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RESEARCH ARTICLE

Toxic heavy metals and nutrient concentration in the milk of goat herds in two Iranian industrial and non-industrial zones

Nasrin Homayonibezi[1] · Sina Dobaradaran[1,2,3] · Hossein Arfaeinia[1,2] · Marzieh Mahmoodi[4] · Ali Mohammad Sanati[5] · Mohammad Reza Farzaneh[6] · Raheleh Kafaei[7] · Maryam Afsari[1] · Moradali Fouladvand[3] · Bahman Ramavandi[1,2]

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Abstract
This work aimed to explore the concentration of nickel, manganese, iron, copper, chromium, and lead in the milk of goat herds in the industrial area of Asaluyeh (southern Iran) and the non-industrial area of Kaki. The milk of 16 goat herds (each herd had at least ten goats) was collected in several villages in each area, and at the same time, the drinking water and forage of goats were sampled. The concentration of elements in the samples was determined by ICP-OES. The mean concentrations of chromium, copper, iron, manganese, lead, and nickel in milk samples of the Asaluyeh area were 16.423 ± 0.349, 0.146 ± 0.118, 6.111 ± 0.501, 0.239 ± 0.016, 0.141 ± 0.030, and 1.447 ± 0.101 mg/kg, respectively. Concentrations of heavy metals (except for copper) in the milk of goats in the industrialized area of Asaluyeh were significantly higher than that of Kaki ($P < 0.05$). Also, the content of heavy metals was significantly correlated with lactose levels ($P < 0.05$). The hazard index for drinking the goat milk was computed to be 0.444 and 0.386 for the Asaluyeh and Kaki area, respectively, which shows a minimal effect of this exposure pathway.

Keywords Goat milk · Asaluyeh · Kaki · Health risk assessment · Heavy metals · Nutrient

Introduction

Although metals such as iron, copper, and manganese are essential elements in the body, in large quantities, they cause toxicity. The metal of chromium, lead, and nickel is listed in the priority list of hazardous substances by the Agency for Toxic Substances and Disease Registry (ATSDR) (Kafaei et al. 2017; Ahmad et al. 2017). Heavy metals accumulate in various tissues of the body and can affect the central nervous system or cause other diseases such as impaired intelligence, behavioral impairment, and cardiovascular and renal disease (Christoforidis and Stamatis 2009). Heavy metals are released from fixed and mobile sources such as petrochemicals, gas extraction, refinery processes, dumping sites of industrial wastes, and agricultural fertilizers and can be transferred to water, air, and soil, and even to plants, animals, and the human body (Jolly et al. 2017; Cechinel et al. 2016; Duong and Lee 2011). Environmental monitoring has shown that the levels of metals in the air, soil, and vegetables are high in areas surrounding gas and petrochemical companies. Therefore, heavy metal accumulation in the food chain, organisms, and people living in the contaminated areas leads to long-term health threats.

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✉ Bahman Ramavandi
ramavandi_b@yahoo.com; b.ramavandi@bpums.ac.ir

[1] Department of Environmental Health Engineering, Faculty of Health and Nutrition, Bushehr University of Medical Sciences, Bushehr, Iran

[2] Systems Environmental Health and Energy Research Center, The Persian Gulf Biomedical Sciences Research Institute, Bushehr University of Medical Sciences, Bushehr, Iran

[3] The Persian Gulf Marine Biotechnology Research Center, Bushehr University of Medical Sciences, Bushehr, Iran

[4] Department of Biostatistics, Faculty of Health and Nutrition, Bushehr University of Medical Sciences, Bushehr, Iran

[5] Department of Environmental Science, Persian Gulf Research Institute, Persian Gulf University, Bushehr, Iran

[6] Department of Pathology, Faculty of Medicine, Bushehr University of Medical Sciences, Bushehr, Iran

[7] Student Research Committee, School of Public Health and Safety, Shahid Beheshti University of Medical Sciences, Tehran, Iran

Food and water are the two main ways to get toxic metals (Hussain et al. 2013). Plants form the first part of the food chain; thus, they are essential components of natural ecosystems and agricultural systems (Jolly et al. 2017). The use of sewage sludge in agriculture and the release of pollutants into the atmosphere (through stack and flaring of factories) gradually increase the metal's toxicity of the soil. Then, metal contaminants in the soil cause severe damage to the health of animals and humans (Javed et al. 2009). The feed of livestock from forage and water containing heavy metals leads to intake and accumulation of these elements in the edible tissues and other products like milk (Pajohi-Alamoti et al. 2017). Norouzirad et al. (2018) concluded that the amount of lead metal in most cows' milk samples was higher than the acceptable limit. In other studies, the level of lead and cadmium in milk exceeded the permissible limit (Imam et al. 2017; Iqbal et al. 2016). Milk contains essential nutrients including calcium, magnesium, sodium, potassium, zinc, riboflavin, vitamin B12, and vitamin D (Fayet et al. 2013; Iannotti and Lesorogol 2014). Milk and its derivatives are an important part of the human diet, especially for the elderly and children, due to the presence of compounds such as essential proteins and minerals that contribute to the development and maintenance of human life and health (de Oliveira et al. 2017). Based on the study, exposure to heavy metals through milk and dairy consumption is clear (Shahbazi et al. 2016). Heavy metals in milk can form a complex with nutrients such as calcium and iron and prevent their absorption by the gastrointestinal tract (Goyer 1995). The amount of heavy metals in the milk of cow (Norouzirad et al. 2018), sheep (Rahimi 2013), and buffalo (Ahmad et al. 2017) has been evaluated in Iran and elsewhere, but the effects of the development of gas and petrochemical industries on the quality of livestock milk are still unclear.

