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The Occurrence and Toxicity of Dioxins and Dioxin-Like Polychlorinated Biphenyls in Foodstuffs Collected From Different Cities of Iran: A Systematic Review

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ABSTRACT

Dioxins compounds are persistent organic contaminants that adversely affect human health and the environmental system. Although the occurrence and toxicity of dioxins congeners are reported worldwide, their status in Iran is very scarce, and no study has been conducted to understand the evidence for occurrence and toxicity of dioxins compounds in Iran yet. Therefore, this systematic review provided a comprehensive report on the levels, occurrence, and health outcome effects of dioxins and dioxin-like Polychlorinated biphenyls (DL-PCBs) in foodstuffs collected from different cities of Iran. We used The Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) to design this review, and six databases (Scopus, Scientific Information Database (SID), PubMed, Web of Knowledge, Scopus, and Google Scholar) were searched from inception until January 2022. A total of 9 articles with data on 396 food samples were considered for the final report. The total toxicity equivalency quantity (TEQ) was ranged from 0.06 ± 0.02 to 15.72 ±16.38 and the mean total dioxin congeners levels in most of the samples were below the standard limits and the measured intake levels of dioxin congeners were in the safe level. Although most of the samples did not exceed the maximum allowable thresholds, 44% of studies reported that the concentration of dioxin compounds in most of their samples was above the thresholds defined by world health organization (WHO) (3 pg TEQ/g fat). Therefore, suitable policy actions and more attention are needed to control and reduce the emission of dioxin congeners and their associated risk factors.

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Introduction

Dioxins and dioxin-like Polychlorinated biphenyls (DL-PCBs) are the main groups of persistent organic pollutants (POPs) including 135 polychlorinated dibenzofurans (PCDF), 75 polychlorinated dibenzo-p-dioxins (PCDDs), and 12 polychlorinated biphenyls congeners, of which 30 have significant toxicity effect on human health [1, 2]. These compounds have a long half-life in the different media environments, and dioxins are highly persistent because of their high lipophilicity[3, 4]. Dioxins compounds are mainly generated by various anthropogenic activities such as industrial processes (manufacturing of chlorine bleaching of paper pulp, smelting, pesticides, and herbicides), waste incinerators (hospital and solid waste). Likewise, small quantities of dioxins can be generated by natural processes, including volcanic eruptions and forest fires [3, 5].

Decreasing dioxin exposure is the main public health target for reducing the type of disease. World Health Organization's International Agency for Research on Cancer (IARC) classified dioxin as a "known human carcinogen" according to human epidemiology data and animal data[4]. Long-term exposure of humans to the high concentration of dioxin is associated with an impairment of the endocrine system, nervous system, immune system, and reproductive functions [5]. Short-term exposure may be linked to skin lesions and altered liver function. Dioxin compounds formation is local, but their distribution in the world is global. The highest concentration of dioxins is found in the sediment, soil, and food supply (fish, meat, and shellfish), especially dairy products, while the lowest concentration is found in air, water, and plants [4, 6]. Several developed countries monitor the level of dioxin compounds in their food supply because early detection of pollutants led to preventing adverse effects on a larger scale [1, 7, 8]. It was evidenced that more than 90% of the exposure of humans to dioxins is through the

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food supply, mainly seafood, dairy products, and meat[6, 9]. Thus, it is critical to protect the food supply to reduce dioxin emissions. Appropriate practices and controls during the production of food supply, their processing, sale, and distribution are all necessary to produce safe food [3, 6].

In low- and middle-income countries like Iran, the available data related to the occurrence and toxicity of dioxins is very scarce yet, and we are not aware of any systematic research on the evidence for occurrence and toxicity of dioxins and DL-PCBs in the food supply[3, 10]. In Iran, several studies monitored the occurrence and toxicity of dioxins and DL-PCBs in environmental samples and their risk that influence human health. However, these studies are disconnected because no general view and systematic investigation on the situation of dioxins and DL-PCBs pollution in Iran. In consequence of adverse health effects posed by dioxins and DL-PCBs pollutants in the food supply, examining the exact concentration of these compounds and their possible health risks is essential to reduce their emission into the food supply and food chain, especially, seafood, meat, and, dairy products [3, 8]. Therefore, conducting a systematic review with the target to provide comprehensive data on dioxins and DL-PCBs' pollution levels and their distribution could be practical to improve food safety programs. This review was performed as a comprehensive database on the levels, occurrence, and health outcome effects of dioxins compounds in foodstuffs collected from different cities of Iran.

