

Toxic pollutants in camel milk from the Mangystau Region of Kazakhstan

—
Results of sampling
conducted in 2015–2016
—

Prague–Aktau 2016



TRANSITION



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Based on the results of environmental sampling conducted in Kazakhstan in 2015–2016 as a part of the project “Enforcing citizens’ rights and public participation in decision making on environmental issues – practical implementation of Aarhus Convention in Mangystau” financially supported by the European Union and Transition Promotion Programme of the Czech Republic.

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1. Introduction

Mangystau Region is among the parts of Kazakhstan with large oil and mining activities where the toxic legacy “hot spots” from the country’s Soviet era can be found. Toxic contamination of food represents one of the major challenges in designing a sustainable future for the region. Nurseitova, Konuspayeva et al. (2016) recently tried to assess the risks of toxic contamination in Kazakhstan for livestock production as a food source and concluded that „*the assessment of contamination risk is not yet known in the situation of Kazakhstan*”. Arnika, EcoMuseum and CINEST too found serious gaps in knowledge about the level of food contamination, particularly by PCBs, PCDD/Fs, and PAHs, in Kazakhstan. We believe that this study, together with previous reports published by Arnika, EcoMuseum and CINEST (Arnika, AWHHE et al. 2015, Arnika, EcoMuseum et al. 2015), contributes to an overall evaluation of toxic contamination risks in certain regions of Kazakhstan.

In this study, we have focused on camel milk contamination as it is a significant part of the diet in Mangystau Region which is the main target area of the project “Enforcing citizens’ rights and public participation in decision making on environmental issues – practical implementation of Aarhus Convention in Mangystau”. It is a joint three-year project of Czech and Kazakhstani NGOs financed by the European Union and the Transition Promotion Programme of the Czech Ministry of Foreign Affairs.

1.1 Milk contamination by toxic chemicals – brief overview of existing studies

It is not very common to monitor levels of contamination by Persistent Organic Pollutants (POPs) and toxic heavy metals in camel milk; however, in Kazakhstan several previous studies analysed the content of different chemicals in camel milk (Diacono, Faye et al. 2008, Meldebekova, Konuspayeva et al. 2008, Konuspayeva, Faye et al. 2009, Konuspayeva, Jurjanz et al. 2011, Konuspayeva, Faye et al. 2011 a). Camel milk is a significant part of the diet in southern and western regions of Kazakhstan.¹ It can be compared to the consumption of cow’s milk in some other countries. Contamination of cow’s milk by various chemicals was studied in relation to specific contaminated sites (Braga, Krauss et al. 2002), particular pollution sources (Liem, Hoogerbrugge et al. 1990, Riss, Hagenmaier et al. 1990, Grova, Feidt et al. 2002, Andre, Marchand et al. 2004, Diletti, Ceci et al. 2008, Esposito, Cavallo et al. 2009) or in particular countries (De Fre and Wevers 1998, Cerkvenik, Doganoc et al. 2000, Hamm, Fuchs et al. 2001, Schaum, Schuda et al. 2003, Schmid, Gujer et al. 2003, Thanner and Moche 2004, Hsu, Chen et al. 2005, Durand, Dufour et al. 2008, Amirova and Shahtamirov 2011, Mocanu, Nistor et al. 2012, Iwegbue and Basse 2013, Concannon 2014, Pietrzak-Fiećko, Gałgowska et al. 2014). Cow’s milk is also commonly included in broader studies focused on the dietary intake of specific contaminants or the monitoring of feed and food contamination (Theelen, Liem et al. 1993, Muntean, Jermini et al. 2003, BiPRO 2004, Schecter, Pöpke et al. 2004, Taioli, Marabelli et al. 2005, European Food Safety Authority 2010, Martorell, Perelló et al. 2010, European Food Safety Authority 2012, Husain, Gevaio et al. 2014). One study from Ireland studied impacts of feeding cows with a supplement of shredded newspapers on the content of heavy metals, PAHs and PCBs in cow’s milk (O’Connell and Meaney 1997). Several studies focused on contamination of goat’s or sheep’s milk (Schulz, Wiesmuller et al. 2005, Costera, Feidt et al. 2006, Perugini, Nuñez et al. 2012, Storelli, Scariano et al. 2012, Arkenbout 2014, Pietrzak-Fiećko, Gałgowska et al. 2014). Butter was analysed for dioxins in the Uzbekistan’s Aral Sea Region (Ataniyazova, Baumann et al. 2001).

Cow’s and sheep’s milk has been found to be a sensitive indicator of POP contamination in feed, soils or dust and is an important exposure pathway from feed (Schulz, Wiesmuller et al. 2005, Malisch and Kotz 2014) or soil pollution (Rychen, Ducoulombier-Crépineau et al. 2005, Schulz, Wiesmuller et al. 2005, Diletti, Ceci et al. 2008, Perugini, Nuñez et al. 2012) to humans. Milk from contaminated areas can

¹ The annual per capita consumption reached 240 l in 2008. Meldebekova, A., G. Konuspayeva, E. Diacono and B. Faye (2008). Heavy Metals and Trace Elements Content in Camel Milk and Shubat from Kazakhstan. Impact of Pollution on Animal Products. B. Faye and Y. Sinyavskiy. Dordrecht, Springer Netherlands: 117-123.



readily lead to exposures exceeding thresholds for the protection of human health (Riss, Hagenmaier et al. 1990, Malisch and Kotz 2014). Some studies focused on bioavailability or transfer of different POPs in goat's milk (Costera, Feidt et al. 2006, Lapole, Rychen et al. 2007, Ounnas, Feidt et al. 2010), cow's milk (McLachlan 1993, McLachlan 1996, McLachlan and Richter 1998, Thomas, Sweetman et al. 1999) or even camel milk (Nurseitova, G. et al. 2014). Lactating animals and their milk might, therefore, be ideal "active sampler" and indicator species for evaluation of the level of contamination in sampled areas by POPs, particularly by dioxins (PCDD/Fs) and PCBs. When contaminated, camel milk in Kazakhstan, as well as cow's milk in some other countries, can lead to a significant body burden in respective countries as it represents a large portion of the human diet.

Based on this assumption, we have chosen camel milk in Mangystau Region (western Kazakhstan by the Caspian Sea) and its analysis for selected POPs as one of the monitoring tools within the project "Enforcing citizens' rights and public participation in decision making on environmental issues – practical implementation of Aarhus Convention in Mangystau". The data and analyses of camel milk discussed in this report were obtained during three field visits in 2015 and 2016. Localities, where samples were collected, are described in section 2.1 of this report. A general description of sampling and analytical methods is provided in section 2.2 of this report.

2. Sites, sampling, and analyses

2.1 Sampled sites

A detailed description of sampled sites and information about collected samples are provided in the following text and tables. The location of sampled localities is also shown on the maps in Figures 1 and 2.



Figure 1: Map of camel milk/shubat sampling locations in Mangystau Region, Kazakhstan.



Figure 2: Satellite image of localities in Baskuduk, Akshukur and Kyzyl Tube in Aktau city vicinities broader area with marked major uranium mines and Lake Koshkar Ata.

2.1.1 Shetpe: Zharmysh farm

GPS location: 44.08926, 52.12111

Sample: KZ-M-15-1

The Zharmysh farm where the sample was taken is located in Mangystau Region about 28 kilometres from the Shetpe town (population 13,364 according to 2012 data).² It is connected with Aktau and with the Zharmysh farm by a road A33. See also map in Figure 3.

A potential source of pollution is the **Caspicement plant** situated 7 kilometres from the centre of Shetpe. Caspicement is a new plant constructed by the multinational HeidelbergCement corporate group which was officially opened in July 2014. It is the only cement production site in Mangystau with a production capacity of 800 thousand tons of cement per year. It is one of the first plants in the world using dry chalk for clinker production. The plant is using oil as a fuel. Local people state that the factory often releases uncontrolled exhaust gases during nights and weekends when the state authorities cannot thoroughly control the facility.

2.1.2 Aktau: Baskuduk

GPS location: 43.69735, 51.20789

KZ-M-15-2

The mixed camel milk sample (as shubat) was taken from a farm situated in Baskuduk, fast growing suburb approximately 10 kilometres north of the centre of Aktau. The settlement is inhabited mainly by Kazakh minorities repatriated from the neighbouring countries (Uzbekistan – Karakalpakstan, Turkmenistan). A substantial part of the newcomers had illegally occupied land on the outskirts of the city due to the insufficient state housing programme. The area is located in the northern part of Aktau city;

² Wikipedia. (2016, 30-03-2016). "Шетпе." Retrieved 31-07-2016, 2016, from <https://ru.wikipedia.org/wiki/Шетпе>.





Figure 3: Satellite image of Zharmysh farm – a sampling location near Shetpe.



Figure 4: Map of surroundings of sampling site in Baskuduk.

squeezed between the Koshkar Ata industrial tailing pond and a municipal landfill in the north and an industrial zone including chemical industry in the east (see maps in Figures 2 and 4).

A potential source of pollution is the **Koshkar Ata** tailing pond (77km²); its southern border is located about 7 kilometres from the sampling point. During the Soviet era, the former Koshkar Ata depression was chosen as a convenient location to accumulate liquid industrial waste from Aktau industrial zone. After decades of the wastewater discharge, a large artificial lake was formed. The lake is considered to be the most hazardous structure in Mangystau Region (Kadyrzhanov, Kuterbekov et al. 2002), although some more recent studies concluded that „*the negative impact is registered only in the immediate vicinity of the perimeter of the tailing lake in the first hundreds of metres*” (Zhanpeissova, Kuterbekov et al. 2005). The lake allegedly contains almost 360 million tons of radioactive waste (Akhmetov, Kadyrzhanov et al. 1999).