Asaluyeh industrial zone is a busy area in terms of petrochemical, gas, and downstream industries as well as loading and unloading on land and sea located in the northern part of the Persian Gulf (southern Iran). Many studies have been published on metal pollution of the Asaluyeh area (Kafaei et al. 2017; Safari et al. 2018). According to the weather conditions of the Asaluyeh area, goats are an important part of the livestock industry and play a vital role in the social-economic structure of rural areas. The quality of goat's milk can be a good indicator of the contamination status of the environment as goat herds are taken to pasture in the surrounding environment. Animals are also exposed to heavy metals through water and feed (grass). Therefore, it is essential to monitor the levels of heavy metals in foods, especially milk, the natural source of mammalian and human infant nutrition and food for humans of all ages. Accordingly, the purpose of this work was to investigate the level of heavy metals (nickel, manganese, iron, copper, chromium, and lead) in the milk of goat herds in villages of the Asaluyeh industrial zone and compare it with that in the villages in the vicinity of the non-industrial area of Kaki. Another goal of this work was to determine the relationship between the content of heavy metals and the level of vitamin D, sodium, potassium, glucose, triglyceride, and cholesterol of the milk samples.

Materials and methods

Study areas

In this work, the study area was selected based on proximity to pollutant sources. The Asaluyeh area, which is adjacent to petrochemical companies and gas refineries, was considered the target area. The Kaki area, which has no polluting industries, highways, and natural pollution, has served as the control area in this study.

Asaluyeh city is located in the south of Iran and on the northern part of the Persian Gulf (27.4721 °N, 52.6146 °E). The city's population is 65,584. The industrial area in Asaluyeh (including gas and petrochemical industries) covers more than 30,000 hectares. Asaluyeh is bounded on the north by the mountains and the south by the Persian Gulf. The map of the study areas is provided in Fig. 1.

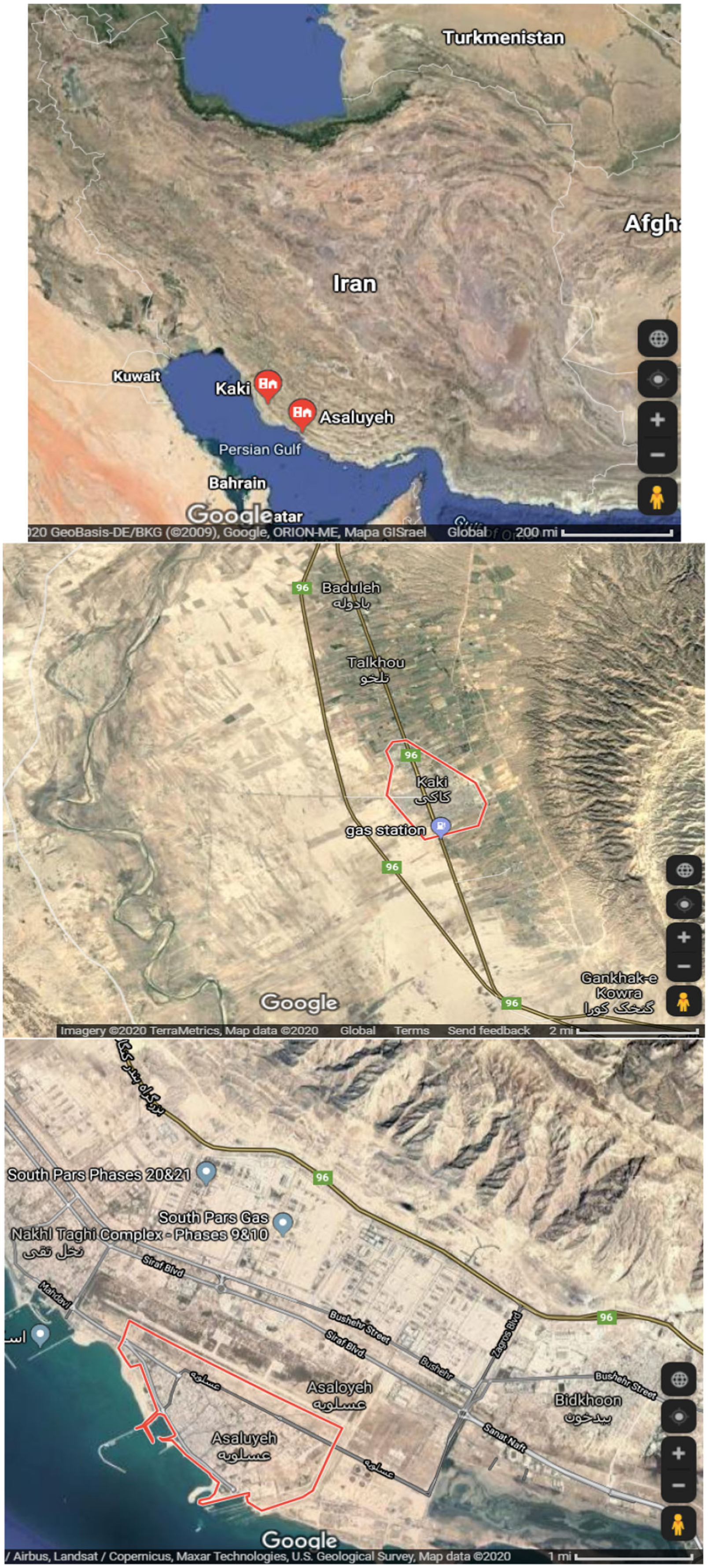
Kaki is an area in southern Iran and is located at 28.3418° N, 51.5229° E. The distance between Asaluyeh and Kaki is approximately 160 km.

