

Migration levels of monostyrene from polystyrene containers to dairy products

Abstract

The level of monomer styrene migrated from polystyrene containers (PS) was measured in different dairy products at various fat contents, storage periods and temperatures. The dairy products were packed in PS containers and divided into groups according to storage period and temperature. The samples were included whole milk (3.6% fat) kept at 100°C for two hours (group1), yogurt (3% fat), sour cream (6% fat), cream (30% fat) and drinking yogurt (3% fat) stored in polystyrene packages at 4°C for 14days (group2), mozzarella cheese (14.0% fat), cheddar cheese (34.4% fat) and butter (81.7% fat) stored at 4°C for 60days (group3) and ice-cream (16% fat) stored at -10°C for 60 ays (group4). Extending storage time and increasing temperature were found to be significantly elevated ($p \leq 0.05$) the migration rate of styrene. Styrene migration was found to be strongly ($p \leq 0.05$) dependent upon the fat content. Maximum migration limit has occurred with the highest fat content; butter (81.7%) at a concentration of 0.102mg/kg.

The calculated consumed quantity of styrene from each dairy food items in adults ranged from 0.018 to 0.170 μ g/kg wt/day which corresponding to 1.1-10.2 μ g/day/person while, total daily consumption of all the tested products is amounted to 50.6 μ g/day. The study showed that the styrene intake from each tested dairy product item is less than the international allowable level. The results postulate that consumption of foods rich in fat content especially, at high storage temperatures in PS packages could represent health hazard while they are highly stimulating migration of monostyrene.

Keywords: migration of monostyrene, dairy polystyrene packages, daily intake

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Introduction

In the modern society, food packaging plays an important role to promote safe transportation, delivery and storage of food. It becomes an indispensable element in the food sector. Among the major polymers used in food packaging, polystyrene (PS) has made up a large volume of the consumption of plastic containers. Polystyrene is widely used as food servicing packaging because it is strong, lightweight, and it provides excellent insulation properties. The usage conditions of polystyrene food packaging range from low temperatures for periods of days or weeks, for example packaged dairy and meat products, to high temperatures approaching the boiling point of water for short periods of time, for example vending cups and instant noodle bowls.¹ The low molecular weight constituents present in the polystyrene plastic have the potential to migrate into the foodstuff in contact with the plastic especially during extended periods of time, or at the elevated temperatures. Substances migrating to foodstuffs are of concern if they present a possible health hazard to the consumer, or cause unacceptable changes to the organoleptic properties of the food or beverages.² Numerous studies have shown that styrene and other volatile compounds present in polystyrene food packaging have the potential to migrate out into the contacting food.^{1,3-10}

Styrene was listed as “reasonably anticipated to be a human carcinogen” by the National Toxicology Panel (NTP).¹¹ Prior to this, the IARC classified styrene as “possibly carcinogenic to humans.”¹² Currently, the USEPA does not have a classification for the carcinogenicity of styrene in its Integrated Risk Information System (IRIS).¹³ As a result of the NTP’s recent classification, the USFDA is updating its safety review of styrene by re-evaluating styrene

concentrations in FCMs and foods contained in PS packaging. Recent study showed that monostyrene (MS) at migrated levels affected performance parameters include weight gain (WG), relative liver weight, food efficiency ratio (FER), protein efficiency ratio (PER) and oxidative stress biomarker; malondialdehyde (MDA) in the plasma of male and female albino rats after 12weeks of feeding.¹⁴

PS is widely used in the manufacture of food contact materials (FCMs), and in 2012, 50% of the domestic consumption of PS was related to food packaging and food service articles.¹⁵ An estimate put domestic consumption at 2600million lbs for 2012.¹⁵ During the production process, the styrene monomer can become occluded in PS products and has the potential to migrate out of the FCMs and into the food. Styrene has known toxic effects on the central nervous system and neurological effects have been observed in workers continually exposed to styrene through inhalation.¹⁶ Although no cases have been reported on the ingestion of styrene by humans, the systemic toxicity is expected to be similar to that of inhalation.¹⁶

The main food contact applications of PS are in dairy products including yogurt, cream, cottage cheese, ice cream and fruit juice, meat trays, biscuit trays, egg cartons, and drink cups.¹⁷ In France the consumer exposure to materials migrating from the packaging components to a number of dietary products which are consumed by one family for one year have been estimated. The consumer exposure to styrene from the yogurt packages have been estimated depending on the yearly purchasing information for more than 5400 dwellings (one million yogurt packages). The results indicated that the family exposure to styrene ranges from one to 35ppm/person/day.¹⁸ In USA and Canada the annual per capita intake of styrene is 3.3mg¹⁹ and

