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Review

Korea Total Diet Study-Based Risk Assessment on Contaminants Formed During Manufacture, Preparation and Storage of Food

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ABSTRACT - Hazardous substances are formed during food manufacturing, processing, or cooking, and may pose a threat to food safety. Here, we present a dietary exposure assessment of Korean consumer's intake of hazardous materials through a Total Diet Study (TDS) which was conducted by the Ministry of Food and Drug Safety (MFDS). The levels of exposure to materials such as acrylamide, furan, biogenic amines, etc., were estimated and risk assessments were then performed. Acrylamide and furan were selected as hazards with high priority of reduction control due to their having a margin of exposure (MOE) lower than 10,000. Risk assessment of exposure to ethyl carbamate, benzene and 3-MCPD showed MOEs higher than 100,000, indicating "safe". Dietary exposure to polycyclic aromatic hydrocarbons and benzopyrene was also found to be safe MOE levels >10,000. In addition, the results indicated safe MOEs (>1,000,000) for heterocyclic amines, nitrosamines, and biogenic amines. Most of the potential food contaminants were being kept at safe levels, however, it is necessary to continue to monitor and control exposure levels in accordance with the 'as low as reasonably achievable' (ALARA) principle.

Key words: Total diet study, Food contaminants, Margin of exposure, Risk assessment

Residual hazardous substances that are formed during food manufacturing, processing, or cooking, may pose a threat to food safety, even in small amounts, as they tend to be ingested for a life-time[1)]. This has heightened anxiety over food safety among the Korean people. Under the existing monitoring system for hazardous substances, the content of a hazardous substance in uncooked food is measured to estimate its exposure dose based on the monitoring results[1)]. This approach fails to capture the true content of a harmful substance accurately because of changes that occur during the processing and cooking, where concentrations can be increased or decreased due to both physical and chemical interactions. For this reason, the alternative risk assessment is practical to determine daily exposure doses more accurately based on a Total Diet Study (TDS), which estimates daily intakes through an analysis of table-ready foods, or an analysis of the content of hazardous substances.

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The TDS has been recognised internationally as the most cost effective way to estimate dietary exposures to food chemicals or nutrients for various population groups and to assess their associated health risks[2)]. The aim of TDS is to provide realistic data on food contamination and chronic exposure levels to chemicals of relevant populations. The TDS approach limits uncertainties in exposure assessment through improved sampling methods and chemical analysis of foods 'as consumed'. TDS generally follows a standard methodology recommended by the World Health Organization (WHO)[2)]. It provides a scientific basis for assessing food safety risks and regulating food supply. Since 1960s, various countries, such as the United States of America (USA), the United Kingdom (UK), Canada and Australia have been conducting their own TDS[3-6)].

In Korea, there was first time TDS on chemicals naturally occurring during cooking and processing like acrylamide, puran, etc. in 2013-2017. The 1st Korea TDS was a large and complex project that comprises sampling and food preparation, laboratory analysis and dietary exposure estimation. It covered the majority of food normally consumed by the Korean population. Accordingly, this review was performed to estimate dietary exposure of Korea population and assess risks on contaminants to be formed during manufacture, preparation and storage of food throughout "TDS project report" conducted by the Ministry of Food and Drug Safety (MFDS)[1]. Also, this study was aimed to seek ways to reduce health risks from those contaminants, and to advise for consumers to have a balanced diet and wise dietary habits.

Materials and Methods

This review study was aimed to present the dietary exposure assessment of Korea population and assess risks on contaminants to be formed during manufacture, preparation and storage of food based on reports published by the Ministry of Food and Drug Safety (MFDS). The main references for this review were “Total Diet Study project report (2013-2017)” and “2016 risk assessment report” conducted by MFDS[1,7]. Accordingly, details of methodology and analytical results including target foods collecting, composite samples preparation and analysis to estimate the dietary exposures and to assess the risks are given in the aforementioned reports[1,7].