Study design and sampling

In both investigated areas (Asaluyeh and Kaki), the herds went to the field in the morning and the afternoon. According to herd owners, goat herds in Asaluyeh used purified water with a reverse osmosis system, and no water was found in the field to feed them. While herds in the Kaki area have used city tap water, springs, and well water. In both areas, alfalfa, wheat straw, barley, and daily food scraps (such as dry bread and fruit shell) were used to feed goats. Goats go to pasture for about 3–6 h, depending on the season.

Samples of milk were collected from villages located in two areas of Asaluyeh and Kaki. For milk sampling, at least 12 villages in each study area were visited. A total of 32 herds of goats (16 herds in Asaluyeh and 16 herds in Kaki) were selected for sampling. One sample was prepared from the milk of each goat flock. In each village, a maximum of 2 milk samples were prepared from two flocks. In this study, a goat flock was sampled that had at least 10 lactating goats, and all the goats had the same water and grass and belonged to one family. All the goats were chosen from one genus and species. Samples were not taken from a goat with an inappropriate physical condition (e.g., excessive weight loss) and the sick ones. Milk samples were not taken from goats whose forage was not supplied from the studied area. Goats that were on the

Fig. 1 Map of study areas (provided from Google Map)



first week of lactation were excluded. Sampling was also done from water and forage of goat flocks. Therefore, in total, 32 milk samples, 32 forage samples, and 32 water samples were prepared and analyzed. Milk samples were collected at sunset when goat flocks came from the pasture. Goats' milk is harvested by their owner and collected in a container. About 200 mL of milk was collected from each flock in an acid-washed polyethylene bottle. The fresh milk was placed beside the ice immediately after being poured into bottles. The amount of 200 g of forage and 300 mL of water was collected at the same time as milk samples were collected. Forage samples were stored in polyethylene bags. Water samples were also collected in the polyethylene bottle. All specimens were transferred to the lab in the least possible time.

The number of samples was calculated according to a study conducted in this field. According to the literature (Rahimi 2013), the average Pb metal in cow milk samples of the two contaminated and non-contaminated sites was 11.4 ± 5.2 mg/kg and 6.81 ± 3.6 mg/kg, respectively. Therefore, considering the probability of the first ($\alpha = 0.05$) and second type of errors ($\beta = 0.2$), the minimum number of samples was calculated to be 32 (each area 16 samples). The sample size was calculated based on the following formula:

$$n = \frac{2\left(z_1-\frac{\alpha}{2}+z_1-\beta\right)^2\left(\sigma_1^2+\sigma_2^2\right)}{\left(\mu_1-\mu_2\right)^2} \qquad (1)$$

Sample preparation and digestion

Perchloric acid (70%) and nitric acid (65%) were purchased from Merck Company. All solutions were prepared using deionized water (resistivity 18.2 MΩ-cm and TOC ≤ 5 μg/L) prepared by American Millipore Company. All dishes used were submerged in diluted nitric acid for 24 h and then rinsed with deionized water before use.

Samples of milk were digested according to Richard's method (Richards 1954; Kabir et al. 2017). The exact amount of 1 mL of milk sample was transferred to a 100-mL flask and then 10 mL of nitric acid was added and heated for 20 min. After that, 5 mL of ultrapure perchloric acid was added. The contents of the flask were then warmed by a direct flame until white fumes appeared and the sample volume dropped to 2~3 mL. The final volume was increased to 50 mL by adding deionized water.

The forage samples were first dried at 50 °C (to reduce the impacts of metal evaporation in the samples) for 24 h. Then, the dried samples were milled using a home mill and sieved to obtain a powder with a particle size of less than 0.5 mm. After use for each sample, the device was thoroughly washed and cleaned to prevent cross-contamination. After that, 1 g of the powder was added to the digestion flasks and then 10 mL of nitric acid was added. For digestion, the mixture was placed at room temperature overnight, then at 100 °C for 4 h, and finally at 140 °C for 4 h. After cooling, the samples were passed through the Whatman 42 filter, and the filtrate was diluted by deionized water to provide a 25-mL sample.

The exact volume of the 30-mL water samples was mixed with 2 mL of nitric acid to reach a pH of 2.

Generally, the digestion of each sample was repeated three times, and the mean of analysis was reported. A control sample was also prepared to determine background contamination during digestion.

Measurement

Concentrations of nickel, vanadium, manganese, iron, copper, selenium, cadmium, and lead were measured by an inductively coupled plasma-optical emission spectrometry instrument (ICP-OES, 730 ES, USA). The most important operating conditions with this device were a plasma gas flow rate of 18 L/min, an auxiliary gas flow rate of 1.5 L/s, carrier gas pressure of 2.10 kg/cm, a peristaltic flow rate of 1.4 mL/min, the detector integration time of 5 s, and the number of integration times 3 for each step. The mean of both injections into the ICP-OES device was considered as a result.