Materials and Methods

Study Design

The Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) was used as a framework to design this study[11]. The main outcome for this systematic review is the occurrence and concentration of dioxins (PCDF and PCDDs) congeners and DL-PCBs compounds in food supply (Table 1) and the level of health risk originating from the effects of dioxins compound. The main question in this study is what is the dioxin level and status in foodstuffs collected from Iranian market?

Search Strategy

We searched five databases were searched, including the scientific information Database (SID), PubMed, Web of Knowledge, Scopus, and Google Scholar were searched from inception until January 2022, from inception until January 2022. Each database was independently searched, and the references list of all studies was hand-searched to find relevant literature, which may be ignored during the search process. The main keywords related to the term "dioxins", "DL-PCBs", and "dioxin-like Polychlorinated biphenyls" were used. Then, these keywords were combined with the terms "foodstuffs", "meat", "dairy products", "fish", "meat", "vegetable", "health risk", and "Iran.

Table 1. Congener's distribution of dioxins, furans and dioxin-like PCBs

Congener	WHO_{2005}-TEF	Congener	*WHO_{2005}-TEF
Polychlorinated dioxins **(PCDDs)**		**Non-*ortho*** polychlorinated biphenyls (PCBs)	
2,3,7,8-TCDD	1	PCB-77	0.0001
1,2,3,7,8-PeCDD	1	PCB-81	0.0003
1,2,3,4,7,8-HxCDD	0.1	PCB-126	0.1
1,2,3,6,7,8-HxCDD	0.1	PCB-169	0.03
1,2,3,7,8,9-HxCDD	0.1	**Mono-*ortho*** polychlorinated biphenyls (PCBs)	
1,2,3,4,6,7,8-HpCDD	0.01	PCB-105	0.00003
1,2,3,4,6,7,8,9-OCDD	0.0003	PCB-114	0.00003
Polychlorinated dibenzofurans **(PCDFs)**		PCB-118	0.00003
2,3,7,8-TCDF	0.1	PCB-123	0.00003
1,2,3,7,8-PeCDF	0.03	PCB-156	0.00003
2,3,4,7,8-PeCDF	0.3	PCB-157	0.00003
1,2,3,4,7,8-HxCDF	0.1	PCB-167	0.00003
1,2,3,6,7,8-HxCDF	0.1	PCB-189	0.00003
2,3,4,6,7,8-HxCDF	0.1	PCB-180	0.00003
1,2,3,7,8,9-HxCDF	0.1		
1,2,3,4,6,7,8-HpCDF	0.01		
1,2,3,4,7,8,9-HpCDF	0.01		
1,2,3,4,6,7,8,9-OCD	0.0003		

*Toxic equivalency factors (TEF) established by WHO in 2005

Inclusion and Exclusion Criteria

In this systematic review, studies were included conducted on detecting the concentration of dioxin compounds in foodstuffs of Iran, publishing in the Persian or English language, reporting average concentration of dioxins and their health risk level, and focused the exposure to dioxins compounds and their adverse effects on human health in Iran. The eligibility of studies and their selection were conducted by monitoring the abstracts, titles, and full texts of manuscripts. All duplicate studies were excluded. There was no time limit in this review.

Data extraction and quality assessment sources

The relevant data were extracted based on the publication author and year, study aim, sample size, study location, time of sampling, analytical technique, type(s) of samples, type of dioxin concentration, and potential contamination. In this study, was used to examine the quality of the selected studies[12]. This checklist includes 6 questions that were designed based on a "yes" or "no" response to estimate a general quality score for each study (Table 2).