Dietary exposure and risk assessment to contaminants

Acrylamide

The formation of acrylamide (AA) in food takes place naturally when carbohydrate-rich, low-protein vegetable foods are cooked at high temperatures. Concentrations of AA in food vary with the type of food, cooking method, water content, and heating time[8]. Acrylamide has been classified as probably carcinogenic (Group 2A) by the International Agency for Research on Cancer (IARC)[9]. AA has shown developmental and reproductive toxicity in animals, as well as neurotoxic effects in humans with occupational exposures. Potato crips, French fries, biscuits, crackers and coffee were reported to contain AA in significant levels ranging on 3 to 1,200 μg/kg in many countries[10]. In a previous study, mean exposure of the Korean adult population to AA has been reported to be 0.076 μg/kg b.w./day resulting in margin of exposure (MOE) of 2,367-4,077 based on the $BMDL_{10}$ (Benchmark Dose Lower Confidence Limit 10%) 0.31 mg/kg b.w./day, set by JECFA, the category of ‘possible concern’ in terms of risk assessment[7].

The daily exposure doses of the average and high consumer (95^{th} percentile) of Korean population to dietary AA are 0.086 and 0.297 μg/kg b.w./day respectively, and the average exposure is less than levels of Canada, France, Hong Kong and Australia (4.3-54.8% levels)[10,11]. The exposure is higher among infants, children, and teenagers under 18 years than in adults over 18 years, taking into account to the fact that confectionary intake is higher among those under 18 years. Among food commodities, the AA contribution from pepper powder is the highest at 18.2%, followed by confectionery (biscuits and cookies) at 10.6%, potatoes at 10.5%, and instant coffee (powder) at 8.1%. The top five “food and cooking method pairs” samples with the highest content of AA were all from pepper powder, cooked with five different methods. Instant coffee (as is), French fries, and potato chips were included in the top 10 list. Among all the pepper powder samples prepared using different cooking methods, the grilled sample showed the highest content of AA, followed by deep-fried, stir-fried, boiled, and pan-fried samples. Results of the TDS-based risk assessment showed that dietary exposure to AA in Korea is at safe levels compared to that in other countries (Table 1)[12]. Their MOEs were all below 4,500, which may indicate “human health concern”. However, as eating habits change, or as the environment changes, exposure levels may exceed the margins of safety.

Furan

Furan is a colorless, highly volatile substance with a boiling point of 31ºC. It is formed from denatured carbohydrates, amino acids, or heated lipids in foods during canning, heat treatments, and cooking[13]. The IARC classified furan as “possibly carcinogenic to humans” (Group 2B). Results of risk assessment indicate that risk from dietary exposure to furan is within the margin of safety. The exposure dose of furan was 0.210 μg/kg b.w./day, and the largest contributors to this exposure were in the following order: wet noodles (7.7%), beer (5.9%), instant jajangmyeon or noodles with black soybean sauce (5.1%), soy beans (4.8%), Makgeolli or rice wine (3.6%), and bread (3.4%). For the hazardous substances assessed, their top 10 contributing foods generally account for 70% or more of the overall exposure in the population; however, in the case of furan, they are detected in a wide variety of foods[14]. According to the 2011 risk reassessment report by the EFSA, dietary exposure to furan is 0.25 μg/kg b.w./day, which is at a similar level to that in this study[2].

Ethyl carbamate (EC)

Ethyl carbamate forms naturally in foods due to chemical reactions during processing and storage, and is present in alcoholic beverages and fermented foods. Examples of alcoholic drinks containing the substance include wine, cheongju (refined rice wine), and whiskey, while fermented foods such as miso (Japanese bean paste), natto (Japanese food made from fermented soybeans), yogurt, cheese, kimchi, and soy sauce are also reported to contain EC. The IARC has classified ethyl carbamate as “probably carcinogenic to humans” (Group 2A). It is absorbed rapidly,

Table 1. Dietary exposure and risk assessment of acrylamide, furan, ethyl carbamate, benzene, 3-MCPD and 1,3-DCP in Korean population[2,17-21)]