Standard solutions VHG-SM80C-100 and VHG-SM90C-100 (supplied by Merck Co.) were used for calibration. The calibration curve was plotted for each of the studied heavy metals. For the measurement purpose, the corresponding wavelengths for Cr, Cu, Fe, Mn, Pb, and Ni were 267.716, 324.754, 259.940, 260.568, 220.353, and 231.604 nm, respectively. Each curve was constructed with three calibration standards and one calibration blank. The calibration standards consisted of solutions with a given concentration of 0.2, 0.6, and 1 μg/L for each metal. For quality assurance (QA) and control (QC), the 0.2-μg/L standard was used as a check solution of the instrument performance and was analyzed after every ten samples and at the end of the analysis. The limit of detection (LD) was about 0.1 μg/L for all the heavy metals analyzed. Five replicates of the VHG-SM80C-100 and VHG-SM90C-100 (supplied by Merck Co.) were used to determine the percent recovery for each heavy metal. The recovery percent for chromium, nickel, lead, copper, manganese, and iron was obtained as 92.7, 91.0, 87.9, 85.6, 102.4, and 88.2, respectively.

The content of vitamin D in milk samples was measured using the enzyme-linked fluorescent assay (ELFA) technique and the Vidas instrument. The level of nutrients (sodium, potassium, glucose, triglyceride, and cholesterol) was also measured using a Hitachi autoanalyzer by photometry and colorimetry methods.

Statistical analysis

Data were statistically analyzed by using the SPSS v.22.0 software (SPSS, Inc., USA). Descriptive statistics (mean, minimum, and maximum) were used to present the data. The Shapiro-Wilk test was used to examine the normality of data. Since data distribution was not normal, logarithm transformation was used to normalize data distribution. Student's *t* test was also performed to compare the concentration of heavy metals between the two areas. The significance level was defined at $P < 0.05$. Health risk assessment was also calculated based on the mean value.

Health risk assessment

In this study, the human health risk was assessed for non-carcinogenic hazards as described by other investigators (Onyele and Anyanwu 2018; Masood et al. 2019; Fathabad et al. 2018). The chronic daily intake (CDI) of heavy metals in milk was evaluated by the following equation (US EPA I 2011):

$$\mathrm{CDI} = \frac{C_w \times I_R \times EF \times ED}{BW \times AT} \tag{2}$$

where CDI is the daily dose of heavy metals (mg/L) to exposed consumers. C_W is the concentration of heavy metals in the milk (mg/L); I_R is the ingestion rate; EF is the exposure frequency (day/year); ED is the exposure duration (year), BW is the bodyweight of the exposed individual, set to 70 kg for adults; and AT is the average time (day).

Hazard quotient

The hazard quotient (HQ) for non-carcinogenic risk was calculated using the equation defined by USEPA:

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RFD}} \tag{3}$$

where RFD is the oral reference dose which is the daily dosage that enables the individual to sustain this level of exposure over a long period without experiencing any harmful effects. The RFD values of Cr (0.003) (Bortey-Sam et al. 2015), Ni (0.02) (Zeng et al. 2015; Elumalai et al. 2017), Pb (0.0036) (Zeng et al. 2015; Elumalai et al. 2017), Cu (0.005) (Elumalai et al. 2017), Mn (0.14) (Zeng et al. 2015), and Fe (0.7) (Javed and Usmani 2016) were considered for health risk assessment.

If HQ > 1, it represents adverse non-carcinogenic effects while HQ < 1 represents an acceptable level (no concern).

Hazard index (HI)

Since more than one toxicant is present, the interactions are also considered. The toxic risks due to potentially hazardous substances present in the same media are assumed to be additive. The HQs may then be summed to arrive at the overall toxic risk, and the hazard index is computed by the following equation:

$$\mathrm{HI} = \sum_{i=1}^{n} (HQ)i \tag{4}$$

where HI is the hazard index for the overall toxic risk, and *n* is the total number of metals under consideration.

If HI < 1.0, the non-carcinogenic adverse effect due to this exposure pathway or chemical is assumed to be negligible.

Results and discussion

Concentration of heavy metals in the milk of goat herds

The concentrations of copper, iron, manganese, nickel, lead, and chromium in goat milk samples collected from the Asaluyeh and Kaki areas is shown in Table 1. The concentration of lead metal in the Kaki area was not measurable. Nickel, chromium, manganese, and iron were found in all the samples in the two studied areas. Copper was present in all the samples collected from the Asaluyeh area, but only in 12.5% of the samples in those in the Kaki area. The lead metal was also present in all Asaluyeh specimens, but not found in the Kaki samples. The mean concentrations of heavy metals in both the Asaluyeh and the control area (Kaki) followed the same pattern of "Cr > Fe > Ni > Mn > Cu > Pb." As seen in Table 1, the most dominant metal observed in both the industrialized and control areas was chromium with mean concentrations of 16.42 mg/kg and 14.21 mg/kg, respectively. In the Asaluyeh area, the lowest concentration of chromium (15.95 mg/kg) is almost twice the maximum iron concentration (7.78 mg/kg). The iron metal was the second most abundant metal in the milk samples. The mean concentrations of chromium, nickel, and lead in the Asaluyeh area were significantly higher than the control area, and the mean concentration of iron, copper, and manganese was higher in the control area than in the industrialized area. This could underscore the release of toxic

