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Radionuclides in tea and their behaviour in the brewing process

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ABSTRACT

Tea plantations may be strongly affected by radioactive fallout. Tea plantations on the Turkish coast of the Black Sea were heavily contaminated by the fallout from the reactor fire at the Chernobyl nuclear power plant in 1986. Two years later, the contamination level was reduced by about 90%. When tea is brewed, the original contamination in the tea leaves is more or less leached into the tea water. While most of the radiocaesium (60–80%) is washed out by brewing, most of the radiostrontium remains in the leaves (70–80%). In food laws, a dilution factor of 40–50 is considered for tea brewing. Most laws only define limit values for radiocaesium. Radiostrontium is not specially regulated, even though its dose coefficients for ingestion are higher than the corresponding coefficients for radiocaesium. Radiostrontium in tea occurs primarily from global fallout (bomb tests from 1945-1965).

1. Introduction

Tea is one of the most popular beverages; after drinking water, it is the most consumed drink worldwide. The tea plant, *Camelia sinensis*, is cultivated in many countries, including the People's Republic of China, India, Kenya, Sri Lanka, Turkey, Indonesia, Vietnam, Japan, Argentina and Iran. Black and green tea are the most consumed and most common teas marketed by commercial brands. Worldwide, it is estimated that over three million tons of tea is produced each year. Tea plantations need large areas for production. The tea plant reaches a height of 1–5 m and carries many leaves, exposing a large surface to fallout (Abd El-Aty et al., 2014; Hirono and Nonaka, 2016). Therefore, tea is an ideal plant for the monitoring and investigation of radioactive fallout. Fallout reaches plantations via air and rain and radionuclides are deposited on the surface of leaves and on the soil. The tea plant takes up the radiocesium from soil and translocates it quickly to the leaves (Takashi et al., 2018; Hinton et al., 1996). The part of ^{137}Cs deposited on the leave surface will be quickly removed by dissolution with water (Yasuhisa et al., 2015; Keiko et al., 2012). ^{137}Cs is reported to be present mainly in cationic form. However, there is also evidence of ^{137}Cs bound to polyphenols in tea (Emine, 2002). During the growing season, tea is harvested weekly. The main harvests are in March/April (first flush), in May/June (second flush) and in autumn (autumnal flush). This results in a continuous renewing of the tea leaves. After harvest, the process of tea manufacturing consists of several steps (withering, rolling, fermentation, drying) (Hirono and Nonaka, 2016; Pintauro, 1977).

From 1945 to 1965, there were over 600 nuclear bomb tests in the atmosphere. In 1963 to 1965, global fallout, consisting mainly of long-lived radionuclides of caesium (^{134}Cs, ^{137}Cs), strontium (^{90}Sr) and plutonium (^{239}Pu, ^{240}Pu), reached a maximum in the northern hemisphere. Most important was the ^{90}Sr fallout. Until 1976, scientists estimate a cumulated deposition of ^{90}Sr of 4,5 10^{17} Bq in the Northern hemisphere and 9.3 10^{16} Bq in the Southern hemisphere (Feely et al., 1978).

In 1963, a partial test ban treaty was enacted (ban on all atmospheric bomb tests). Therefore, fallout was reduced remarkably after 1963. In 1964, the contamination level of tea cultivated in India was about 37 ± 36 Bq/kg dry weight (d.w.) of ^{137}Cs and 26 ± 11 Bq/kg d.w. of ^{90}Sr (Lalit et al., 1983). The contamination level started to decline, and from 1961 to 1985, activity levels in Indian tea did not exceed 20 Bq/kg (Lalit et al., 1983). Table 1 summarises the contamination levels of tea from 1961 to 2011 found in literature and results from own investigations.

A further global input of radionuclides came from the fallout released in 1986, when the nuclear catastrophe at the Chernobyl NPP happened. The reactor fire caused a rise of the radio-contamination level in whole Europe and Turkey. Turkish tea, harvested in 1986, showed activities up to 30,000 Bq/kg d. w. of radiocaesium ($^{134+137}$Cs) and up to 430 Bq/kg d.w. of radiostrontium (^{90}Sr) (Zehringer, 2016; Gökmen et al., 1995). Turkish tea plantations on the south coast of the Black Sea were most affected, and 57% of analysed Turkish teas exceeded the limit for radiocaesium (500 Bq/kg in tea water) (Gökmen et al., 1995). After 1987, the activity level in Turkish tea was reduced again to a level of 1–70 Bq/kg d.w. of ^{137}Cs (Zehringer, 2016; Gökmen

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Table 1
Radio contaminations in tea from 1961 to 2011. All activities in Bq/kg dry weight. n: number of analysed tea samples, s.d.: standard deviation, n.a.: not analysed. *mean values, no medians available