Table 2. Quality assessments of the included studies based on Newcastle–Ottawa scale

Code	Authors (years)	Standard sampling protocol indicated	Period of sampling indicated	QA/QC conducted	All objectives achieved	Report on Dioxin sources	Report on mean Dioxin levels	Overall quality score
1	Zeynab Samadi Jirdeh, 2013[17]	No	Yes	Yes	Yes	No	Yes	4/6
2	Mohsen Rezaei, et al, 2012[18]	Yes	Yes	Yes	Yes	No	Yes	5/6
3	Reza Ahmadkhaniha et al, 2017[13]	Yes	Yes	Yes	Yes	No	Yes	5/6
4	Zeinab Samadi Jirdeh et al, 2013[18]	No	Yes	Yes	Yes	No	Yes	4/6
5	Azadeh Nakisa et al, 2012[19]	Yes	Yes	Yes	Yes	No	Yes	5/6
6	Ayub Ebadi Fathabad et al , 2019[20]	Yes	No	Yes	Yes	No	No	3/6
7	Seyede Pegah Azarchehry et al, 2021[14]	Yes	Yes	Yes	Yes	Yes	Yes	6/6
8	Sara Bayat et al., 2011[15]	Yes	No	Yes	Yes	No	Yes	4/6
9	Shakoorzadeh, A. et al., 2017[16]	No	Yes	No	Yes	Yes	Yes	4/6

Results

Search Outcome and Quality Assessment

In this study, we selected 47 published articles for quality assessment from which 9 articles with data on 396 food samples were considered for the final report because these articles met inclusion criteria (Figure 1, Table 2). All data were extracted from 7 different provinces of Iran as follows: Tehran (n= 4 studies)[13-16], Qazvin (n= 2)[17], Khuzestan Province (n=2)[18, 19], provinces of North of Iran (n=2)[14, 20], and provinces of Central Iran (n=1)[14]. Our results showed that the highest number of studies was conducted in Tehran (capital of Iran) [13-16] (Table 3). According to the quality assessment information, 44% of the eligible studies (n=4) were in good-quality categories and rated from 5 to 6 out of 6[13, 14, 18, 19]. These studies used valid protocol and methods for sampling and measurement of dioxin congeners provided reliable results and all objectives were achieved in their studies. Likewise, 44% of the eligible studies (n=4) were in moderate-quality categories with an average score of 4[15-17], and about 12% of the studies showed low quality (from 2 to 3) [20] because non-indication of average concentration of dioxin compounds, as well as period of sampling and, were not clear in their researches. The first studies related to evaluating dioxin in foodstuffs were published in 2011[15] (Table 2).