Table 1 Concentration of heavy metals in the milk of goat herds (mg/kg)

Metal	Area	Min-max	Mean ± SD	*P* value
Cr	Asaluyeh	15.954–17.021	16.423 ± 0.349	< 0.001
	Kaki	13.740–14.511	14.211 ± 0.205	
Ni	Asaluyeh	1.334–1.646	1.447 ± 0.101	< 0.001
	Kaki	1.239–1.370	1.319 ± 0.036	
Pb	Asaluyeh	0.110–0.212	0.141 ± 0.030	–
	Kaki	BDL	BDL	
Cu	Asaluyeh	0.066–0.522	0.146 ± 0.118	0.762
	Kaki	0.070–0.273	0.171 ± 0.143	
Mn	Asaluyeh	0.221–0.279	0.239 ± 0.016	0.025
	Kaki	0.219–0.334	0.257 ± 0.027	
Fe	Asaluyeh	5.693–7.786	6.111 ± 0.501	< 0.001
	Kaki	8.370–11.042	9.135 ± 0.558	

BDL below detection limit

Table 2 Comparison between mean concentrations of heavy metals in the goats' milk in the present work with samples in previous studies

Country/organization (area)	Unit	Cr	Ni	Pb	Mn	Cu	Fe	Reference
WHO	mg/kg	0.50	0.40	0.02	55.50	0.4 0	0.50	Ahmad et al. 2017; Iqbal et al. 2016
Iran (Asaluyeh)	mg/kg	16.42	1.44	0.141	0.239	0.146	6.11	This work
Iran (Kaki)	mg/kg	14.21	1.31	BDL	0.171	0.257	9.13	This work
Nigeria	mg/kg	–	–	0.168–1.394	0.047–1.965	–	0.365–0.688	Garba et al. 2018
Pakistan	mg/kg	1.52×10^{-3}	5.15×10^{-3}	–	1.152×10^{-3}	0.223×10^{-3}	0.094 ×	Ahmad et al. 2017
Turkey	mg/kg	–	–	–	0.014–0.105	0.094–26	0.307	Bakircioglu et al. 2018
Egypt	mg/kg	–	–	0.044–0.751	–	0.888–18.316	1.320–45.619	Meshref et al. 2014
Iran (southwest of Iran)	mg/kg	–	–	$47.0 \times 10^{-3} \pm 0.0039$	–	–	–	Norouzirad et al. 2018
Iran (industrial cities)	mg/kg	–	–	$1.93 \times 10^{-3} \pm 1.48 \times 10^{-3}$	–	–	–	Rahimi 2013
Iran (industrial areas)	mg/kg	–	–	14×10^{-3}	–	427×10^{-3}	–	Shahbazi et al. 2016

The unit for some studies was converted to mg/kg. One liter of milk was estimated to equal to 1 kg

BDL below detection limit

metals from the gas and petrochemical companies into the ambient air of the Asaluyeh area. Higher concentrations of these metals were also observed in the forage samples of the exposed area than in the control (discussed below). Similarly, Norouzirad et al. (2018) reported the most important factors affecting the increase in lead and cadmium concentrations in cow's milk samples due to air pollution caused by oil extraction activity and contaminated forage. However, in addition to the contaminated environment, factors such as the period of lactation and the amount of nutrition also affect the increase in the levels of metals (Garba et al. 2018).

Lead is one of the most harmful heavy metals for humans. Lead can cause cancer, bleeding, and kidney disorders. Based on the recommendation of the WHO, the lead limit for milk is 0.02 mg/kg. The average lead concentration of goat milk samples in Asaluyeh was 0.141 mg/kg while it was not observed in Kaki (Table 2). In this study, the average concentration of lead in the Asaluyeh area was higher than the WHO limit. The causes of lead contamination in goat milk in Asaluyeh cannot be related to the water and fodder. Because lead was not found in the drinking water of Asaluyeh goats, and the amount of this element in the forage of the two regions was equal. Therefore, a third path (such as lead-contaminated air) can contaminate goat milk with lead in the Asaluyeh industrial area. In a study in Asaluyeh, lead was found in the atmosphere and dust (Safari et al. 2018). Similar results for areas close to pollution sources such as roads (Simsek et al. 2000; Perween 2015), mining (Giri et al. 2011; Perween 2015), coal-fired power plants (González-Montaña et al. 2012; Tunegová et al. 2016), waste, metropolises, and industrial units (Simsek et al. 2000; Perween 2015; Swarup et al. 2005; Patra et al. 2008; Hyseni and Musaj 2014) have been reported. Our results showed that lead content in the goat milk samples was lower than that observed in Nigeria (Garba et al. 2018) and Iran (Shahbazi et al. 2016; Rahimi 2013) and higher than the values reported in Egypt (Meshref et al. 2014) and Iran (Norouzirad et al. 2018).