from 2.8 to less than 14.8mg respectively,²⁰ while the study²¹ valued the average annual intake of styrene for the people of USA by about 0.08-0.54 mg/person or 1.0-6.5 μ g/kg of the body weight for adults i.e. equivalent to the average annual intake of 0.2-1.2 μ g /person or 3-17ng/kg of the body weight. This value was estimated by FDA using the factor of dietary intake based on assuming that only 10% of the diets wrapped with polystyrene. The daily dietary intake of styrene from the polymer, contacting the food has been estimated for the USA people, and about 3 ppb or equivalent to 9 μ g/day were found as average styrene intake from the daily diet. This value is approaching the dietary styrene intake in the UK, 1983. At the level of the Arab World, Sharja Municipality prohibited- in 2006- the usage of polystyrene cups in delivering the hot drinks. The circulation distributed to the food establishments indicated that the polystyrene cups will not endure high temperature of hot beverages which may lead to seepage of components to the foodstuff and that will affect the safety of foods and beverages in those cups.²²

The work in this research paper are aimed to evaluate the level of dairy products contamination by styrene monomer derived from dairy contact materials into most PS refillable foodstuffs, explore the effect of interacted parameters including the fat content, temperature and storage time on the migration level, and assess the monostyrene daily intake level and potential risk.

Materials and methods

Samples

Different dairy products involved in this study included, whole milk (3.6% fat), yogurt (3% fat), sour cream (6% fat), cream (30% fat), drinking yogurt (3% fat), mozzarella (14.0% fat), cheddar cheese (34.4% fat), butter (81.7% fat), and Ice-cream (16% fat) were obtained from manufacturer and packed in polystyrene containers. The test conditions of the samples are shown in Table 1.

Table 1 Dairy products samples and storage conditions used through present study

Group	Sample type	Number of samples	Storage conditions
1	Whole milk (3.6% fat)	3	2 h at 100°C
	Yogurt (3% fat)	3	14 days/4°C
2	Sour cream (6% fat)	3	14 days/4°C
	Cream (30% fat)	3	14 days/4°C
	Drinking yogurt (3% fat)	3	14 days/4°C
3	Mozzarella cheese (14.0% fat)	3	60 days/4°C
	Cheddar cheese (34.4% fat)	3	60 days/4°C
4	Butter (81.7% fat)	3	60 days/4°C
	Ice-cream (16% fat)	3	60 days/-10°C

Chemicals

Styrene (99%) and α -methyl styrene (99%) were obtained from Fluka and Sigma-Aldrich (Steinheim, Germany). All other chemicals and solvents used were analytical grade, i.e. purity>99%, obtained from Merck (Darmstadt, Germany).

Preparation of styrene standard solutions

Styrene standard stock solution at a concentration of 1000 mg/l was prepared by dissolving 0.2778 \pm 0.01ml of styrene standard (99%) methanol (HPLC-grade). The quantitative analysis of styrene in samples was achieved using a calibration curve.²³

HPLC conditions

Styrene concentration was estimated by high performance liquid Chromatography (HPLC) (Bio-TEK) with UV/VIS absorbance detector and a data module integrator/recorder. The column (250x4.6mm) was packed with Lichrosorb RP-18(10 μ m) and the mobile phase was distilled water-acetonitrile (25%-75%) at flow rate of 1.0ml/min. The system is connected with injector (Bio-TEK), the peak areas under the curve were calculated using a computer (Chromatopac Software, Karosystem 2000, version 1.83 integration). Sample injection volume was 50 μ l. The detector was set at 245nm.

Styrene analysis in food samples

A 50g measure of the homogenized sample was placed into a 250ml round bottom flask, mixed with 50ml of distilled water and 25ml of acetonitrile (HPLC grade). Anti-bumping granules and 2ml of calcium chloride (20%) were added, plus 2ml of α -methyl styrene (4mg/liter in methanol) were added as internal standard. The flask was fitted with a splash head, straight condenser and take-off and the solution was slowly distilled over a period of 1 hour, collecting about 24ml of the distillate in a 25ml volumetric flask. The distillate was made up to 25ml with distilled water (24). The distillate was made up to increments (up to 100 μ l) of styrene working solution (90.6mg/liter in methanol) to a styrene-free sample of the food (calibration standard). An identical volume of internal standard was added to each sample. The retention times for styrene and α -methyl styrene were approximately 6 and 7.5min respectively. Known amounts of styrene monomer (0.05mg/kg) were added to some food samples to determine recovery of styrene from different foodstuffs (Table 2). The limit of detection of the analytical method for styrene analysis in food samples is 0.001mg/kg.