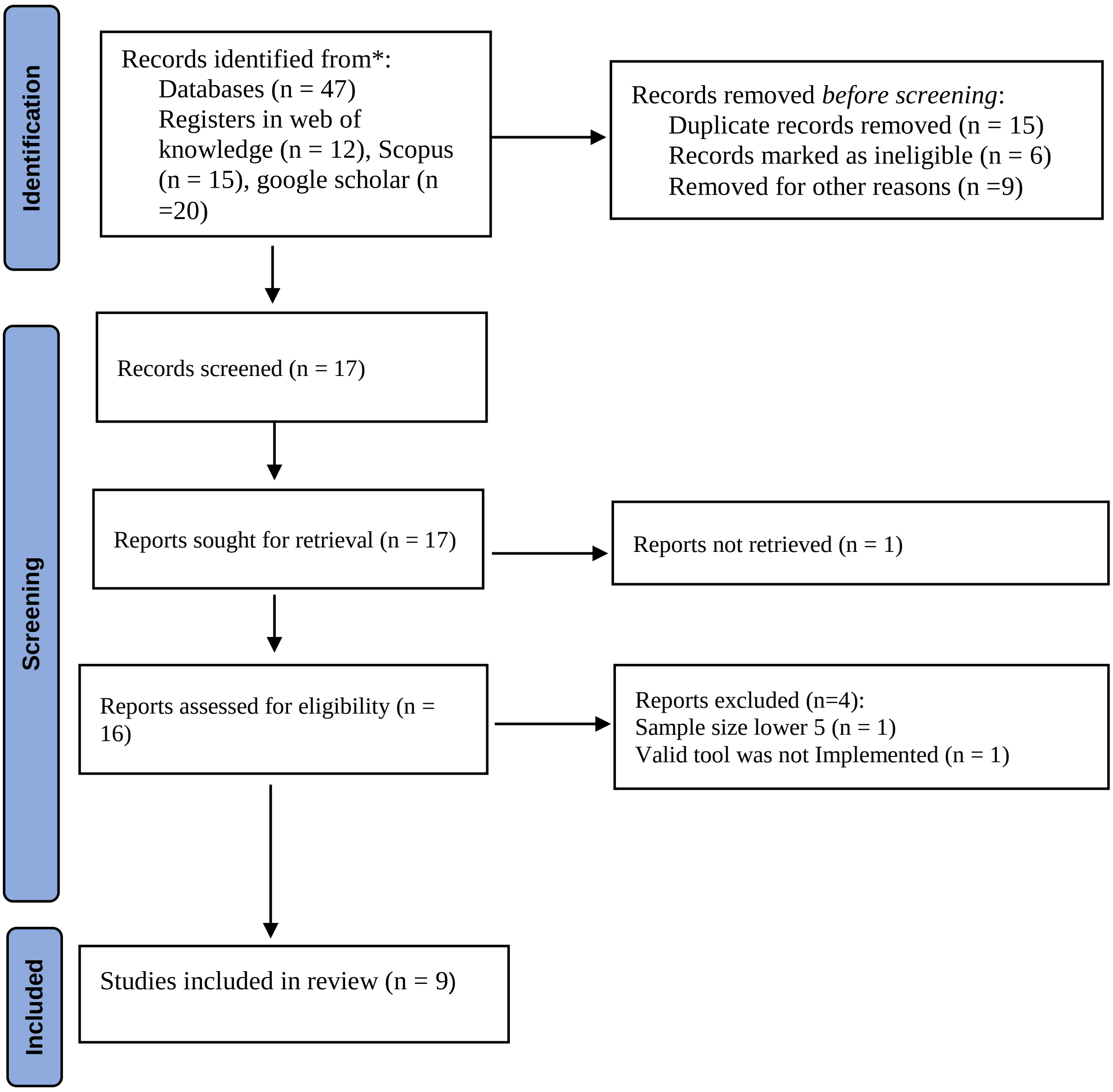

Figure 1. PRISMA flowchart for studies selection

Study Samples Characteristics

Of 396 food samples included studies, 245 (62%) samples included raw or fresh milk (28%) [14, 17, 19] and pasteurized milk (34%) [13-15, 17, 18], 125 (32 %) samples included fishes of Caspian Sea [20], and 26 (6%) samples included pinto beans [16](Table 3). Most of the studies (67%) have measured Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) as dioxin compounds[13, 14, 16-19] and 5 studies examined DL- PCBs[13-15, 17, 18, 20]. In this review, 6 (67 %) studies measured dioxin compounds using Gas Chromatography-mass spectrometry (GC–MS) or by High-Resolution Gas Chromatography coupled with High-Resolution Mass Spectrometer (HRGC/HRMS)[13, 14, 16, 17, 20] . Likewise, 3 studies measured dioxin compounds by using liquid chromatography (HPLC)[15, 17-19]. Based on our findings, HRGC/HRMS is the most common valid method, which was recently implemented to measure dioxin congeners in Iran, while other methods were rarely used.