Chromium is an essential element because it absorbs sugar, protein, and fat. It also maintains blood cholesterol levels and regulates insulin activity in the body. But, it should be noted that excessive amounts of chromium cause cancer in humans. The total chromium content in foods is generally 0.5 mg/L while the acceptable daily intake is 5–200 μg/day (Iqbal et al. 2016). The concentration of chromium in the goat milk in the Asaluyeh industrial area and Kaki non-industrial area was 16.42 and 14.21 mg/kg, respectively (Table 2). The reasons for the difference between chromium metal in goat milk of the two regions can be found in the significant difference of this metal in water and forage, which will be explained in detail in the following discussions. Ahmad et al. (2017) reported lower levels of chromium in milk than in our study (Ahmad et al. 2017).

Also, the study by Anastasio et al (Anastasio et al. 2006) and AbdulKhaliq et al. (Abdulkhaliq et al. 2012) reported lower chromium content than ours.

Copper is mainly absorbed in the brain, liver, and bone. High concentrations of copper lead to vomiting, diarrhea, and the collapse of the cardiovascular vessels. The daily tolerable amount for copper is 3 mg while its permissible limit in milk is 0.40 mg/kg (Shahbazi et al. 2016). Low concentrations of copper can be due to zinc in foods that can interfere with the absorption of copper. The mean concentrations of copper in Asaluyeh and Kaki were 0.146 mg/kg and 0.171 mg/kg, respectively, in which the difference in values was not significant. In this study, copper was lower than the permissible limit. Other studies reported higher levels of copper than in this study (Iqbal et al. 2016; Ahmad et al. 2017; Bakircioglu et al. 2018; Meshref et al. 2014). A study in Iran reported lower levels for copper than the current investigation (Shahbazi et al. 2016).

Manganese is an essential nutrient for the normal physiological functions of the body, with minimal side effects such as neuromuscular disorders at higher doses. The recommended value for manganese in milk is 55.5 mg/kg. The concentration of Mn in milk samples in Asaluyeh and Kaki areas was 0.239 mg/kg and 0.257 mg/kg, respectively. The significance of the difference in manganese content in the two regions can be linked to goat drinking water, which is explained in the relevant section. Other researchers reported lower contents of copper than in this work (Ahmad et al. 2017; Garba et al. 2018; Bakircioglu et al. 2018). Heavy metals such as manganese, nickel, copper, and iron are important for completing the life cycle but are toxic when they are above the permitted level.

The permissible limit for iron is 0.5 mg/kg (Boudebbouz et al. 2020). The mean iron concentration in Asaluyeh and Kaki area was 6.11 mg/kg and 9.13 mg/kg, respectively. In this study, the iron content in goat milk samples was higher than the limit values. The mean iron concentration in the two regions was statistically significant. According to the results, the presence of iron in water and fodder cannot cause this difference. The presence of metallothioneins and the relationship between metals can be the reason for the high iron content in goat milk (Leotsinidis et al. 2005; Ren-ju et al. 2015), which needs to be studied further. Similar results in the studies of Garba et al. (2018), Giri et al. (2011), and Meshref et al. (2014) indicate high concentrations of iron in milk samples. Some studies reported lower iron levels than those detected in the work (Bakircioglu et al. 2018; Ahmad et al. 2017).

The prescribed limit for nickel in milk is 0.4 mg/kg (Potortì et al. 2013). The mean nickel concentrations in Asaluyeh and Kaki were 1.44 and 1.31 mg/kg, respectively. The high concentration of nickel in goat milk of the Asaluyeh industrial zone is probably related to the content of this metal in forage. The studies of Ahmad et al. (2017), Giri et al. (2011), and Iftikhar et al. (2014) showed higher concentrations of nickel in milk samples than of ours. The result of the Gougoulias et al. (2014) study showed a low concentration of nickel in the samples.

Concentration of heavy metals in drinking water of goat herds

The concentrations of studied heavy metals in drinking water samples of goat herds for the industrial area of Asaluyeh and the non-industrial area of Kaki are listed in Table 3. Chromium, manganese, iron, and nickel metals were detected in all the water samples collected from the Asaluyeh and control area. The amount of copper in both areas was below the detection limit. Also, none of the water samples in the Asaluyeh area contained lead, while this metal was detected in 12.5% of the samples in the Kaki area. The absence of lead in Asaluyeh water is due to the use of advanced treatment methods such as membrane filtration, while the drinking water of goats in Kaki areas is supplied from groundwater without treatment. The concentration pattern of elements in water of Asaluyeh and control samples was “C r> Ni > Fe > Mn” and ‘Ni > Cr > Fe > Mn > Pb,” respectively. These patterns, like the milk samples, imply the dominance of chromium in the water in both areas. Also, lead and copper, the lowest metals measured in the milk, were reported below the detection limit for most water samples. Chromium concentration in the industrialized area was also significantly higher than in the control area (< 0.001). Thus, although no statistically significant relationship was found between the content of metals in the water and milk samples, evidence indicates the effect of water on the presence of metals in the milk samples. Zhou et al. (2019) in China also found that the transfer of metals to cow’s milk occurs through environmental pathways such as water (2019). However, in contrast, Norouzirad et al. (2018) pointed out the lack of correlation between lead and cadmium levels in water and cow milk samples. The permissible levels of lead, chromium, nickel, copper, and manganese in water are 0.01, 0.05, 0.07, 2, and 0.4 mg/L, respectively, and no limit has been set for iron (WHO 2006). The mean concentrations of chromium, lead, nickel, copper, and manganese in the Asaluyeh and Kaki area are lower than those defined by the World Health Organization.