Table 2 Recovery of added monostyrene from various dairy products under experiment conditions

Sample	No. of replica	Amount added (mg/kg)	Recovery %
Whole milk (3.6% fat)	5	0.05	92.1
Yogurt (3% fat)	5	0.05	97.5
Sour cream (6% fat)	5	0.05	93.2
Cream (30% fat)	5	0.05	94.9
Drinking yogurt (3% fat)	5	0.05	95.7
Mozzarella cheese (14.0% fat)	5	0.05	90.8
Cheddar cheese (34.4% fat)	5	0.05	96.6
Butter (81.7% fat)	5	0.05	89.9
Ice-cream (16% fat)	5	0.05	98.1

Results and discussion

Migration of monostyrene from polystyrene (PS) packages into dairy products

Table 3 shows the quantity of monostyrene migrating from the packages of polystyrene to some packed dairy products including, whole milk (3.6% fat), ice-cream (16% fat), cream (30% fat), sour cream (6% fat), yogurt (3% fat), drinking yogurt (3% fat), cheddar cheese (34.4% fat), butter (81.7% fat), mozzarella (14.0% fat).

Table 3 Migration levels of monostyrene from dairy PS packages into different dairy products

Group*	Samples	Monostyrene residues** (mg/kg)
1	Whole milk (3.6% fat)	0.071±0.09 ^d
2	Yogurt (3% fat)	0.011±0.22 ^e
	Sour cream (6% fat)	0.028 ±0.05 ^f
	Cream (30% fat)	0.074±0.17 ^d
3	Drinking yogurt (3% fat)	0.015±0.13 ^e
	Mozzarella cheese (14.0% fat)	0.079±0.11 ^c
	Cheddar cheese (34.4% fat)	0.082±0.06 ^b
	Butter (81.7% fat)	0.102±0.05 ^a
4	Ice-cream (16% fat)	0.044±0.07 ^e

*For group conditions revise Table 1

**Average±standard error (n=3). Average carrying similar letters implies that no significant differences exist ($P\leq 0.05$).

The results indicate that the higher level of monostyrene is found in butter with statistically significance of ($P\leq 0.05$) as it reached (0.102±0.05)mg/kg of the foodstuff weight compared to other tested foods. This high level of monostyrene attributed to the increase of the fat ratio in butter and to the extended storage period in the polystyrene pack before extraction, and this is considered as one of the factors enhancing the increase of migration, as the migration increase directly proportional with time and with the increment of fat ratio.

These results agreed with previous¹⁰ study explained the relation between the range of migration and food ingredients i.e. the fat content, as it was found that the styrene migration in the full fat milk (3.6% fat) is higher than in the skimmed milk. This study also agreed with another study results²⁵ which concluded that, whenever the fat ratio increases the migration ratio increases also. It was Ramshaw (1984) indicated that the quantity of migrated styrene was higher in the product of high fat contents.²⁶

Table 3 shows the styrene migration in dairy products such as cream, sour cream, yogurt, drinking yogurt, ice-cream, cheddar cheese, and mozzarella reach 0.074±0.17, 0.028±0.05, 0.011±0.22, 0.015±0.13, 0.044±0.07, 0.082±0.06, 0.079±0.11µg/kg respectively. The styrene migrated to the cream was higher with statistically significant ratio of ($P\leq 0.05$) compared to sour cream, yogurt and drinking yogurt due to increment of the fat ratio in the cream (30%) more than in sour cream (6%), yogurt and drinking yogurt (3%). The same trend was shown with butter, cheddar cheese and mozzarella. According to the time

and temperature the styrene migrated to cheddar cheese was higher with statistically significant ratio of ($P\leq 0.05$) compared to cream due to increment of storage time in cheddar cheese (60days) more than in cream (14days). In addition the styrene migrated to whole milk (2h at 100°C) and cream (14days/4°C) with not significant difference. Previous study²⁷ proved that styrene migrated to the high fat cream with a level of 59 parts in billion after 24hours while the lower migration occurred in cheese, sour cream, and yogurt and naturalized milk. Another study showed that the styrene migrated to the cream of (19.5% fat) with a less rate than the increment of the storage period,²⁸ and this agrees with the results of the present study i.e., with increase of fat rate the styrene migration increases. Khansari et al.²⁹ showed that the styrene migrated to full fat milk (3.6% fat) higher than the low fat milk (1.5-2.5 fat%). The study of Flanjak et al.²⁴ showed that the average values of styrene migration of all yogurt samples in New South Wales survey ranged between 0.021 and 0.032mg/kg of the food weight. In other studies the value of monostyrene migration in yogurt ranged from 0.003 to 0.220mg/kg of the sample weight.^{27,28,30} The World Health Organization¹² showed that styrene exists with concentration of 2.5-80µg/kg of the food weight in yogurt and other dairy products in polystyrene packages.

Daily dietary intake of monostyrene of foods packed in polystyrene packages

Table 4 shows the daily intake (µg/kg of the body weight) of monostyrene of studied foods. Dietary intake of monostyrene was calculated by assuming that a 60kg weight person consumed 100g of these foods, thus the consumed quantity of styrene ranged from 0.018 to 0.170µg/kg of the body wt/day i.e. equivalent to 1.1-10.2µg per day (Table 5). The styrene intake of each tested dairy products according to the present results is less than the international allowed level; 40µg/day/person.³¹

Table 4 Monostyrene intake (µ/kg of the body weight/day) of different dairy products packed in PS packages.