A major operator in the industrial zone was a uranium processing plant which means that the wastewaters and sludge are contaminated by radioactive matter. Another large factory located in the area was an Aktau Plastic Plant. There are also several other factories, mainly in the chemical industry, discharging various toxic wastes.

Nowadays, wastewater from the industrial zone is still discharged into the lake by the open canal. Sewage water is also discharged into the lake because the city failed to finish the construction of wastewater treatment plant. The vicinity of Koshkar Ata is widely used as an illegal dumping ground for industrial and household waste. The dumpsite contains a diverse mixture of waste: used furniture, animal carcasses, fluorescent tubes, food leftovers, glass bottles, oil sludge, etc. The animals such as horses and camels come here to drink as the Mangystau Region is short on water. The highest concentration of contaminants, solid waste and high levels of radioactivity are concentrated in the southern part of the lake.

Other sources of pollution include several **former and current chemical plants** located in the industrial zone of the city of Aktau. For example, former unsecured uranium extraction plant situated 6 kilometres from the sampling point. The former sulphuric acid production plant is located 6 kilometres from the sampling point. Media reported that about 4 thousand tons of sulphur and about 96 thousand tons of other chemical waste had been found on the site of the former plant. The place is unsecured, freely accessible, and the wind can transfer the chemicals to the neighbourhoods (Kazakhstan Today 2013). Former nitrogen fertilizer plant (AT3) is located about 13 kilometres away from the sampling point. The plant used to be one of the largest nitrogen fertilizer producers in the Soviet Union and it has currently restarted the production as the only such plant in the Republic of Kazakhstan (under the new name KazAzot).

Another contaminated spot in the Aktau area and in the vicinity of Baskuduk is the so-called **Dead Lake** or **Malaya Oymasha Lake**, about 10 kilometres south of the sampling point. The lake has no tributary and is divided into two parts that differ in colour: red and turquoise blue. The water of the lake is salty but used by local inhabitants for unpermitted cattle bath. Some sources cited that radioactivity of natural radionuclides in water sample from this lake is exceeding allowed limits of activity. It might have originated as a disposal of waste waters from the uranium extraction plant operating in Aktau in the Soviet era or by generally high level of natural radioactivity in uranium-rich region. There is also an illegal dump of construction and household waste on the shores of the lake.

Municipal waste dump located right on the edge of Baskuduk may also contribute to the overall pollution of the site.

2.1.3 Kuryk

GPS location: 43.17447, 51.67954

Sample: KZ-M-15-3

Kuryk town is situated about 70 kilometres south of Aktau city, connected to it by an asphalt road. Kuryk is an administrative centre of the Karakiya District of Mangystau Region. According to the data from 2015, the population of Kuryk is 10,318 inhabitants.³

Insufficient **municipal waste management** is the most significant source of pollution at this site. Waste containers are overfilled and open, freely accessible to domestic animals. As the vegetation is scarce in the semi-desert area, the domestic animals are often looking for leftovers in the containers. Dumpsites are also spread around the town. Local residents report that in winter, when there is a lack of vegetation, farmers feed their cows and camels with a carton from cardboard boxes, cheaply obtained in the freight port.

³ Wikipedia. (2016, 31-07-2016). "Құрық" Retrieved 02-08-2016, 2016, from <https://kk.wikipedia.org/wiki/Құрық>.



2.1.4 Akshukur

GPS location: 43.77333, 51.06527

Sample: KZ-M-15-4

Akshukur is a village located on the shore of the Caspian Sea, 20 kilometres north of Aktau and about 8 kilometres from the **Koshkar Ata tailing pond** as the crow flies. In 2009, the village had 6,230 inhabitants and the number probably increased significantly in 2016, as the extensive construction of new family houses is visible.⁴

A possible source of pollution might be the Koshkar Ata tailing pond described above (see section 2.1.2 and map in Figure 2). Another very likely source of milk contamination is the inadequate waste management.

There is no official waste management organized by the local council in Akshukur. Residents can either carry their waste to Aktau and – as they are not residents of Aktau – throw it “illegally” to some waste container in the city, or dump it illegally in Akshukur. There are also fire pits in the inner yards of the houses, and part of the waste is burned in the households.

Illegal waste dump is located 2 kilometres northeast of the sampling site, next to the local cemetery. It is not fenced, so the animals can roam there freely, and it contains both industrial and municipal waste, car wrecks, fluorescent tubes as well as animal carcasses (camels, dogs, horses, cows). Herds of cows were spotted grazing at the location as, most likely, do the camels.

In 2016, the municipality announced in the press a clean-up project and the construction of the official landfill.

2.1.5 Tauchik

GPS location: 44.34837, 51.3531

Sample: KZ-M-15-5

The sampling site is located within the Tauchik village, a settlement in the Tuparkagan district, 100 km north from Aktau. The population of Tauchik, as of the 2009 census, is about 2,600 people.⁵ It was founded in the early 1930s near the deep coal reserves. In the period 1932-39, **the coal mining** flourished and the Tauchik mine was an important coal supplier during the World War II; however, it stopped operation after the 1950s and 60s.⁶ Large mineshafts and associated equipment, now abandoned, are located about 3 kilometres from the sampling point (see map in Figure 5).

The area is also associated with **oil extraction and transport**, which could be another pollution source. Nearby oil fields are called Karazhanbasmunay, Karakudukmunai, Buzachi Oilfield. About 4 km from Tauchik to the south, a small settlement with the KazMunaiGaz gas station is located at the junction of the Tauchik-Zhyngyldy road and the major road connecting Aktau and Kiyakty with the Khalamkhas oil extraction field. The field, situated on the shelf of the Caspian Sea, is owned by the MangystauMunaiGaz Company operating together with the KazMunaiGaz at the northern extraction site. The Khalamkhas-Karajanbas-Aktau pipeline runs along the road.

4 Wikipedia. (2016, 22-02-2016). “Акшукур.” Retrieved 31-07-2016, 2016, from <https://ru.wikipedia.org/wiki/Акшукур>.

5 Wikipedia. (2015, 08-09-2015). “Таушық.” Retrieved 31-07-2016, 2015, from <https://ru.wikipedia.org/wiki/Таушық>.

6 Wikipedia. (2015, 17-05-2015). “Таушық.” Retrieved 31-07-2016, 2015, from <https://kk.wikipedia.org/wiki/Таушық>.



Figure 5: Satellite image of Tauchik sampling site and nearby area.

2.1.6 Kyzyl Tube

GPS location: 43.75444, 51.53333

Sample: KZ-M-16-6

The sampling site is located in an open steppe northeast from Aktau, about 18.5 km from the nearest settlement – Bayandy village with about 2,050 inhabitants (as of 2009) – and about 20 km from the Mangystau town (14.8 thousand inhabitants, as of the 2009 census) on the outskirts of Aktau city.^{7,8} The nearby Aktau–Karman Ata road runs 10 km from the sampling site, and the Aktau–Shetpe railway lies approximately 9 km from the site (see map Figure 2).

There are 2 **abandoned uranium mines**⁹ in the vicinity that used to supply a uranium processing plant and a nuclear reactor in Aktau. The mines do not operate since 1990 and their facilities are not preserved. The volume of extracted uranium is unknown.

There are oil wells also visible on the horizon.

2.2 Sampling and analytical methods

2.2.1 Sampling

Samples of camel milk mostly in the form of fermented milk product – shubat¹⁰ were collected at six localities in Mangystau Region (see Figure 1 and section 2.1). Only one was a sample of the raw camel milk.

Where it was possible, pooled samples from more individual lactating camels were collected at each of the selected sampling sites in order to get more representative samples. At farms in Baskuduk and Akshukur, shubat had already been mixed from the milk of a large number of camels (up to 200). In Tauchik, only one lactating camel was available when camel milk was sampled in autumn 2015. One additional

7 Wikipedia. (2015, 22-02-2015). "Баянды (Мангистауская область)." Retrieved 31-07-2016, 2015, from [https://ru.wikipedia.org/wiki/Баянды_\(Мангистауская_область\)](https://ru.wikipedia.org/wiki/Баянды_(Мангистауская_область)).

8 Wikipedia. (2015, 04-04-2015). "Mangistau." Retrieved 31-07-2016, 2015, from <https://en.wikipedia.org/wiki/Mangistau>.

9 The Uranium Mine No. 2.3 is located about 8km northwest and the Uranium Mine No. 4 lies 12km southwest of the site.

10 Shubat or chal is a popular Kazakh and Turkmen beverage of fermented camel milk, sparkling white with a sour flavor. It is sold mixed with certain amount of water, therefore its fat content is lower than in raw camel milk.



site, Kyzyl Tube, was sampled in spring 2016. This difference has to be taken into consideration when evaluating the observed levels of certain chemicals as camels can excrete different chemicals in milk unproportionally in different seasons of the year (Nurseitova, G. et al. 2014).

All lactating camels were grazing most of the time and in winter they were getting additional mixed feed bought from the shop and/or mixture of kitchen leftovers. They were of different age as marked in Table 1 for each of the sampling locations. All farmers or families consumed their own milk/shubat and sold the rest at the markets.

Table 1 summarizes the basic data regarding the size of samples and measured levels of fat content in each of the samples. In total, five pooled samples of camel milk plus one sample from an individual lactating animal were taken. Samples were collected at the farms and local market and already mixed. One liter of mixed milk was then taken from pooled (mixed) sample and kept frozen (approx. – 10°C) until it was sent to a laboratory for analysis.

Table 1: Overview of samples: one sample of raw camel milk (from Kuryk) and five samples of shubat made from camel milk from selected sites in Kazakhstan.