Table 3. Characteristics of included studies

Code	Study area	Sample/ sample size	Analytical Methods	Concentration (pg TEQ/g fat) PCDD/F	DL-PCBs	Total TEQ	Outcomes
1	Qazvin	Row milk (n=19)	GC-MS/ Kawashiro *Methods*	15.3 ±16.18 (2.6–73.7)	0.40± 0.304 (0-0.92)	15.72 ±16.38 (2.5–74.57)	Mean of total TEQ is above the thresholds defined by WHO/2,3,7,8 -TCDD, 1,2,3,7,8 –PeCDD and 1,2,3,4,7,8 –HxCDD were the dominant congeners
2	Khuzestan Province	Pasteurized milk (n=45)	HPLC/ A liquid-liquid extraction	4.02 ±0.21 (0.59-3.17)		4.02 ±0.21	Concentrations of dioxins in summer were more than other seasons/2,3,7,8 -TCDD and 1,2,3,4,7,8 –HxCDD were the dominant congeners/Mean of total TEQ is above the thresholds in all samples.
3	Local market in Tehran	10 brand Pasteurized milk (n=20)	GC-MS-Ms		0.49 ± 0.12	0.49 ± 0.12	Mono-ortho congeners were more abundant than the non-ortho ones and congener 118 and 180 was the dominant chemicals (over 80%)/Mean of total TEQ is lower the thresholds in all samples/ There is no significant difference between seasons /Non-carcinogenic risk was in safe level (HQ < 1).
4	Qazvin	Pasteurized full-fat milk (n=7)	*HPLC/ HRMS	0.74±0.30 (0.34-1.10);	0.137±0.040	0.88±0.34 (0.36-1.14)	Mean of total TEQ was lower than the thresholds/ 2,3,7,8-TCDD and 1,2,3,4,7,8-HxCDD were dominant
5	Khuzestan Province 2012	Fresh milk (n= 60)	HPLC/ Kawashiro	1.77 (0.35 - 3.03)		1.77 (0.35 - 3.03)	The mean total content was below the recommended threshold /2,3,7,8-TCDD and 1,2,3,4,7,8-HxCDD were dominant /There were statistically significant between differences seasons with highest concentration in summer and the lowest in winter.
6	Caspian sea	Fish (n=125)	HRGC/HRMS/ USEPA 1668		0.59 – 0.82	0.59 – 0.82	The mean total content was below the recommended threshold/ Mono-ortho congeners were more abundant and congener 118 and 180 was the dominant chemicals (over 80%)/Non-carcinogenic risk was in safe level (HQ < 1)/ Cancer risk exceeded the value of $1×10^{-6}$
7	Central Iran	Raw milk (n= 12)	HRGC_HRMS/EC/252/2012	2.42 ± 0.97 0.63-4.08		2.42 ± 0.97 0.63-4.08	The mean concentration is higher in some samples
7	North of Iran	Raw milk (n= 9)	HRGC_HRMS/EC/252/2012	1.13 ± 0.4 0.63-1.89		1.13 ± 0.4 0.63-1.89	The mean total content was below the recommended threshold in all samples/2,3,7,8-TCDD and 1,2,3,4,7,8-HxCDD were the dominant chemicals
7	Tehran	Pasteurized milk (n= 15)	HRGC_HRMS/EC/252/2012	0.06 ± 0.02 0.03 to 0.1		0.06 ± 0.02 0.03 to 0.1	The mean total content was below the recommended threshold in all samples
8	Tehran	Pasteurize milk (n=58)	AOAC method		0.75± 0.24	0.75± 0.24	The mean total content was below the recommended threshold in all samples
9	Tehran	Pinto beans (n= 26)	GC/ MRLS METHOD	2.13 ± 0.02		2.13 ± 0.02 0.62 to 3.67	Samples are cultivated near factories and are exposed to their wastewater were made toxic by dioxins; while dioxins was not observed in the other samples, which were located far from factories

PCDD/Fs: Polychlorinated dibenzo-p-dioxins and dibenzofurans; and DL- PCBs: dioxin-like polychlorinated biphenyls; TEQ: Toxic equivalent/ HRGC/HRMS: High Resolution Gas Chromatography coupled with High Resolution Mass Spectrometer; *the national standard of Iran with no. 8262/2005; HPLC: High Performance Liquid Chromatography.

Likewise, 3 studies measured dioxin compounds by using liquid chromatography (HPLC)[15, 17-19]. Based on our findings, HRGC/HRMS is the most common valid method, which was recently implemented to measure dioxin congeners in Iran, while other methods were rarely used. Three (33%) and two studies (22%) extracted dioxin compounds using the method of Commission Regulation (EU) No 252/2012, and Kawashiro et al., 2008 method, respectively [13, 14, 16, 17, 20]. Other studies used different protocols for the extraction process [15, 17-19]. Our finding showed that 44% of the surveys were performed in Tehran[13-16] and the sample size was ranged from 7 to 125 (Table 3).