Group*	Samples	Monostyrene Quantity (µg/kg of body wt./day)**
1	Whole milk (3.6% fat)	0.118 ^d
	Yogurt (3% fat)	0.018 ^e
2	Sour cream (6% fat)	0.047 ^f
	Cream (30% fat)	0.123 ^d
	Drinking yogurt (3% fat)	0.025 ^e
3	Mozzarella cheese (14.0% fat)	0.131 ^c
	Cheddar cheese (34.4% fat)	0.137 ^b
	Butter (81.7% fat)	0.170 ^a
4	Ice-cream (16% fat)	0.073 ^e

*For group conditions revise Table 1

**Computation based on consumption of 100g daily dietary intake. Average (n=3) carrying similar letters implies that no significant differences exist ($P\leq 0.05$).

Table 5 Daily monostyrene intake ($\mu\text{g}/\text{day}$) per person in diet from different dairy products packed in PS packages

Group*	Samples	Monostyrene quantity ($\mu\text{g}/\text{day}$)**
1	Whole milk (3.6% fat)	7.1 ^d
2	Yogurt (3% fat)	1.1 ^s
	Sour cream (6% fat)	2.8 ^f
	Cream (30% fat)	7.4 ^d
	Drinking yogurt (3% fat)	1.5 ^s
3	Mozzarella cheese (14.0% fat)	7.9 ^e
	Cheddar cheese (34.4% fat)	8.2 ^b
	Butter (81.7% fat)	10.2 ^a
4	Ice-cream (16% fat)	4.4 ^e

*For group conditions revise Table 1

**This limit is computed depending on 60 kg body weight per person. Average (n=3) carrying similar letters implies that no significant differences exist ($P \leq 0.05$).

Compiling more dairy products in a single diet the consumption styrene figure will increase the amount of styrene migrated. We have to say that these results were calculated depending on 100g intake of each food per day; meanwhile the daily styrene intake could be three to four times higher with more frequent food consumption, especially that some of tested foods are dominant in most fast foods restaurants. It is noticeable that dairy products rich in fat content like butter (81.7%) represented the highest risk of styrene intake of the total calculated daily intake.

Addicted consuming of specific food type regular packaged in PS cups could lead to public health hazard. In a study evaluating the consumers exposed to styrene from yogurt for more than 5400 houses in France with a purchasing power of 2 million, it appeared that the rate of dietary intake per person is about $12 \mu\text{g}/\text{day}$ and average of house or family exposure to styrene lies between $1\text{--}35 \mu\text{g}/\text{day}$.¹⁸ The fact that styrene can adversely affect humans in a number of ways raises serious public health and safety questions. One way of styrene exposure is through human daily intake: the assumption, only via food, is estimated to range from 3 to $7 \text{ ng}/\text{kg}$ body weight.³²

Lickly et al.⁹ stated that rate of monostyrene (MS) intake of the daily diets in the USA is $9 \mu\text{g}/\text{day}$, and in UK from 1 to $4 \mu\text{g}/\text{day}$, and this rate is four times less than the daily Research Center (SIRC). FAO and WHO organizations indicated that the maximum permissible intake of styrene in different diets is $40 \mu\text{g}/\text{day}$. This limit is computed depending on 60 kg body weight.³¹ In the present study, based on the assumption of consumption rate and analytical MS data, the calculated daily MS intake could amount to $50.6 \text{ MS} \mu\text{g}/\text{day}$; if all tested dairy products are consumed *per day*, a situation imposes health risk.

Conclusion

In this study, the re-evaluation of styrene monomer in dairy products and Food Contact Materials (FCMs) was necessary due to the recent labelling of styrene as “reasonably anticipated to be a

human carcinogen” by the National Toxicology Panel (NTP). Styrene monomer was selected because of the toxicological properties of this monomer. Monostyrene migrates from the polystyrene packages to different dairy products, and the higher migration has been found in butter, then cheddar cheese, mozzarella and cream. Migration rate increased with the increment of fats rate as in butter (81.7%) and cheddar cheese (34.4%). It is clearly observed that the level of migration depends upon the fat content. Further, the migration of styrene was affected by temperature (whole milk). Quantity of styrene intake from 100g of each tested foods packed in polystyrene package ranged between $(1.1\text{--}10.2) \mu\text{g}/\text{day}$. The consumption rate, food ingredients, and storage conditions are the most crucial factors need to be monitored and controlled to manage the potential health risk of dietary styrene intake.

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Conflict of interest

The author declares no conflict of interest.

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