Sample	Locality	No. of camels in pooled sample	Age (in years)	Fat content	Date of sampling	Shubat/milk source
KZ-M-15-1	Shetpe	3	5 to 8	0.62%	30/09/2015	farm
KZ-M-15-2	Baskuduk	max. 200	various	0.85%	05/10/2015	farm
KZ-M-15-3	Kuryk	3	3 to 5	3.13%	07/10/2015	farm
KZ-M-15-4	Akshukur	max. 200	various	2.21%	08/10/2015	farm
KZ-M-15-5	Tauchik	1	5	1.06%	08/10/2015	family
KZ-M-16-6	Kyzyl Tube	25	2 to 20	1.63%	25/05/2016	farm

2.2.2 Chemical analyses

Analyses for dioxins, PCBs, PAHs and OCPs were carried out in the Czech Republic in Axys Varilab laboratory.¹¹ A 30g sub-sample was dried with anhydrous sodium sulphate, spiked by internal standards and extracted by toluene in a Soxhlet apparatus. A small portion of the extract was used for gravimetric determination of fat. The remaining portion of the extract was cleaned on a silica gel column impregnated with H₂SO₄, NaOH, and AgNO₃. The extract was further purified and fractionated on an activated carbon column. The fraction containing dioxins (PCDD/Fs), PCB, OCPs, and PAHs was analysed by HR GC-MS on Autospec Ultima NT. Analysis of PCDD/F meets requirements of European Standard EB 1948 1, 2 and 3 (Stationary source emissions – Determination of mass concentration of PCDDs/PCDFs)

When analyzing PCDD/F and dioxins like PCBs in food and feed, laboratory meets the requirements of the European Council:

- COMMISSION DIRECTIVE 2002/70/EC of 26 July 2002 establishing requirements for the determination of levels of dioxins and dioxin-like PCBs in feeding stuffs
- COUNCIL DIRECTIVE 2001/102/EC of 27 November 2001 amending Directive 1999/29/EC on the undesirable substances and products in animal nutrition
- COUNCIL REGULATION (EC) No 2375/2001 of 29 November 2001 amending Commission Regulation (EC) No 466/2001 setting maximum levels for certain contaminants in foodstuffs

Analyses for heavy metals were carried out at the specialized chemical laboratory of the State Vet-

¹¹ Laboratory Axys Varilab jointly owned by a Czech-Canadian company, which provided the analysis, is a laboratory certified by the Czech Office for Standards, Metrology and Testing (ÚNMZ) for analysis of POPs in air emissions, environmental compartments, wastes, food and biological materials. Its services are widely used by industry as well as the Czech governmental institutions. It also works for agencies outside of the Czech Republic.

erinary Institute, Prague. The total mercury (Hg) content was determined directly in the sample units by the selective mercury analyser (Advanced mercury analyser, AMA-254) based on atomic absorption spectroscopy (AAS). Other toxic metals (Pb, Cd, and Cr) were measured by the means of electrothermal (flameless) atomic absorption spectrometry with Zeeman background correction (graphite furnace atomic absorption spectrometry (GF-AAS, SpectrAA 220Z, Varian) after microwave mineralisation of the samples (EN13 804, 13805 and 14084). Remaining heavy metals (Al, Zn, Cu and Mn) were measured by the means of inductively coupled plasma mass spectrometry (ICP-MS, Varian) after microwave mineralisation of the samples (EN13 804, 13805 and 14084). The concentrations of all target analytes in the samples were determined and expressed in wet weight (w.w.).

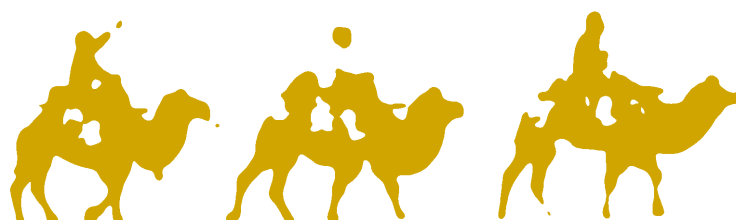
3. The Kazakhstani, EU, and other limits for POPs and heavy metals in milk

Camel milk (processed into shubat or added fresh to the tea) is a relatively common part of the diet in southern and western regions of Kazakhstan and it is also common practice that people in Kazakhstan sell leftover camel milk at local markets. Shubat (and sometimes milk) from camel farms is sold in specialized shops. Shubat from large-scale farms can be also bought bottled in ordinary stores.

There are no specific limit values for various toxic compounds in camel milk, however, maximum limit values for milk as such and/or milk products exist in different countries which can be applied to camel milk as it is done for milk of other ruminants. Limit values for various POPs and heavy metals are summarized in Table 2.

Table 2: Limit concentration values for OCPs, heavy metals, PAHs, PCBs and PCDD/Fs TEQs in milk

Limit values for POPs	Milk				
	Kazakhstani MAC ⁷	Russian MAC ¹	Russian MAC ¹	EU ML ²	EU MRL ³
Unit	ng g ⁻¹ *	ng g ⁻¹ fat	ng g ⁻¹ *	ng g ⁻¹ fat	ng g ⁻¹ fresh weight
WHO-PCDD/Fs TEQ	0.003	0.003		0.0025	
WHO-PCDD/Fs-dl-PCB TEQ				0.005	
PCBs ⁵				40	
DDT and its metabolites	50 (1,000 for cream)	10			40 ⁶
p,p'-DDT			50		
gama-HCH (lindane)					1
alfa-, beta-HCH					4; 3**
alfa-, beta-, gama-HCH as a sum	50 (1,250 for cream)	20	50		
HCB					10
Aldrin					10***
Heptachlor					4****
	Infant formulae and follow-on formulae, including infant milk and follow-on milk ⁴				
4 PAHs					1.0



Limit values for heavy metals	Milk			
	Kazakhstani MAC ⁷	Russian MAC ¹	Russian MAC ⁴	EU ML ¹⁰
Unit	ng g ⁻¹ *	ng g ⁻¹ fat	ng g ⁻¹ *	ng g ⁻¹ fresh weight
Mercury	5	5	5	
Copper	300–600 for children 0–5 months of age for adopted milk based mixtures; 400–1,400 in milk-based food for premature children; 400–1,000 for children 5–12 months of age; 400–1,000 in partly adopted milk based mixtures for young children; 600–1,000 in milk-based food for nursing mothers and pregnant women; 400 in milk butter for long storage	300–600 for children 0–5 months of age 400–1,000 for children 5–12 months of age; 300–1,000 for children 0–12 months of age;	1,000 (for children and adults)	
Lead	100	20	100	20
Zinc		4,000–10,000 for children 0–12 months of age for all food) ¹¹	5,000	
Cadmium	30	20	30	
Arsenic	50	50	50	

1 Current Russian СанПиН 2.3.2. 2401-08 Hygienic safety and nutrition value requirements for food. Sanitary-epidemiologic rule and normatives (СанПиН 2.3.2. 2401-08 Гигиенические требования безопасности и пищевой ценности пищевых продуктов Санитарно-эпидемиологические правила и нормативы)

2 EU Regulation (EC) №1259/2011

3 Regulation (EC) №149/2008. Maximum residue level (MRL) means the upper legal level of a concentration for a pesticide residue in or on food or feed set in accordance with the Regulation, based on good agricultural practice and the lowest consumer exposure necessary to protect vulnerable consumers.

4 Russian Federation GN 1.2.2701-10 Hygienic normatives (standards) pesticides concentration in environmental media (ГН 1.2.2701-10 "Гигиенические нормативы содержания пестицидов в объектах окружающей среды")

5 sum of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180

6 sum of p,p'-DDT, o,p'-DDT, p,p'-DDE and p,p'-DDD

7 Kazakhstan SanPin Hygienic safety requirement and nutrition value for food from 6 August 2010, No 611

8 Commission Regulation (EU) No 835/2011 of 19 August 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs

9 Sum of benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene

10 Regulation (EC) No 1881/2006

11 The upper limit of zinc intake for children 1–3 years of age is 7mg/day

* not clear whether calculated for fat content or not

** for each congener is MRL set separately

*** for aldrin and dieldrin combined

**** sum of heptachlor and heptachlor epoxide expressed as heptachlor

4. Results and discussion

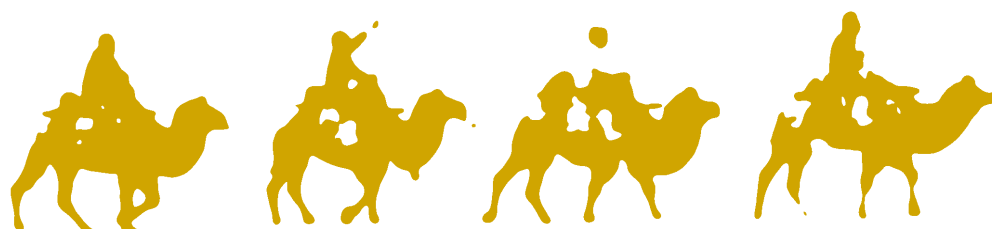
More precise HRGC-HRMS analyses were chosen for analyzing the contamination by dioxins and dioxin-like PCBs, as well as 6 indicator PCB congeners of camel milk samples. The same analysis was used also to analyze samples for 16 PAH congeners and other POPs: group of OCPs: hexachlorobenzene (HCB), other chlorobenzenes (TeCIB, 1,2,3,4-TeCIB, QCIB), hexachlorocyclohexanes (HCHs) and DDT and its metabolites, heptachlor, aldrine, octachlorostyrene, heptachloroepoxyde trans and cis, chlordane trans and cis, oxychlordane, metoxychlor and mirex. The sixth sample from Kyzyl Tube was not analysed for OCPs.