Chemicals	Dietary exposure (μg/kg b.w./day)		Oversee exposure (μg/kg b.w./day)	Safety reference (mg/kg b.w./day)
	Mean/95th	MOE		
Acrylamide	0.086/0.297	4,366	0.21 (Hong Kong, 2013) 0.43 (France, 2012) 0.3-1.1(EFSA. 2011)	$BMDL_{10}$, 0.31
Furan	0.210/0.597	4,752	0.234 (USA, 2007) 0.25 (EFSA, 2011)	$BMDL_{10}$, 0.96
Ethyl Carbamate	0.0021/0.0058	>100,000	0.0082 (Hong Kong, 2009) 0.033 (EFSA, 2007)	$BMDL_{10}$, 0.3
Benzene	0.0069/0.028	>100,000	0.020 (Belgium, 2012) 2.4 (UK, 2013)	$BMDL_{10}$, 1.2
3-MCPD*	0.3122/1.4041	>100,000	0.2-0.53 (Hong Kong, 2013)	TDI 2 μg/kg/day (WHO, 2001)
1,3-DCP*	0.0057/0.0244	>100,000	0.012-0.041 (Australia, 2003) 0.00-0.019 (Hong Kong, 2007)	$BMDL_{10}$, 3.3 (JECFA, 2006)

MOE: Margin of exposure.
3-MCPD: 3-Monochloropropane-1,2-diol, 1,3-DCP: 1,3-dichloro-2-propanol.
$BMDL_{10}$:Benchmark Dose Lower Confidence Limit 10%.
TDI: Tolerable daily intakel.

and almost completely, from the gastrointestinal tract and the skin, and more than 90% is eliminated as CO_2, with 2% to 8% excreted in urine[15,16)].

Based on the results of risk assessment for dietary exposure to EC (Table 1), risks posed by the ingestion of food were at safe levels in both the average exposure group and the extreme exposure group. The average daily exposure level of Koreans to EC was 0.0021 μg/kg b.w./day, while the average for the extreme exposure group (P95) was 0.0058 μg/kg b.w./day. Major contributors to ethyl carbamate exposure are soy sauce (61.7%), Maesil ju or green plum liquor (30.9%), and whiskey (5.5%). Ethyl carbamate is usually detected in fruit wines and soy sauces[2)].

Benzene

Benzene is a volatile compound found in very low concentrations in the environment, and reports indicate that it forms from the combination of vitamin C and sodium benzoate used in beverages as a preservative[17)]. In addition to vitamin C, benzene can be also formed in small quantities from beta-carotene, amino acids (phenylalanine, tyrosine, tryptophan), and aromatic flavonoid compounds (pinene, limonene, carene). It can also be created during the manufacturing and processing of heat-treated juices and preserved food products. The IARC has classified benzene as "carcinogenic to humans" (Group 1). In the case of human exposure, 12% of the exposure dose passes out of the respiratory system through the lungs in the form of benzene, while 0.1% is excreted in urine, also remaining in the form of benzene.

The risk of benzene in Korea was assessed against a reference dose of 4 μg/kg b.w./day, which is the health-based guidance values (HBGV) set by the U.S. EPA. Exposure levels of benzene in Korea are lower than those in other countries. The average detection rate for benzene in food was 27.5%. The daily exposure dose per Korean person was 0.0069 μg/kg b.w./day, which is 0.17% and 0.66% (P95) of HBGV of U.S. EPA. MOEs are >100,000. Major contributors to dietary benzene exposure were pork belly (15.7%), mackerel (13.8%), pork (7.5%), and anchovy (dried after boiling, 7.3%).

3-MCPD and 1,3-DCP

3-Monochloropropane-1,2-diol (3-MCPD) and 1,3-dichloro-2-propanol (1,3-DCP) are chemical compounds that belong to the group of chloropropanols, which are created during food manufacturing processes. For instance, these compounds are created by the acid hydrolysis of proteins during the production of soy sauce. The most common chloropropanols, formed from acid-hydrolyzed vegetable proteins, are 3-MCPD, followed by 2-MCPD, 1,3-DCP, and 2,3-DCP. The compounds 3-MCPD and 1,3-DCP are detected in processed foods such as soy sauce, sauces, soups, doughnuts, burger patties, roasted grains, and beer[22)]. IARC has classified 3-MCPD and 1,3-DCP as "possibly carcinogenic to humans" (Group 2B).