Dioxin Concentration in Samples

According to our findings in this review, the concentration of PCDD/F and DL-PCBs ranged from 0.06 ± 0.02 to 15.3 ±16.18 and from 0.137±0.040 to 0.75± 0.24 pg TEQ/g fat, respectively. The total TEQ was also ranged from 0.06 ± 0.02 to 15.72 ±16.38. Of the 9 studies that measured concentration of dioxin in foodstuffs collected from different cities of Iran, 4 studies (44%) reported that the concentration of dioxin compounds in most of their samples were above the thresholds defined by WHO (3 pg TEQ/g fat)[15-18]. Most implicated dioxin included 2,3,7,8 -TCDD, 1,2,3,7,8 –PeCDD and 1,2,3,4,7,8 –HxCDD were the dominant congeners[14, 17-19] (Table 3).

Risk Assessment of Dioxin Congeners

In this systematic review, only two studies examined carcinogenic and non-carcinogenic risks (CR) in foodstuffs samples[13, 20]. This study showed reported acceptable or negligible non-carcinogenic risk or hazard quotient (HQ < 1) through consumption of fish in Caspian Sea sample and pasteurized milk. However, carcinogenic risk exceeded the threshold value of 1×10-6 in fish samples (Table 3).

Discussion

The primary aim of this systematic review was to evaluate the levels, occurrence, and health outcome effects of dioxins and DL-PCBs in foodstuffs collected from different cities of Iran. In fact, we tried to conduct systematic research to verify whether the level of dioxins and DL-PCBs in the foodstuffs samples comply with WHO and European Union (EU) maximum allowable thresholds for milk (3 pg TEQ/g fat) and other foodstuffs (0.492 pg/g of fat)[2, 4]. This is the main question that must be replied to before deciding which measures are practical to reduce and control dioxins congeners in the food chain and environment.

In the present study, 9 published articles met inclusion criteria identified that measured dioxins and DL-PCBs in foodstuffs marketed Iran; among them, only two studies estimated the carcinogenic and non-carcinogenic risk effect of dioxins and DL-PCBs exposure on human health [13, 20]. Our finding showed that dioxins and DL-PCBs concentration in most of foodstuffs samples (56%) were below the thresholds defined by WHO and EC legislation [13-15, 17, 18, 20]. These studies examined dioxin congeners in pasteurized milk and fresh milk, and fish from 2011 to 2019. Overall dioxins level in these samples ranged from 0.06 to 0.88 pg TEQ/g fat. Therefore, it seems that the consumption of these samples which are commercially available in Iranian markets is not a threat for consumers in Iran. The mean of the sum of PCDD/F and DL-PCBs in milk products (n= 5640) which were collected in different European countries ranged from 0.07 to 5.68 pg TEQ/g fat[2]. According to our findings, the concentration of PCDD/F and DL-PCBs in most of the foodstuffs samples in Iran is significantly lower than the EU samples[2, 4]. Likewise, contamination levels observed in this review are similar to those reported by some other researchers: 0.65 in Catalonia, 0.43 in the United Kingdom, 0.31 in Sweden, 0.18 in Greece, and 0.16 pg/g of fat in Finland [1, 2, 13].

Although most of the samples did not exceed the maximum allowable thresholds provided by WHO and EU Regulation, 4 studies (44%) on pasteurized milk, raw milk, and pinto beans reported that the concentration of total dioxin compounds in most of their samples were above the standard thresholds for milk [15-18]. Dioxin levels were ranged from 2.42 to 15.72 pg TEQ/g fat. The high concentration of dioxin congeners in these samples may occur via different pathways. The main pathways of dioxin compounds are related to animal feed and emission of industrial wastewater in environmental media (air, soil, and water)[7, 14]. All of these studies on milk were conducted in industrial cities in Iran (Qazvin and Khuzestan) where the main oil gas and petrochemical industries were developed[17, 18]. Likewise, other industries related to the production of ferrous and nonferrous metal, mineral products, open burning processes, and chemicals are more abundant in these cities compared to other cities in Iran [5, 18]. It was evident that from 1990 to 2010, ferrous/nonferrous metal production and open burning processes significantly contributed to 24.4% to 32.2% and 45.8% to 35.7% of the total PCDD/PCDFs emissions into the environment of Iran[5]. Likewise, the study on pinto beans samples showed that samples are cultivated in farms where located near factories were significantly toxic by dioxins while dioxins were not observed in the other samples, which were located far from factories[16]. Therefore, the observations confirm that industrial wastewater and animal feed have a significant contribution to food contamination. This result is in agreement with findings from other studies on dioxin compounds that were conducted in other countries such as China, the Brazilian citrus pulp, and the Belgian dioxin crisis, which show, small industries are able to cause severe health consequences if a high concentration of pollutants in foodstuff is detected quite late in processed food of animal origin on the market[1, 2]. Considering the high amount of this product on the national or international market, it is essential to pay more attention to the contamination of dioxin compounds in feed, particularly in industrial cities of Iran.