The results of the analyses for POPs by using HRGC-HRMS are summarized in Table 3. The Table 4 compares the results of the analyses for 6 PCB indicator congeners and Table 5 does the same for 12 DL PCB congeners. In Table 8 are results for PAH congeners. The results for OCPs on a fresh weight basis are summarized in Table 9 and compared with the respective EU limit values. The results for analyses of heavy metals content in collected camel milk samples are in Table 10.

Levels of analysed chlorobenzenes other than HCB (TeCIB, 1,2,3,4-TeCIB, QCIB), heptachlor, aldrine, octachlorostyrene, heptachloroepoxyde trans and cis, chlordane trans and cis, oxychlordane, metoxychlor, and mirex were below LOQ (= 0.1 ng g⁻¹ fat) in all samples, thus they are not included in the Tables 3 and 8.

Table 3: Summarized results of analyses for POPs for six pooled camel milk samples from Mangystau Region collected in 2015–2016.

Locality	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	KZ-M-16-6	EU standards/limits
Sample	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube	
Fat content	0.62 %	0.85 %	3.13 %	2.21 %	1.06 %	1.63 %	–
PCDD/Fs (pg WHO-TEQ g ⁻¹ fat)	0.45	1.33	1.30	0.01	0.31	0.24	2.50
DL PCBs (pg WHO-TEQ g ⁻¹ fat)	3.02	14.94	5.25	2.07	47.30	3.24	–
Total PCDD/F + DL PCBs (pg WHO-TEQ g ⁻¹ fat)	3.47	16.27	6.55	2.08	47.61	3.48	5.00
HCB (ng g ⁻¹ fat)	3.60	3.40	0.52	1.20	1.80	NA	–
6 PCB congeners (ng g ⁻¹ fat)	15.70	22.20	7.98	3.54	44.61	0.82	40.00
sum HCH (ng g ⁻¹ fat)	20.40	13.74	8.42	1.38	3.91	NA	–
sum DDT (ng g ⁻¹ fat)	1.74	2.32	0.28	0.57	0.88	NA	–
sum 16 PAHs (ng g ⁻¹ fat)	717.80	481.10	421.00	392.20	389.30	< LOQ	–



4.1 Unintentionally produced POPs: Dioxins (PCDD/Fs), dioxin-like PCBs (DL PCBs) and chlorobenzenes

Dioxins belong to a group of 75 polychlorinated dibenzo-p-dioxin (PCDD) congeners and 135 polychlorinated dibenzofuran (PCDF) congeners, of which 17 are of toxicological concern. Polychlorinated biphenyls (PCBs) are a group of 209 different congeners which can be divided into two groups according to their toxicological properties: 12 congeners exhibit toxicological properties similar to dioxins and are therefore often referred to as 'dioxin-like PCBs' (DL PCBs). The other PCBs do not exhibit dioxin-like toxicity but have a different toxicological profile and are referred to as 'non dioxin-like PCB' (NDL PCBs) (European Commission 2011). Levels of PCDD/Fs and DL PCBs are expressed in total WHO-TEQ calculated according to toxic equivalency factors (TEFs) set by WHO experts panel in 2005 (Van den Berg, Birnbaum et al. 2006). These new TEFs were used to evaluate dioxin-like toxicity in six samples of camel milk from Mangystau Region of Kazakhstan.

None of the six samples exceeded the EU and/or Russian MAC levels of PCDD/Fs congeners in milk, however, three out of the six samples exceeded the EU limit value for total TEQ of both PCDD/Fs and DL PCBs in milk (European Commission 2011).

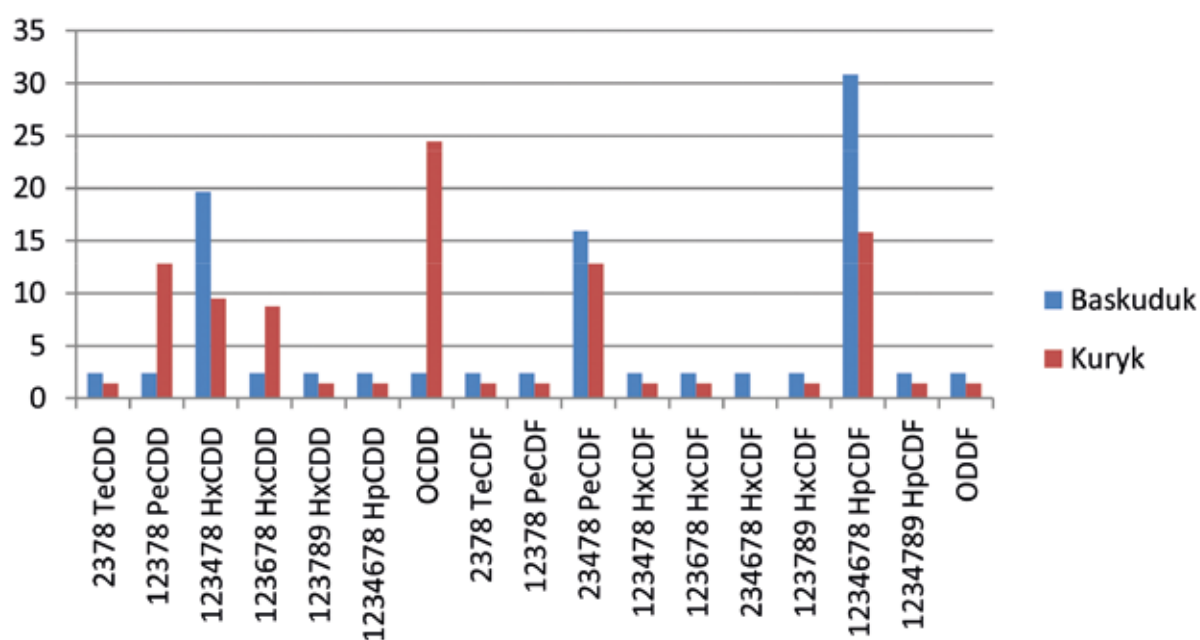


Figure 6: Graph showing PCDD/Fs patterns in camel milk samples from Baskuduk and Kuryk. Congeners with levels below LOQ were counted as half of LOQ in this graph. In % share on absolute level of 17 PCDD/Fs congeners in total.

Toxicity of DL PCBs has prevailed over PCDD/Fs congeners toxicity in all camel milk samples collected for this study. Sample from an individual lactating camel from Tauchik showed the highest level of DL PCBs (47.30 pg WHO-TEQ g⁻¹ fat). It was almost 23-times higher than the lowest level of DL PCBs (2.07 pg WHO-TEQ g⁻¹ fat) observed in a sample from Akshukur accompanied also by the lowest level of PCDD/Fs (0.01 pg WHO-TEQ g⁻¹ fat). Sample from Baskuduk also contained a high level of DL PCBs (14.94 pg WHO-TEQ g⁻¹ fat) accompanied by the highest level of PCDD/Fs of all six samples (1.33 pg WHO-TEQ g⁻¹ fat). A comparable level of PCDD/Fs was observed in the sample from Kuryk (1.30 pg WHO-TEQ g⁻¹ fat); however, the dioxin congeners patterns for these samples were different as is evident from the graph in Figure 6. It suggests a different source of camel milk contamination by dioxins for each sample or a combination of sources.

Total WHO-TEQ levels of PCDD/Fs and DL PCBs in samples collected for this study were much higher than in those collected by Konuspayeva, Faye et al. (2011 a) . The highest level of DL PCBs expressed in WHO-TEQ is more than 8.5-fold higher than the maximum level (5.33 pg WHO-TEQ g⁻¹ fat) reported by Konuspayeva, Faye et al. (2011 a) , although the levels of PCDD/Fs were reported within comparable range (0.53–1.49 pg WHO-TEQ g⁻¹ fat) or slightly higher than in this study.

In general, Konuspayeva, Faye et al. (2011) observed that the levels were higher in the region of Atyrau, and that *“this point could be linked to the importance of oil extraction in the area which is considered as the most risk for air and soil pollution in steppe regions around”*. Our findings are in agreement with that conclusion as we observed significantly higher levels of DL PCBs in particular.

The background levels for PCDD/Fs and DL PCBs in camel milk were not set up yet, and none of our samples can be considered to come from the environment without the influence of industrial or mining activity (see section 2.1). Although Konuspayeva, Faye et al. (2011) aimed in their study to establish background level for PCDD/Fs and DL PCBs in camel milk in Kazakhstan, no such level resulted from their study due to lack of information about potential sources of milk contamination by U-POPs. A more remote area with no industrial or mining activity and no potential influence of waste disposal needs to be sampled for camel milk in order to get some idea about real background levels of U-POPs in camel milk.

The lowest observed levels of total PCDD/Fs and DL PCBs were between 1.31 pg WHO-TEQ g⁻¹ fat in Konuspayeva, Faye et al. (2011) and 2.08 pg WHO-TEQ g⁻¹ fat in this study (detected in the sample from Akshukur). We believe that the background level for Kazakhstan can be provisionally established within the range of 2 – 3 pg WHO-TEQ g⁻¹ fat, which is between the lowest level in this report and the mean concentration for samples collected by Konuspayeva, Faye et al. (2011). For comparison of results in both studies see also Table 11.

HCB is also considered to be an unintentional product of POP (U-POP) in the same processes as dioxins and DL PCBs (Stockholm Convention on POPs 2008), although it is commonly measured together with other OCPs for which the hygienic limit values are established on fresh weight basis (see Table 2). HCB content in all samples was well below this limit value (see Table 9). Highest levels of HCB expressed per gram of fat in milk were detected in samples from Shetpe and Baskuduk.