Results of the TDS-based risk assessment showed that dietary exposure to 3-MCPD and 1,3-DCP at safe levels within the HBGVs. Average daily exposures of Koreans to 3-MCPD and 1,3-DCP are 0.3122 and 0.0057 μg/kg b.w./day respectively, which are similar to those from Hong Kong, Austria and Brazil (Table 1). The dietary exposure to 3-MCPD is 15.6% of TDI. MOEs are >100,000 based on $BMDL_{10}$, 3.3 mg/kg/day. Major food commodities contributing to dietary exposure were chicken (31.0%), pork (13.0%), processed pork (ham) (11.0%), imported beef (11.0%), and Korean beef (9.8%) for 3-MCPD. The key contributors for 1,3-DCP were red pepper powder (13.0%), squid (10.8%), mackerel (8.7%), and eomuk or fish cake (8.0%).

Polycyclic aromatic hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) are formed during the incomplete combustion of carbohydrates, fats, and proteins present in food when the food is grilled, deep-fried, or stir-fried. Benzo[a]pyrene (B(a)P) is a PAH that is listed as "carcinogenic to humans" (Group 1) by the IARC. When PAHs are orally administrated in rats, they are rapidly absorbed *in vivo* and their bioavailability is between 20% and 50%. Research shows that the half-life of the radioactive isotope B(a)P intravenously administered in rats is less than one minute, which means that it is eliminated quickly from the blood[23].

Results of risk assessment show that dietary exposure to PAHs is maintained at safe levels (0.0001-0.0050 μg/kg b.w./day) and that it is lower than those of the EFSA and other countries. For Korea, the average daily exposure to benzopyrene was estimated at 0.0035 μg/kg b.w/day, with the average exposure to PAH8 at 0.0226 μg/kg b.w/day (Table 2). Both of these exposure levels are similar to, or lower than those for other countries. Their MOEs were at >10,000 levels based on $BMDL_{10}$. Major dietary contributors to PAHs exposure were pork, Samgyeopsal (pork belly), and cabbage kimchi. Benzopyrene, which has the highest toxicity of all PAHs, was found in the highest concentrations in pork (13.0%), followed by beer (9.1%) and pork belly (8.6%). The food with the highest PAH content turned out to be pepper powder, although differences did exist with the cooking method used. The "food and cooking method pair" with the highest B(a)P content was half-dried skipjack tuna (as is).

Heterocyclic amines (HCAs)

Heterocyclic amines (HCAs) are chemicals formed from the reaction of amino acids and creatine, or the pyrolysis of creatine when meat or fish is cooked at temperatures of 200°C or higher. HCAs are divided into two types depending on their chemical structures: (1) amino-carbolines, which are formed in the pyrolytic reaction of pure amino acids, and (2) amino-imidazo-azaarenes, which have an aminoimidazole ring linked to a quinoline, a quinoxaline, or a pyridine[24]. The IARC has classified imidazoquinoline as "probably carcinogenic to humans" (Group 2A) and other HCAs as "possibly carcinogenic to humans" (Group 2B).

Since HCAs are created in meat or fish during high-temperature cooking, target food commodities were cooked at different temperatures for different lengths of time to analyze their HCA content. Although risk posed by HCAs cannot be assessed because there is no HBGV set for HCAs, there is literature available containing data on the determination of PhIP's risk. Results of applying these data to PhIP and the other HCAs indicate that the dietary exposure were at safe levels (MOE >1,000,000). While major HCAs (IQ and MeIQ) were not detected, exposure doses of other HCAs were 0.315 ng/kg b.w./day for PhIP , 0.188 ng/kg b.w./day for A-α-C, and 150.376 ng/kg b.w./day for harman. HCA exposure levels for Korea are 10 or more times less than

Table 2. Dietary exposure and risk assessment of polycyclic aromatic hydrocarbons

Chemicals	Dietary exposure (μg/kg b.w./day)		Oversee exposure (μg/kg b.w./day)	Safety reference (mg/kg bw/day)
	Mean	MOE		
Benzopyene	0.0035	>10,000	0.001(Spain, 2008) 0.004 (EFSA, 2008)	$BMDL_{10}$, 0.1 (JECFA, 2010)
PAH2 (BaP, Chry)	0.0069	>10,000	0.0009 (France, 2013) 0.0107 (EFSA, 2008)	$BMDL_{10}$, 0.17 (EFSA, 2008)
PAH4 (BaA, BaP, BbF, Chry)	0.0147	>10,000	0.0015 (France, 2013) 0.0195 (EFSA, 2008)	$BMDL_{10}$, 0.34 (EFSA, 2008)
PAH8 (BaA, BaP, BbF, Chry, BkF, DahA, BghiP, IcdP)	0.0226	>10,000	0.0288 (EFSA, 2008) 0.0022 (France, 2013)	$BMDL_{10}$, 0.49 (EFSA, 2008)