Health risk assessment examines the effect of hazard, risk level, and exposure, according to the potential toxicity of dioxin pollutants, the severity of exposure, and risk characterization[10]. Based on our results on risk evaluation for dioxin compounds, 2 studies reported safe risk levels via consumption of pasteurized milk and fish in the Caspian Sea[20]. However, studies on dioxin risk and its effect on human health are limited in Iran; therefore, conducting more studies is important to better understand a potential health risk related to dioxin contamination in foods products.

The highest contribution to the total concentration of dioxin compounds is from 2,3,7,8-TCDD , 1,2,3,4,7,8-HxCDD, and 1,2,3,7,8 –PeCDD [14, 17-19]. Among DL-PCBs, Mono-ortho congeners were more abundant than the non-ortho ones and congener 118 and 180 showed the highest concentration (over 80%)[13, 20]. A similar dioxin profile was observed in dairy products examined in Italy [1, 8]. Likewise, several studies on food samples reported that the mono-ortho congeners were more abundant than the non-ortho congeners in dairy products[2, 9]. This same profile of the dioxin pattern could be due to the same sources of emissions into the environment and food chain including the presence of similar factories and industries in different countries and the use of the same source of crop and silage for cow feeding [2, 9, 20].

In addition, seasonal variations were reported for dioxin levels in tested samples with the highest concentration in summer and the lowest in winter[18, 19]. However, one study reported an insignificant difference between seasons[13]. These differences between seasons could be due to differences in climate conditions (atmospheric deposition, dust storm, and dust particles), air pollution conditions, the status of soil contamination, and the amount of fertilizers that are used in agricultural activities[6, 12]. However, seasonal trends need repeated tests throughout the year.

Conclusion

This present study tried for the first time systematically reviewed the levels, occurrence, and health outcome effects of dioxins and DL-PCBs in the Iranian market, which is important to better understand the situation of dioxin pollution in a national context. This present review showed that the mean total dioxin congeners (PCDD/F and DL-PCBs) levels in pasteurized milk and fresh milk, and fish in most of the samples were below the standard limits defined by the WHO or EU legislation and the measured intake levels of PCDD/F and DL-PCBs were in the safe level. However, our findings highlighted samples were collected from industrial cities of Iran are significantly toxic by dioxin congeners because the concentration of total dioxin compounds in their samples was above the standard limitation. Considering the existence of several sources for dioxin pollution, their adverse effects on human health and the ecosystem, and their bio-accumulative properties, suitable policy actions, and more attention are needed to control and reduce the emission of dioxin congeners and their associated risk factors. Likewise, we recommended continuous monitoring of dioxin compounds in foodstuffs and environment media especially in industrial reigns in Iran. This residual monitoring is useful in controlling emission points for ensuring the safety of food. Further studies on other foodstuff-based repeated analyses in different seasons are suggested to find out the spatial and temporal emission trends of dioxin congeners.

Abbreviations

Dioxin-like Polychlorinated biphenyls (DL-PCBs); Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA); Scientific Information Database (SID); toxicity equivalency quantity (TEQ); world health organization (WHO); persistent organic pollutants (POPs); polychlorinated dibenzofurans (PCDF); polychlorinated dibenzo-p-dioxins (PCDDs); Cancer Risk (CR); hazard quotient (HQ); European Union (EU).

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Conflict of Interest

There is no conflicts of interest in this work.

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