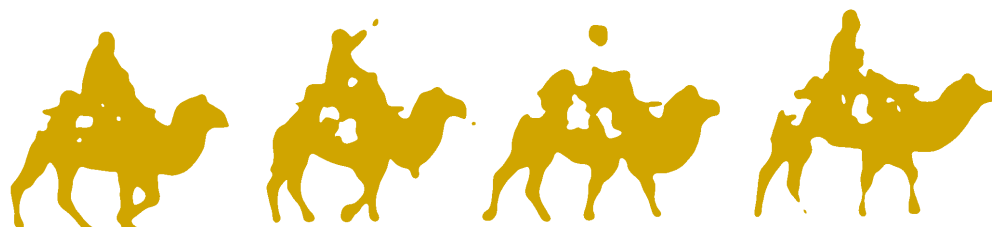
Another group of U-POPs is PAHs released, for example, during oil processing or as a result of incomplete combustion. Results of analyses for their congeners are discussed below in section 4.3.

4.2 Polychlorinated biphenyls (PCBs)

4.2.1 Indicator PCBs

Levels of 6 indicator PCB congeners correspond to the finding of high levels of DL PCBs and are much higher in comparison with the study by Konuspayeva, Faye et al. (2011 a) who reported for camel milk samples from Kazakhstan levels of 6 PCB congeners within the range of 0.60–17.44 ng g⁻¹ fat. Results of analyses for 6 individual PCB congeners for six samples collected within our research are summarized in Table 4 and graph in Figure 7. Sums of these congeners were in the range of 3.54–44.61 ng g⁻¹ fat. These are significantly higher levels than those found in previous studies. Mean of 15.8± 16.2 ng g⁻¹ fat is 2.5-times higher than 6.3 ± 2.7 ng g⁻¹ fat, respective median of 11.84 2 ng g⁻¹ fat is almost 2.5-times higher than 5.1 ± 2.0 ng/g⁻¹ fat reported by Konuspayeva, Faye et al. (2011 a); however, our research was done at smaller number of localities and the highest level was found in just one individual sample at one of selected locations. Samples were also taken in the season when excretion of certain congeners of PCBs was observed to be higher (Nurseitova, G. et al. 2014), however, Konuspayeva, Faye et al. (2011) observed higher levels of DL PCBs in spring samples compared to autumn samples.

Transfer of different POPs and their excretion by ruminants through milk can vary. Therefore, it is also difficult to follow specific congener profiles of certain sources of pollution in the milk of ruminants. This topic is further discussed in section 4.2.3.



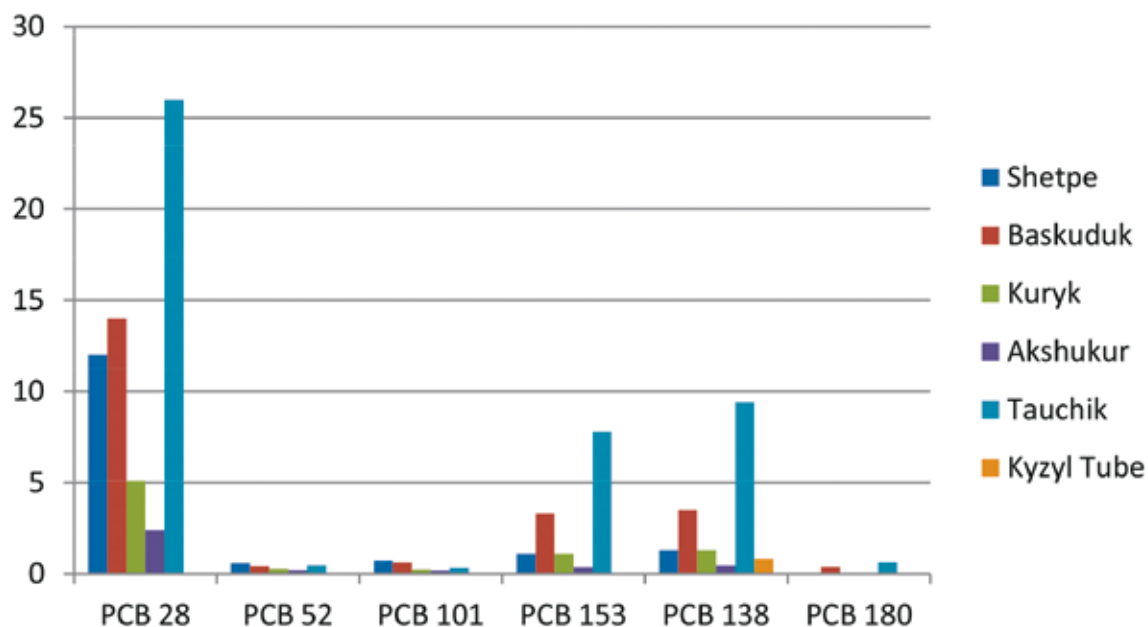


Figure 7: Indicator PCB or NDL PCB congeners patterns in six camel milk samples from Mangystau Region. Congeners below LOQ were considered "0" in this graph. In ng g⁻¹ fat.

Table 4: Summarized results of analyses for 6 PCB indicator congeners in six camel milk samples from Mangystau Region collected in 2015–2016 and their comparison with results of analyses presented in Konuspayeva, Faye et al. (2011 a). Levels are in ng g⁻¹ fat. Levels below LOQ were considered "0" for the calculation of the mean.

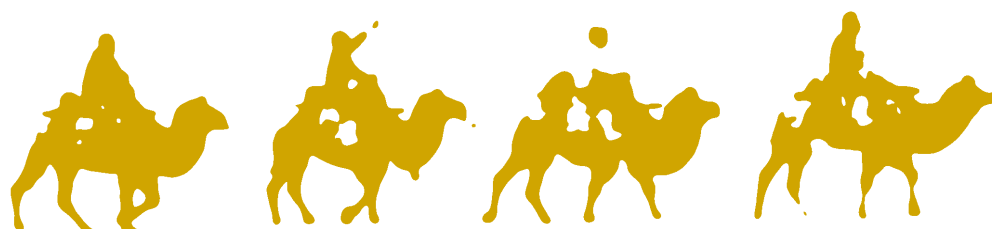
Sample	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	KZ-M-16-6		Konuspayeva, Faye et al. (2011)
Locality	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube	Mean	Mean from several locations
Fat content	0.62%	0.85%	3.13%	2.21%	1.06%	1.63%		
PCB 28	12.00	14.00	5.10	2.40	26.00	< 0.8	9.92	0.80
PCB 52	0.58	0.41	0.26	0.18	0.46	< 0.8	0.32	2.61
PCB 101	0.72	0.62	0.22	0.16	0.32	< 0.8	0.34	1.32
PCB 153	1.10	3.30	1.10	0.35	7.80	< 0.8	2.28	0.80
PCB 138	1.30	3.50	1.30	0.45	9.40	0.82	2.80	0.52
PCB 180	< 0.3	0.37	< 0.2	< 0.05	0.63	< 0.8	0.17	0.21
Sum of 6 PCB	15.70	22.20	7.98	3.54	44.61	0.82	15.81	6.26

4.2.2 Dioxin-like PCBs (DL PCBs)

The mean levels of DL PCBs concentrations in camel milk samples presented in this study were almost 10 times higher than the levels found in the previous study by Konuspayeva, Faye et al. (2011 a). For more detailed comparison of these studies see Table 5. Levels of DL PCB congeners are also shown in graph in Figure 8. Results for DL PCBs were already discussed in section 4.1.

Table 5: Summarized results of analyses for 12 DL PCB congeners in six camel milk samples from Mangystau Region collected in 2015–2016 and their comparison with results of analyses presented in Konuspayeva, Faye et al. (2011 a). Levels are expressed in ng g⁻¹ fat. Levels below LOQ are shown as “0”.

Sample	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	KZ-M-16-6		Konuspayeva, Faye et al. (2011 a)
Locality	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube	Mean	Mean from several locations
PCB 81	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
PCB 77	0.150	0.046	0.025	0.011	0.028	0.003	0.044	0.000
PCB 126	0.029	0.140	0.051	0.020	0.450	3.200	0.648	0.017
PCB 169	0.000	0.013	0.000	0.000	0.010	0.000	0.004	0.001
PCB 123	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.014
PCB 118	2.300	11.000	2.900	1.200	38.000	0.036	9.239	0.974
PCB 114	0.000	0.000	0.000	0.050	1.100	0.000	0.192	0.041
PCB 105	1.100	5.000	1.400	0.680	20.000	0.000	4.697	0.509
PCB 167	0.000	0.730	0.250	0.100	1.800	0.000	0.480	0.038
PCB 156	0.000	1.300	0.380	0.130	4.100	0.000	0.985	0.089
PCB 157	0.000	0.240	0.000	0.058	1.400	0.000	0.283	0.022
PCB 189	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.006
Sum 12 DL PCB	3.579	18.469	5.006	2.249	66.888	3.239	16.572	1.711



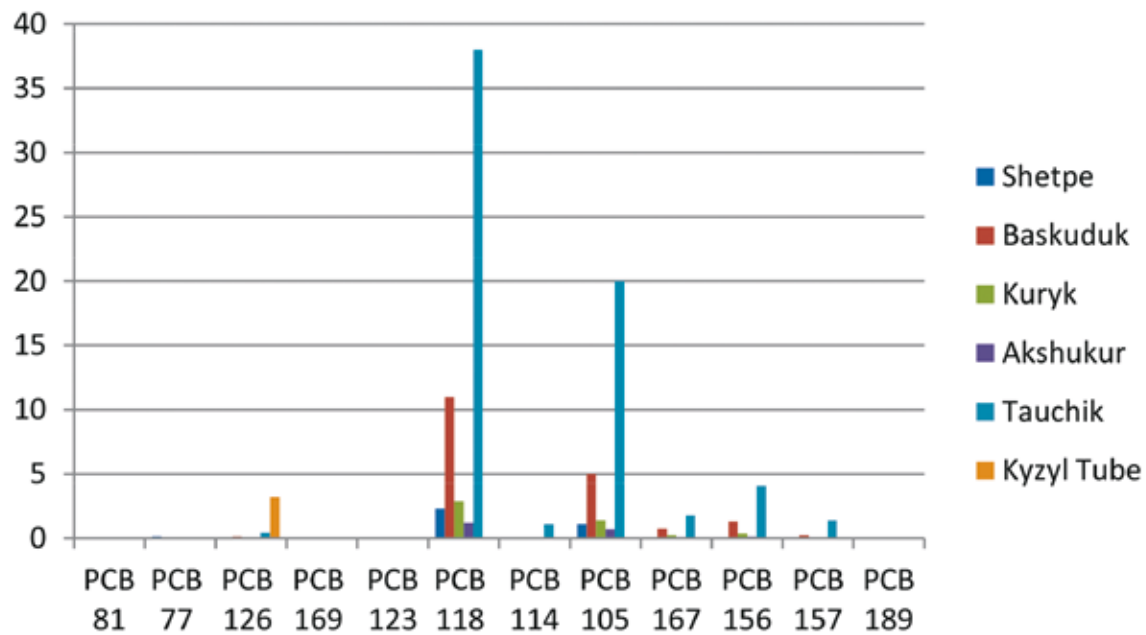


Figure 8: DL PCB congeners patterns in six camel milk samples from Mangystau Region. In $ng\ g^{-1}$ fat.