BaA; benzo[a]anthracene, Chry; chrysene, BbF; benzo[b]fluoranthene; BkF; benzo(k)fluoranthene; 3MC; 3-methylcholanthrene, BaP; Benzo[a]pyrene, DahA; dibenzo[a,h]anthracene, BghiP; benzo(ghi)perylene; IcdP; indeno[1,2,3-cd]pyrene.
MOE: margin of exposure.
PAHs: Polycyclic aromatic hydrocarbons.
$BMDL_{10}$: Benchmark Dose Lower Confidence Limit 10%.

those for other countries. Contributing foods to exposure were pork belly (samgyeopsal), pork, beef, cutlassfish, etc.

Biogenic amines (BAs)

Biogenic amines (BAs) are nitrogenous compounds formed mainly by the decarboxylation of amino acids or by the amination and transamination of aldehydes and ketones during the metabolic processes of microbes, plants, and animals, and are naturally found in live cells. Biogenic amines are found in fermented soybean products, kimchi, fermented livestock products, and fish. Histamine, the most well-known biogenic amine, is used as an indicator of hygiene to measure the extent of decomposition in fish[25,26]. The Codex standard for histamine levels in fish and processed fish products is set at less than 100 mg/kg. Putrescine, spermidine, tyramine, tryptamine, and cadaverine can react with nitrites in food to form a carcinogen called nitrosamine. The diverse metabolites of histamine have little pharmacological activation, and most of them are passed out of the body in urine.

There is no HBGV set for BAs. For this reason, the toxicity benchmark value of $BMDL_{10}$ 36,920 μg/kg b.w./day, proposed by the Codex for histamine, was also applied here to estimate the MOE for BAs. The results of the assessment showed that dietary exposure to BAs and histamine were 0.047-0.287 and 0.140 mg/kg/day, respectively, being maintained at safe levels, with findings similar to, or lower, than those of Japan, Australia, and EFSA[10,27]. Contributors to BAs exposure include soju, beer, canned tuna, Ramyeon (instant noodle), Makgeolli (rice wine), kimchi (napa cabbage kimchi), soybeans, and garlics. Contributors to histamine for which a standard exposure level is set include white rice at 21.5%, kimchi at 20.3%, and Ssamjang (mixed paste) at 13.5%. The food and cooking method pairs with the highest histamine content is pepper powder (boiled), followed by pepper powder (prepared by grilling and frying) and Aekjot (liquid salted and fermented seafood) made of fish and shellfish (as is/pan-fried/boiled).

Nitrosamines (NAs)

Nitrosamines occur due to the reaction of secondary amines and nitrites under acidic conditions. They may be formed *in vivo* from precursors following the ingestion of foodstuffs, and bacteria in the gastrointestinal tract contribute to the formation of nitrosamines from nitrites and amines. Nitrosamines are listed as “probably carcinogenic to humans” (Group 2A) and “possibly carcinogenic to humans” (Group 2B) by the IARC. Studies have shown that, when administered orally, nitrosamines are quickly absorbed in the gastrointestinal tract, and their metabolites exhibit carcinogenicity in hamsters and rats[28].

The dietary exposure to total nitrosamines and N-nitrosodimethylamine (one of nitrosoamines) were 0.0189, 0.0116 μg/kg b.w./day, respectively, which is about 81.1% of the level for the German population. Major food commodities contributing to nitrosamine exposure include kimchi, potatoes, Doenjang (soybean paste), pepper powder, processed pork (ham), and confectionery (snack confectionery). There have been few studies to assess dietary exposure to nitrosamines. Since there is no HBGV for nitrosamines, it is necessary to observe trends in dietary exposure levels through continuous monitoring.