Heavy metal concentration in forage of goat herds

The concentrations of the studied heavy metals in forage samples collected from the Asaluyeh industrial area and Kaki non-industrial area are shown in Table 3. Copper, nickel, chromium, manganese, and iron were observed in all the samples collected from the Asaluyeh and Kaki areas. The Pb metal was found in 68.75% of Asaluyeh samples, while it was detected in 12.5% of Kaki samples. Therefore, it is concluded

Table 3 Concentration of heavy metals in the drinking water (μg/L) and forage of goat herds (mg/kg)

Metal	Area	Min–max	Mean ± SD	*P* value
Drinking water of goat herd				
Cr	Asaluyeh	0.66×10^{-5}–0.82×10^{-5}	$0.76 \times 10^{-5} \pm 0.51 \times 10^{-6}$	< 0.001
	Kaki	0.51×10^{-5}–0.57×10^{-5}	$0.55 \times 10^{-5} \pm 0.16 \times 10^{-6}$	
Ni	Asaluyeh	0.36×10^{-5}–0.59×10^{-5}	$0.50 \times 10^{-5} \pm 0.68 \times 10^{-6}$	< 0.001
	Kaki	0.69×10^{-5}–0.80×10^{-5}	$0.74 \times 10^{-5} \pm 0.37 \times 10^{-6}$	
Pb	Asaluyeh	BLD	BLD	–
	Kaki	0.099×10^{-5}–0.113×10^{-5}	$0.10 \times 10^{-5} \pm 0.94 \times 10^{-7}$	
Cu	Asaluyeh	BLD	BLD	–
	Kaki	BLD	BLD	
Mn	Asaluyeh	0.09×10^{-5}–0.22×10^{-5}	$0.12 \times 10^{-5} \pm 0.32 \times 10^{-6}$	0.036
	Kaki	0.11×10^{-5}–0.50×10^{-5}	$0.17 \times 10^{-5} \pm 0.96 \times 10^{-6}$	
Fe	Asaluyeh	0.38×10^{-5}–0.51×10^{-5}	$0.44 \times 10^{-5} \pm 0.36 \times 10^{-6}$	0.323
	Kaki	0.42×10^{-5}–1.8×10^{-5}	$0.53 \times 10^{-5} \pm 0.34 \times 10^{-5}$	
Forage of goat herd				
Cr	Asaluyeh	2.818–29.886	12.405 ± 7.08	< 0.001
	Kaki	1.175–9.686	4.896 ± 2.37	
Ni	Asaluyeh	1.827–8.187	4.608 ± 2.03	< 0.001
	Kaki	0.845–6.037	2.227 ± 1.27	
Pb	Asaluyeh	0.036–0.501	0.108 ± 0.13	0.919
	Kaki	0.047–0.125	0.086 ± 0.05	
Cu	Asaluyeh	4.222–93.817	15.946 ± 23.87	0.053
	Kaki	1.900–13.555	6.767 ± 3.90	
Mn	Asaluyeh	47.990–188.329	82.086 ± 33.45	< 0.001
	Kaki	16.176–11576	46.005 ± 29.99	
Fe	Asaluyeh	4.253–2494.57	1044.53 ± 669.02	0.005
	Kaki	4.775–1269.55	262.52 ± 316.415	

BDL below detection limit

that the irrigation water of the fodder or the atmosphere of the Asaluyeh region was contaminated with lead, which caused the contamination of the fodder. The decreasing trend of metal concentrations in forage samples in both areas was similarly reported as Fe > Mn > Cu > Cr > Ni > Pb. The mean concentrations of chromium, nickel, manganese, and iron were significantly higher in the exposure area than in the non-industrial area. These results show the obvious effect of petrochemical emissions on increasing the amount of metals in forage. Therefore, an increase in the concentration of nickel,

Table 4 R^2 value of linear regression to show the relationship between heavy metal, vitamin D, and nutrients concentrations in milk and those in milk/forage/water

Kaki	Pb in milk vs Pb in forage	0.325
Asaluyeh	Vitamin D in milk vs Cr in milk	0.238
Asaluyeh	K in milk vs Cr in milk	0.526
Asaluyeh	Na in milk vs Cr in milk	0.286
Asaluyeh	Cholesterol in milk vs Cr in milk	0.233
Asaluyeh	Triglyceride in milk vs Cr in milk	0.54
Asaluyeh	Lactose in milk vs Mn in milk	0.224
Asaluyeh	K in milk vs Ni in milk	0.235
Asaluyeh	K in milk vs Pb in milk	0.235
Asaluyeh	Lactose in milk vs Pb in milk	0.308

Only R^2 values greater than 0.2 are listed in this table.

Table 5 Concentration of vitamin D and nutrients (sodium, potassium, cholesterol, lactose, and triglyceride) in milk of goat herds (mg/kg).