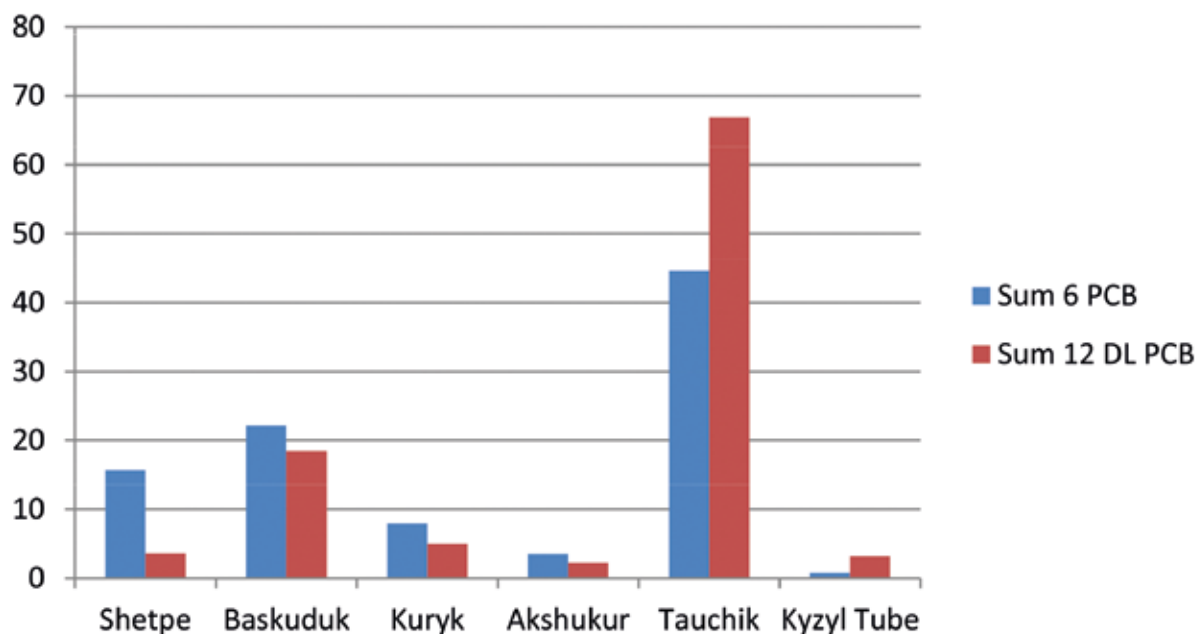


Figure 9: Comparison of total indicator PCBs and DL PCBs in six camel milk samples from Mangystau Region. In $ng\ g^{-1}$ fat.

4.2.3 Intake of PCB congeners and other POPs by different species of ruminants

To identify sources of pollution of certain dairy products, such as milk, it is important to follow presence and quantification of certain congeners as some industrial sources can show specific profiles comparable to fingerprints in forensic science. Therefore, it is also important to look at the kinetics of each chemical compound. Applying this approach on the milk of ruminants seems difficult because their intake of different POPs, their congeners, and metabolites can vary. Specific POPs congeners patterns recognized in soil, dust or other environmental matrices can be significantly changed in milk due to the specific metabolism of ruminants. McLachlan (1993), for example, concluded that: *"the contaminant absorption in the cow and hence the carry-over rate (COR) of persistent compounds was found to be a function of K_{ow} ¹², with approximately constant values up to a log K_{ow} of 6.5 and thereafter rapidly decreasing absorption with increasing lipophilicity of the contaminant"*.

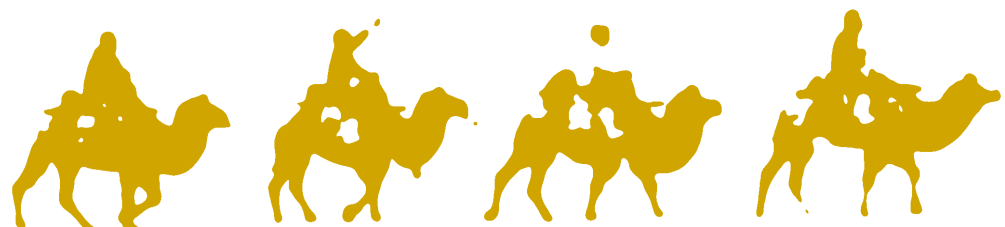
Costera, Feidt et al. (2006) studied the feed to milk transfer of 17 PCDD/Fs and 18 PCBs in goats exposed to a 10-week long-term intake of contaminated hay collected in the vicinity of a municipal waste incinerator. They concluded that: *"For PCDD/Fs, 2,3,7,8-TCDD appeared as the compound having the highest COR (38.8 %). Within dioxin-like-PCBs, the highest COR were found at a similar level (higher than 80 %) for PCBs 105, 118 and 157. Concerning indicator-PCBs, COR ranged from 5 % (PCB 101) to more than 40 % (PCBs 118, 153 and 180). The intensity of this transfer appeared to be a function of physico-chemical properties (chlorination or log K_{ow}) of the molecules and their metabolic behaviour."* Rychen, Jurjanz et al. (2008) confirmed these findings: *"Transfer rates to milk have been established: for PCBs, the rate of transfer varies from 5 % to 90 %, for PCDD/Fs from 1 % to 40% and for PAHs from 0.5 % to 8 %."*

Nurseitova et al. (2014) focused on the transfer of PCB indicator congeners and DDT metabolite DDE and their excretion in camel milk in Kazakhstan. They exposed 3 Bactrian camels to controlled doses of PCBs and DDT for 56 days following 4 months of decontamination period at the end of summer and autumn. The trial has been carried out in Suzak region of South Kazakhstan, close to the Moyun-Kum desert with approximately 100 mm of annual rainfall.

Principal findings of their study were summarized as follows: *"The daily excreted amounts of tri- or tetrachlorinated congeners (PCB28, PCB52) were not statistically increased during the exposure period in comparison to the previously measured background levels. Nevertheless, the much lower excretion amounts after the decontamination period let suppose an overestimated background level, possibly linked to environmental presence to these congeners. At the end, the daily excreted amounts of these low chlorinated congeners increased again in autumn when animals reconstructed fat reserves in humps for winter. This would suggest an intermediate storage of fat (and lipophilic compounds as POPs) in another tissue (for ex. visceral or perirenal fat which could represent up to 30 % of the fat storage in camel; Faye et al. (2001) before being reinserted in blood circulation what would allow to transport them to humps but also to the udder and therefore increase their excretion in milk.*

Penta- (PCBs 101 and 118), hexa- (PCBs 138 and 153) and heptachlorinated (PCB 180) congeners increased significantly during the exposure period (table 1). Then the excretion decreased significantly during two months without reaching again completely the numeric values of the background levels of daily excretion (Table 6). The excretion of these congeners also rose on, right up from September in the same time as fat storage in humps grew on. To our knowledge, such phenomenon has not been reported in other mammals but Konuspayeva, Jurjanz et al. (2011) reported amazingly low or no contamination of indicator PCBs in camel milk of areas where at least background contamination was likely."

¹² " K_{ow} " or " $\log K_{ow}$ " = concentration in octanol phase/Concentration in aqueous phase. Octanol/water partition coefficient is very important index in biological, toxicological and environmental area. The animal bodies are made from water and lipids. If you know the distribution ratio of the chemicals to the octanol, you can estimate bio accumulations. Yamamoto, H. (2011, 28-06-2011). "Properties Estimation: $\log P$, $\log K_{ow}$: Octanol-water partition coefficient." 2016y.



The calculated carry over rates (CORs) based on plateau excretion at the end of the exposure period from a study by Nurseitova et al. (2014) in comparison with some other studies focusing on ruminants are shown in Table 6.¹³

Table 6: Carry over rate (%) of indicator PCBs and DDT in camel milk in comparison to milk of other species in the literature by Nurseitova et al. (2014).