Year	Country	Reference	Radiocaesium (^{137}Cs) median ± s.d.	Radiostrontium (^{90}Sr) median ± s.d.
1961	India, Sri Lanka	Lalit et al., 1983	14 ± 29	3.4 ± 4.3
1962	India	Lalit et al., 1983	6.5 ± 3.8	11 ± 8.5
1964	India	Lalit et al., 1983	73 ± 29	38 ± 16
1968	Taiwan	Chu et al., 1969	3.1 ± 3.2	14 ± 8.5
1974	India	Lalit et al., 1983	5.0 ± 2.8	19 ± 25
1975	India	Lalit et al., 1983	4.7 ± 1.9	18 ± 3.8
1977	India	Lalit et al., 1983	1.9 ± 2.9	10 ± 5.8
1979	India	Lalit et al., 1983	1.1 ± 0.1	n.a.
1985	Peop. Rep. China	Sha et al., 1993	2.2 ± 0.2*	17 ± 9*
1986	Turkey	Zehringer, 2016	8580 ± 10,290	430
1986	Turkey	Gökmen et al., 1995	12,870 ± 6266	n.a.
1987	Turkey	Gökmen et al., 1995	7250 ± 3340	n.a.
1995	Peop. Rep. China	He, 1995	n.a.	18*
2000	Peop. Rep. China	Lu et al., 2006	0.3 ± 0.05*	3.0 ± 1.1*
2006	Korea	Dersee, 2008	0.7*	n.a.
2007	Turkey, Sri Lanka, Lebanon	Dersee, 2008	35 ± 30	n.a.
2008	Peop. Rep. China	Dersee, 2008	0.5 ± 5.8	n.a.
2008	India, Russia			
2009	Turkey	Zehringer, 2009	23 ± 16	n.a.
2010	Turkey	Görür et al., 2011	35 ± 28	n.a.
2010	Turkey	Sahin et al., 2011	n.a.	14 ± 19
2015	Turkey	Zehringer, 2016	34 ± 7	31 ± 8

et al., 1995). With the exception of the heavily contaminated Turkish tea from the Black Sea coast, there were no more violations of the Swiss food law. However, contamination levels remained elevated even more than 30 years after the catastrophe in Chernobyl. In 2015, after more than one half-life of ^{137}Cs and ^{90}Sr later, some Turkish teas still contained up to 50 Bq/kg d.w. of ^{137}Cs and 38 Bq/kg d.w. of ^{90}Sr (Zehringer, 2016a).

With regard to food safety, it is important to understand whether tea water prepared with contaminated tea is still safe. Therefore, the behaviour of radionuclides during the brewing process was studied.

2. Material and methods

2.1. Sampling

All tea samples were collected on the local market in Basel, Switzerland, by our food inspection team. From 1986 on, Turkish tea has been under constant scrutiny because of many violations to the limits for radiocaesium (^{134}Cs and ^{137}Cs) due to the heavy fallout in northern Turkey. After the melting of the core at the Fukushima Daiichi NPP in 2011, the state-laboratory of Basel-City changed their focus to Japanese tea, and started a monitoring of importations to Switzerland.

Without further treatment, the dry, broken tea was placed into well-defined counting devices. The date of the sampling was taken as the reference date. All activities were corrected to this date.

2.2. Tea brewing experiments

We chose two Turkish teas for the investigation of the behaviour of radionuclides during the brewing process. In all experiments, the leaching behaviour of ^{137}Cs was studied in a threefold extraction process. Ten g of dried tea leaves were brewed with 1 L of boiling tap water for 10 min. The tea leaves were then filtered off using a paper filter. Both, the tea leaves and the tea water, were analysed with gamma ray spectrometry. The used tea leaves were brewed again for a second time. In a second similar brewing test, ^{90}Sr was analysed with beta spectrometry as described above. We used two of the most contaminated Turkish teas for these experiments: Yuvam Gold black tea from the province Manisa and Caykur black tea from the province Rize. Both teas were from the harvest of 2009.

2.3. Gamma spectrometry

The radiation levels of tea were analysed with high-resolution gamma spectrometry. We used germanium detectors in coaxial geometry with relative efficiencies of 35% and 50%. All detectors were shielded with 10 cm of lead. Due to different sample volumes, several efficiency-calibrated geometries were used: petri dishes of 12 mm and 24 mm height (diameter of 60 mm), cylindrical bottles of 250 and 500 mL volume, and 1 L Marinelli beakers for the tea water samples. Background spectra of the water-filled devices were subtracted from the sample spectra. Detector efficiencies were determined with ^{241}Am/^{152}Eu-reference sources (Czech Metrology Institute, Prague). All detectors were calibrated from 59 to 1408 keV. The peak resolution, full width at tenth maximum (FWTM) versus full width at half maximum (FWHM), was 1.2–2.5 keV. Each sample was counted for at least 86,000 s. ^{134}Cs was identified and quantified using the following gamma ray energies (emission probability in brackets): 569 keV (15.4), 605 keV (97.6) and 796 keV (85.5). ^{137}Cs was measured using the gamma-ray energy at 662 keV (84.6). ^{40}K was measured at 1460.5 keV (10.67). We used Interwinner 7.0 from ITEC INSTRUMENTS for data recording and spectra analyses. Attenuation effects due to sample matrix and sample density were corrected, whereas we did not correct for coincidence summing effects of ^{134}Cs. As an accredited laboratory (according to the European Guidelines ISO/IEC 17025:2005) we are obliged to take part at yearly-organised proficiency tests. As in the last years the matrix tea was never tested, we came back to a reference material from the International Atomic Energy Agency (IAEA), a moss-soil material (IAEA-447). The ^{137}Cs-activity of the test material was recovered within a z-score of two.

2.4. Beta spectrometry

All reagents used for sample preparation were of analytical grade. The pure beta-emitter ^{90}Sr was determined indirectly via its daughter yttrium-90 (^{90}Y) with beta spectrometry. Aliquots of 10–20 g of tea were dried at 120 °C and then ashed at 600 °C. The ashes were then dissolved in 10% HCl. A defined portion of YCl_3-solution was added to the solution as a carrier. Strontium and other earth alkaline metals were precipitated as oxalates. The oxalates were separated by filtration and ashed again at 850 °C. The residue was dissolved in 25% HCl and $SrCl_2$