Nutrient	Area	Min–max	Mean ± SD	*P* value
Vitamin D	Asaluyeh	0.005–1.290	0.57 ± 0.41	0.054
	Kaki	0.464–1.101	0.71 ± 0.20	
K	Asaluyeh	255.0–574.0	498.1 ± 80.2	0.47
	Kaki	180.0–611.0	476.6 ± 121.5	
Na	Asaluyeh	210.0–900.0	343.1 ± 197.7	0.375
	Kaki	190.0–1030.0	308.1 ± 216.4	
Cholesterol	Asaluyeh	40.0–270.0	118.7 ± 58.9	0.825
	Kaki	40.0–460.0	127.5 ± 110.4	
Lactose	Asaluyeh	10.0–70.0	36.8 ± 18.5	0.007
	Kaki	20.0–170.0	68.7 ± 41.6	
Triglyceride	Asaluyeh	420.0–3170.0	1943.1 ± 796.7	0.554
	Kaki	770.0–2940.0	2090.6 ± 579.7	

Table 6 HQ and HI values for the studied heavy metals in the milk of goat herds

Area	HQ value						HI value
	Cr	Ni	Pb	Cu	Mn	Fe	
Asaluyeh	0.4264	0.0054	0.003	0.0022	0.00013	0.0067	0.444
Kaki	0.3692	0.005	–	0.0017	0.000142	0.0101	0.386

chromium, and lead in the samples of milk from the contaminated area compared with the control could be related to an increase in the concentration of these metals in the forage. Higher concentrations of iron, copper, manganese, chromium, nickel, and lead in the forage of the Asaluyeh area than of the control area might be indicate the effect of metal-pollutant emissions from gas and petrochemical industries on crops/forage in the area. Although according to R^2 value (Table 4) the concentration of metals in milk did not show a strong correlation with those in forage and water, higher amounts of chromium, lead, and nickel in Asaluyeh milk samples than in control confirmed the release of metals by industry and entering the food cycle. Air pollution from industrial areas, as well as unsuitable disposal of industrial wastewater, can have a huge impact on the heavy metal content of plants. For example, Roba et al. (2016), while studying the amount of heavy metals in vegetables and fruits around mines in Romania, concluded that the likelihood of metal contamination in vegetables is very high which can pose potential hazards to residents. Another study also found that heavy metals in the soil around sugarcane and paper industries were high and that chromium and nickel in sugarcane were higher than recommended by the World Health Organization (Pandey et al. 2016).

Vitamin D and nutrients (sodium, potassium, cholesterol, lactose, triglyceride) in the milk of goat herds

The concentrations of vitamin D and nutrients (sodium, potassium, cholesterol, lactose, and triglyceride) in the goat milk samples collected from the Asaluyeh industrial area and the non-industrial Kaki area are shown in Table 5. The decreasing trend of nutrients in milk samples in both areas was similarly reported as "triglyceride > potassium > sodium > cholesterol > lactose." The mean concentration of sodium, potassium, cholesterol, and triglyceride in the Kaki area and the industrial area of Asaluyeh was not significant. However, the lactose concentration in the control area was significantly (< 0.007) higher than in the Asaluyeh industrial area. The mean concentration of vitamin D in the Kaki and Asaluyeh area was not significantly different. These results showed that petrochemical emissions had no significant effect on vitamin D and nutrient levels.

Health risk assessment of drinking goat milk

The HQ for the evaluated heavy metals is presented in Table 6. These results are less than 1, indicating an acceptable level of risk for all elements. The hazard index (HI) was 0.444 and 0.386 for Asaluyeh and Kaki areas, respectively. Because the results are less than 1, we consider the health risks of this route (drinking of milk) to be negligible. Although the target health quotient (THQ) indicates milk samples safe for drinking, special attention should be paid to metal concentrations. It is difficult to reduce metals to an acceptable level if their concentration is higher than the permitted concentration. In a study, acceptable levels of risk (HQ) and carcinogenic risk (CR) have been reported (Muhib et al. 2016). In Bangladesh, the health risk assessment through consumption of powdered milk and cow's milk was investigated, and it was concluded that the powdered milk contains a higher concentration of heavy metals than liquid one (Jolly et al. 2017). Kabir et al. (2017) have conducted that cadmium, chromium, arsenic, and mercury were the most dangerous toxic elements present in the cow's milk samples.

Conclusions

The results showed that the amount of heavy metals in milk samples in the Asaluyeh industrial zone is significantly higher than that in the non-industrial Kaki area. Also, a significant difference in milk lactose content for both studied areas was detected. The water samples for goat herds had high chromium and nickel concentrations similar to milk samples. Heavy metal sequences in goats forage samples in both areas were observed as Fe > Mn > Cu > Cr > Ni > Pb. Results of health risk assessment showed that the goat milk in the two areas has no health risk (risk index < 1). But the role of milk and dairy consumption in Iran should not be overlooked.

Authors' contributions N. Homayonibezi: formal analysis, writing—original draft. S. Dobaradaran: methodology. H. Arfaeinia: data curation. M. Mahmoodi: methodology. A.M. Sanati: methodology. M.R. Farzaneh: data curation. R. Kafaei: data curation, writing—original draft. M. Afsari: data curation. M. Fouladvand: conceptualization, methodology. B. Ramavandi: supervision, conceptualization, methodology.

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Data availability All data in this article will be available upon request.

Compliance with ethical standards

Ethical approval The study protocol and ethics of this work have been approved by the Ethical Committee of the Bushehr University of Medical Science.

Consent to participate No human or animal specimens were used in this work. To avoid damage to the environment, the lowest weight of the algae sample was collected.

Consent to publish All the authors agreed to publish the data in this journal.

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