	Nurseitova et al. (2014)	McLachlan (1993)	Thomas et al. (1999)	Costera, Feidt et al. (2006)	Ounnas, Feidt et al. (2010)
specie	3 Bactrian camels	1 cow	5 cows	3 goats	3 goats
Duration of exposure (days)	56	permanent	permanent	70	45
PCB 28	nd	nd	nd	25	nd
PCB 52	6	nd	< 2	10	nd
PCB 101	2	nd	4	5	nd
PCB 118	19	33	94	85	59
PCB 138	14	63	69	41	36
PCB 153	13	78	75	45	nd
PCB 180	71	63	63	55	nd
DDT (DDE included)	3.6	4	NA	NA	NA

Nurseitova et al. (2014) commented on results presented in Table 6 as follows: “Although hierarchic order between the CORs of congeners seems to be the same, the general transfer rate is clearly lower than in other studies. Non-coplanar, low chlorinated PCBs (i.e. 52 and 101) were weakly transferred (<10 %) into camel milk as previously reported in cows and goats. Contrarily, the only mono-ortho congener (PCB 118) was transferred into camel milk to a much lesser extent (19 %) than in goats and cows. The same tendency has been observed for hexachlorinated indicator PCBs: transfer rates seem clearly lower than in other ruminants and finally, the heptachlorinated PCB 180 has been transferred at a rate at least as high as in other ruminants. At the end, the transfer rate of DDT in our camels seems low but consistent to this reported by McLachlan (1993).

That means that low chlorinated compounds but also heptachlorinated PCB 180 have similar transfer patterns in milk from camels in comparison to other ruminants coplanar PCB 118 and hexachlorinated congeners are less transferred. This difference could be partially due to the difference in exposure dose between the studies. Indeed, we used much higher exposure doses (2.2 µg kg⁻¹ BW & day) in our camels in comparison to studies on goats using only 0.03 µg kg⁻¹ BW & day (Costera, Feidt et al. 2006, Ounnas, Feidt et al. 2010) during a similar duration of exposure (56 days for our camels in comparison to 45 to 70 days in goat studies). We cannot exclude that some transfer mechanisms, especially for highly transferred compounds, reached a saturation of absorption what would reduce mathematically the transfer rates.”

¹³ Carry over rate as defined in Costera, A., C. Feidt, P. Marchand, B. L. Bizec and G. Rychen (2006). “PCDD/F and PCB transfer to milk in goats exposed to a long-term intake of contaminated hay.” Chemosphere 64(4): 650-657.: „COR = [m.fy/f. F].100. COR is the carry-over rate (%); m is pollutant concentration in milk fat at steady state (ng kg⁻¹); fy is fat yield (g d⁻¹); f is pollutant concentration in diet (ng k g⁻¹ DM); F is daily feed intake (g d⁻¹).

4.2.4 PCB congeners pattern in camel milk samples from Mangystau Region

The dominance of the indicator congeners in camel milk samples presented in this report was observed in the following order: PCB 28 > PCB 138 > PCB 153 > (PCB 101; PCB 52) > PCB 180. This can be influenced by both 1) specific metabolism of lactating camels as described above, and 2) level of environment contamination. The order of dominant congeners observed in camel milk samples in this report is partly contrary to findings of their CORs by Nurseitova et al. (2014): the level of PCB 180 was very low, and the concentrations of PCB 28 congener were found very high. It is also different in comparison with findings by Konuspayeva, Faye et al. (2011 a) who found higher levels of PCB 52 and 101, and at the same time much lower level of PCB 28 congener among NDLCB congeners than observed in this report (see Table 4). This can point to a specific pattern of PCB contamination in the areas where some camels are bred. This pattern is characterized by the prevalence of PCB 28 congener.

Proportion of DL PCBs congeners present in camel milk samples collected for our study was similar to that found by Konuspayeva, Faye et al. (2011 a), and "PCB 105 and PCB 118 appeared to be present at higher concentrations in camel milk (>80% of the sum of the 12 DL-PCBs)".

All three dominant NDLCB congeners (PCB 28, 138 and 153) as well as two dominant NDLCB congeners (PCB 118 and 105) in our camel milk samples were present in high levels also in commercial mixtures of PCB oils produced by Sovtol in the former USSR (Brodsky, Evdokimova et al. 2005). A certain amount of capacitors containing PCB oils was reported also from Mangystau Region (323) according to the National Implementation Plan of the Stockholm Convention for Kazakhstan (Republic of Kazakhstan 2009). Company KazMunaiGas operating in this region (close to Tauchik – see section 2.1.5) was reported in 2009 to also hold 682 capacitors containing PCB oils (Republic of Kazakhstan 2009)¹⁴.

4.2.5 Potential contribution of contaminated camel milk to total daily intake of PCBs and PCDD/Fs by local residents in Mangystau Region

People may be exposed to PCBs by inhaling contaminated air and ingesting contaminated water and food. Food is a major route of human exposure to PCBs. Camel milk and/or shubat represent a significant part of the diet for Kazakhstani population. Therefore, it is important to assess risks related to consumption of camel milk contaminated by PCBs. This can be done by calculating the daily intake of PCBs and PCDD/Fs via the consumption of camel milk from analysed samples.

Calculation of daily intake levels was made by using following formula:

$$DI_{adult} = (((C \cdot F \%) / 100) \cdot (240 \cdot 1030)) / 365 / 70;$$
$$DI_{child} = (((C \cdot F \%) / 100) \cdot (240 \cdot 1030)) / 365 / 35,$$

where DI = daily intake; C = concentration of certain group of chemicals (6 PCB congeners, PCDD/Fs, DL PCBs), and F% = fat content in sample

For calculation, we used the following data: the consumption of milk 240 liters per person per year = 247.2 kg of milk/year considering the density of milk to be approx. 1 liter = 1030g (Nurseitova, Konuspayeva et al. 2014). We used the body weight of 70kg for an adult and 35kg for a child (approximately 10 years old). Results of daily intake calculations for samples from different localities, as well as the mean and median for the whole set of samples, can be found in Table 7. However, this is an estimate based on available (limited) data mentioned above which can give some idea about the potential risk of contamination of food by PCBs, dioxins and dioxin-like PCBs in Mangystau Region. It does not draw full picture of real health risks.

¹⁴ The Atyrau refinery (ANPZ) owned by KazMunaiGas also was reported in 2016 to hold 4 PCB-transformers. Химатчов, Р. (2016). "В одном векторе с защитой природы края, страны, планеты. (https://azskmg.kz/upload/iblock/61d/22_.pdf). " Новатор (Novator) 22: 4.

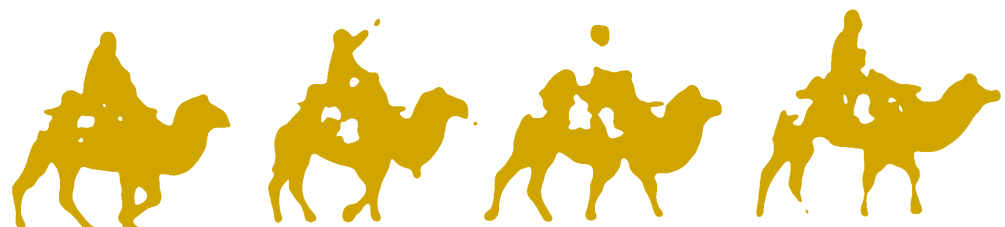


Table 7: Calculation of potential daily intake of PCBs and dioxins (PCDD/Fs) by consumption of contaminated camel milk/shubat in Mangystau Region based on available data from this study. The calculation was made for each location/pooled sample as well as for the whole set of samples presented in this study. (Explanation for calculation is in text above).

Locality	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube	Mean/ median
Data for samples of shubat and/or raw camel milk from Kuryk							
Fat content	0.62%	0.85%	3.13%	2.21%	1.06%	1.63%	1.58%/1.35%
Sum 6 PCB	15.7	22.2	7.98	3.54	44.61	0.82	15.81/11.84
PCDD/Fs + DL PCBs	3.47	16.27	6.55	2.08	47.61	3.48	13.24/5.02
Daily intake of toxic chemicals from camel milk/shubat per kg of body weight by adults (Dadult)							
Sum of 6 PCB	0.94	1.83	2.42	0.76	4.58	0.13	2.42/1.55
PCDD/Fs + DL PCBs	0.21	1.34	1.98	0.45	4.88	0.55	2.02/0.66
Daily intake of toxic chemicals from camel milk/shubat per kg of body weight by children approx. 10 years of age (body weight 35 kg); (Dchild)							
Sum of 6 PCB	1.88	3.65	4.83	1.51	9.15	0.26	4.83/3.09
PCDD/Fs + DL PCBs	0.42	2.68	3.96	0.89	9.76	1.10	4.05/1.31

There is almost no data available on the total dietary intake of PCBs and/or dioxins in Kazakhstan; therefore, it is difficult to evaluate how large proportion represents the intake of PCBs of PCDD/Fs by camel milk. It is certain that Kazakhstani population is not exposed to PCBs only through the consumption of camel milk as high levels of PCBs were also observed, for example, in free range chicken eggs in other parts of country (Petrлік, Kalmykov et al. 2015), as well as significant levels of PCBs detected in some fish samples (Šír 2015). The proportion of milk in total food consumption in Kazakhstan in 2007 was close to 34% of total food basket per day according to the World Atlas – Food Security data (Knoema 2012),¹⁵ but there is no available data on the contamination of other foods forming the Kazakhstani diet by dioxins and dioxin-like compounds. The consumption of meat, which can also contain significant levels of PCBs, represents an important part of the diet (over 8% according to Knoema, 2012). For example, UNECE assumed that the main source of high exposure to organochlorine compounds including PCBs in Kyzylorda Region is probably the consumption of contaminated animal fat from beef, goat, chicken and milk products (UNECE 2000).

In 1978, the estimated dietary intake of PCBs by adults in the USA was 27 ng kg⁻¹ body weight per day, but it declined to 0.5 ng kg⁻¹ body weight per day in 1982–1984 and <1 ng kg⁻¹ body weight per day for the period of 1986–1991 (WHO and IPCS 2003). Inhabitants of Netherlands consume through diet 5.6 ng of indicator congeners of PCBs per kg bw daily (Baars, Bakker et al. 2004). The PCBs intake through the consumption of shubat/camel milk from three of six samples presented in this study could exceed the total daily intake of PCBs by an average American in 1990; the mean intake for all samples can reach half of the total PCBs dietary intake in Netherlands.

