K in edible mushrooms collected in Japan and radiation dose due to their consumption, *Health Phys.*, **72**, 384–389.
10) Shimizu, M., Anzai, I., Fukushi, M. and Nyuui, Y. (1997) A study on the prefectural distribution of radioactive cesium concentrations in dried *Lentinula edodes* (Shiitake) produced in Japan. *Radioisotopes*, **46**, 272–280.
11) Suzuki, A., Morimoto, I. and Okitsu, T. (1982) Elution of metals from crude drugs. *Shoyakugaku Zasshi*, **36**, 190–195.
12) The Resources Council of the Science and Technology Agency of Japan (2000) *Standard Tables of Food Composition in Japan, Fifth Revised Edition*, Printing Bureau, Ministry of Finance, Tokyo.
13) Malm, J., Rantavaara, A., Uusi-Rauva, A. and Paakkola, O. (1991) Uptake of Caesium-137 from Peat and Compost Mould by Vegetables in a Greenhouse Experiment. *J. Environ. Radioact.*, **14**, 123–133.