Prevention and risk reduction

Acrylamide increases rapidly at 160ºC or higher temperatures. Therefore, as a method of reducing the AA formation during cooking, it is recommended to cook at lower temperatures (160ºC or lower for frying and 200ºC or lower for oven cooking). You can further reduce the formation of AA by choosing steaming or boiling over grilling as your cooking method. Adding or sprinkling pepper to season a food before stir-frying or deep-frying increases the AA content in the food; therefore, it is advisable to add pepper to taste after cooking is complete[29]. Also, AA formation can be reduced if potatoes are soaked in water at 60ºC for 45 min, and using less sugar during the cooking process will help reduce the formation of AA.

Furan is highly volatile, therefore, after opening a processed food that has been hermetically sealed, it is recommended that it remain open for some time before it is consumed to allow the furan to evaporate. A baby drink also should not be ingested immediately after purchase. Instead, it should be kept in the refrigerator, with the lid open, for at least one day before drinking. Meat products should be heated at 50ºC~70ºC in the microwave, or using other methods, before eating. In the case of instant coffee or brewed coffee, it is advisable to let it “breathe” with the lid open for one to five minutes to allow furan to evaporate before drinking, rather than drinking it immediately after adding hot water.

In Europe, ethyl carbamate exposure levels were about four times higher for drinkers than non-drinkers. Considering the Korean diet, with relatively high intakes of fermented foods such as kimchis, fermented seafoods, and alcoholic drinks, the possibility of human health risk due to diet-derived EC cannot be ruled out. As mentioned previously, benzene can be formed in small quantities when both vitamin C and sodium benzoate are present in beverages. For this reason, in 2006, the food and beverage industry was urged by the MFDS to use an alternative preservative other than sodium benzoate or replacing it with a natural preservative.

Methods for reducing dietary exposure to 3-MCPD and

1,3-DCP include adjusting production processing conditions (e.g., the acid density used for acid-hydrolyzed soy sauce), neutralization conditions, and reaction times. At home, you are advised to cook at lower temperatures and to cut down on edible oils and fats. In order to reduce the formation of PAHs and HCAs, in cooked meat, boiling is a better cooking method than roasting or grilling, and it is recommended to cook food items over medium heat (150ºC~160ºC), and not to bring the meat into direct contact with the fire over a grate. When meat is grilled over charcoals, the smoke will contain PAHs and HCAs, and should not be inhaled. Also, the meat should not be burned; any burned parts should not be consumed.

It is also advisable to reference the following when cooking: marinating meat or fish in salt or garlic helps reduce HCA formation. Moreover, sulfur compounds (from garlic and onions) and antioxidants (e.g., anthocyanin in the red wine and catechin in the tea) inhibit the formation of HCAs. To reduce the formation of BAs, the MFDS recommends (1) lowering fermentation and storage temperatures, (2) selecting high-quality strains for fermentation, and adding additives to the processing steps during manufacturing.

Currently, most of all contaminants are controlled at safe levels, but it is necessary to continue to monitor and control exposure levels according to the as low as reasonably achievable (ALARA) principle.

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국문요약

식품의 제조, 가공, 조리 및 저장 중 많은 유해물질이 발생하며 이들은 소량이지만 장기간 노출되면 식품안전에 위협이 될 수 있다. 본 연구에서는 식품의약품안전처에서 수행한 총 식이조사(TDS) 자료를 기반으로 우리나라에서 식품섭취를 통한 이들 주요 유해물질에 대한 노출 및 위해평가 상황을 파악하고 소비자 및 기업으로 하여금 관련 위험을 저감하는 방안을 제시하고자 하는 것이다. 식품의 제조, 가공, 조리 및 저장 중 생성 유해물질 중 대표적인 아크릴아미드, 퓨란, 에틸 카바메이트, 3-MCPD, 바이오제닉 아민류, 니트로아민류 화합물, 다환 방향족화합물, 벤젠 등에 대한 식이를 통한 노출량을 조사하고 위해평가를 수행한 바, 아크릴아미드 및 퓨란의 경우 노출안전역(MOE) 10,000이하로 저감화 우선 물질로 판단되며, 나머지 물질 등의 경우 모두 노출안전역이 10,000 또는 100,000 이상으로 안전한 수준에 있는 것으로 나타났음. 그러나 향후 지속적으로 모니터링을 수행하고 ALARA 원칙에 따라 가능한 노출 저감화를 위해 노력을 기울여야한다.

Conflict of interests

The authors declare no potential conflict of interest.

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