¹⁵ The food consumption refers to the amount of food available for human consumption as estimated by the FAO Food Balance Sheets. However the actual food consumption may be lower than the quantity shown as food availability depending on the magnitude of wastage and losses of food in the household. Food consumption per person is the amount of food, in terms of quantity, for each individual in the total population. Consumption of milk relates to the amount of milk including the fresh milk equivalent of dairy products, but excluding butter.

There are different values for Tolerable Daily Intake (TDI) for PCBs: in the USA, TDI is set at a level of 3–5 ng kg⁻¹ body weight per day for adults and 2–12 ng kg⁻¹ body weight per day for children (GreenFacts 2016). In Mangystau Region, these levels can almost be reached just by daily consumption of camel milk/shubat. This is worrying as it is not only shubat, what contributes to the total daily intake of PCBs of local people.

World Health Organization (WHO) established less strict TDI for PCBs at the level of 20 ng kg⁻¹ body weight per day (averaged over the whole life); (WHO and IPCS 2003, GreenFacts 2016). Levels of DI for 6 indicator PCBs from camel milk in this study represents from 1/100 (sample from Kyzyl Tube) to one half (sample from Tauchik, calculated for children) of this TDI (see Table 7).

TDI for PCDD/Fs and DL PCBs was set by WHO experts within the range of 1–4 pg WHO-TEQ kg⁻¹ of body weight per day (van Leeuwen, Feeley et al. 2000). Daily consumption of camel milk/shubat from Tauchik can result in exceeding of this TDI; similarly, the median of concentrations in the whole set can exceed 1–4 pg WHO-TEQ kg⁻¹ of body weight per day for children (see Table 7). Loutfy, Fuerhacker et al. (2006) developed a broader comparison of daily intakes of PCDD/Fs and DL PCBs in different countries. Based on the comparison of our data with published daily intakes of dioxins and DL PCBs in some other countries in the end of 1990s, the consumption of camel milk represented by samples from Mangystau Region can lead to the equivalent of full exposure to these compounds in some countries, e.g. the USA, UK or Belgium. We must note that this comparison is not at the same level of data complexity as we compare data for one type of food calculated for a limited set of measurements with the more overall picture. However, it can help us to imagine the scale of the problem with DL PCBs contamination in Mangystau Region, particularly at certain sites, such as Tauchik, Baskuduk, and Kuryk.

Hooper, Chuvakova et al. (1999) observed high levels of TCDD in food in areas where pesticides are applied in the cotton-growing region of southern Kazakhstan, and daily intake by women reached 175 pg WHO-TEQ per day (2.5 pg WHO-TEQ kg⁻¹ of body weight for a person weighing 70kg). The mean value for daily intake of PCDD/Fs and DL PCBs from camel milk in our study is close to that level and the median value exceeds one quarter of that level. For the sample from Tauchik, the daily intake of dioxin-like compounds in camel milk is almost double the daily intake of these compounds by women from the cotton-growing region of southern Kazakhstan (Hooper, Chuvakova et al. 1999). The difference is in the prevalence of chemicals in total WHO-TEQ: DL PCBs prevail in camel milk samples from the Mangystau Region (see Table 11), while PCDD/Fs prevailed in the study by Hooper, Chuvakova et al. (1999) from the cotton-growing region.

4.3 Polycyclic aromatic hydrocarbons (PAHs)

Results of analyses for 16 PAH congeners in our six camel milk samples are summarized in Table 8. Six out of sixteen PAH congeners (benzo[a]anthracene, chrysene, benzo[a]pyrene, indeno[1,2,3-c,d]pyrene, benz[g,h,i]perylene and dibenz[a,h]anthracene) were below LOQ in all six camel milk samples; this is common for ruminants' milk samples in general (Jurjanž, Rychen et al. 2008).

Konuspayeva, Jurjanž et al. (2011) reported the prevalence of phenanthrene, fluoranthene and pyrene in camel milk samples from southwestern part of Kazakhstan, however, they analysed samples for 13 PAH congeners not including naphthalene. Phenanthrene is second dominant congener in all six camel milk samples collected for our study while naphthalene is the most dominant as is also evident from the graph in Figure 10.

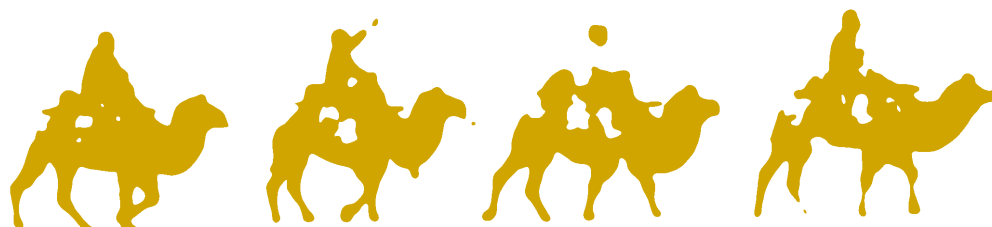


Table 8: Summarized results of analyses for 6 PCB indicator congeners in six camel milk samples from Mangystau Region collected in 2015–2016 (in ng g⁻¹ fat; except for the last two rows where levels are in ng g⁻¹ fresh weight of shubat/milk).

Sample	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	KZ-M-16-6
Locality	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube
Fat content	0.62%	0.85%	3.13%	2.21%	1.06%	1.63%
Naphthalene	410	300	270	160	250	< 30
Acenaphthylene	18	20	15	23	15	< 30
Acenaphthene	13	12	< 5	12	6	< 30
Fluorene	25	20	17	17	15	< 30
Phenanthrene	200	110	100	130	88	< 30
Anthracene	27	12	11	30	8	< 30
Fluoranthene	15	7	8	9	7	< 30
Pyrene	< 8	< 6	< 5	5	< 5	< 30
Benzo[a]anthracen	< 8	< 6	< 5	< 2	< 5	< 30
Chrysene	< 8	< 6	< 5	< 2	< 5	< 30
Benzo[b]fluoranthene	10	< 6	< 5	4	< 5	< 30
Benzo[k]fluoranthene	< 8	< 6	< 5	2	< 5	< 30
Benzo[a]pyrene	< 8	< 6	< 5	< 2	< 5	< 30
Indeno[1,2,3,-c,d]pyrene	< 8	< 6	< 5	< 2	< 5	< 30
Benz[g,h,i]perylene	< 8	< 6	< 5	< 2	< 5	< 30
Dibenz[a,h]anthracene	< 8	< 6	< 5	< 2	< 5	< 30
Sum of 16 PAHs	718	481	421	392	389	< LOQ
Sum of 4 PAHs	10	< LOQ	< LOQ	4	< LOQ	< LOQ
Sum of 16 PAHs in ng g ⁻¹ fresh milk	4.45	4.09	13.18	8.67	4.13	< LOQ
Sum of 15 PAHs ¹ in ng g ⁻¹ fresh milk	1.91	1.54	4.73	5.13	1.48	< LOQ

¹ Naphthalene is excluded for better comparison with the previous study by Konuspayeva, Jurjanz et al. (2011).

Levels of individual PAH congeners were few tenths of ng per milliliter of fresh milk in samples collected by Konuspayeva, Jurjanze et al. (2011) according to the graph in their study. It is approximately comparable to tenths of ng per gram of fresh milk if we consider that one liter of camel milk weights 1030 grams (Nurseitova, Konuspayeva et al. 2014) or less in the case of shubat. Table 8 shows calculated total sums of 16 PAH congeners measured in camel milk in this study as well as sums of 15 PAHs without naphthalene, which Konuspayeva, Jurjanz et al. (2011) did not analyze in their study.

It is evident that total 15 PAH congeners are 1.5 or more ng g^{-1} in fresh camel milk/shubat samples collected in Mangystau Region for our research. The highest levels of 15 PAHs (without naphthalene) were found in samples from Kuryk and Akshukur due to the high content of fat as well as lower levels of naphthalene. Levels of 15 PAHs in these two samples are most likely several times higher than those observed by Konuspayeva, Jurjanz et al. (2011), who concluded that *“the oil forages in Atyrau do not seem to affect PAH contents in milk of this area”*. In opposition to their conclusion, we assume that the most likely reason for such high levels of PAHs in our milk samples is the usage of oil in the cement kiln in Shetpe and the oil industry concentrated in the Mangystau Region. Additionally, the levels of PAHs in camel milk samples from the Mangystau Region are higher than those observed in a previous study from Kazakhstan. Different levels of PAHs in the sample from Kyzyl Tube compared to the other five samples can be explained by different time of sampling: sample in Kyzyl Tube was collected in spring, while the other samples were obtained in autumn when metabolism of camels is changing (see also section 4.2.3); however, fewer pollution sources in the vicinity of Kyzyl Tube may also play the role. Different time of sampling in this study, as opposed to the research of Konuspayeva, Jurjanz et al. (2011), could partly explain the difference in results.

A sample of milk from Shetpe was the sample most contaminated by 16 PAHs including naphthalene when considering the value per gram of fat, with the levels significantly higher than in other samples (see Table 8). The lowest level was detected in the sample from Kyzyl Tube, however, much higher LOQ value for all congeners for this sample has to be taken into account.

PAHs concentrations in samples from the Mangystau Region are also one order higher than those observed in cow’s milk by Grova, Laurent et al. (2000) and Grova, Feidt et al. (2002) in Italy.

N. Grova and her team also observed that *“PAH distributions in milk show higher relative concentrations of naphthalene, and the absence of high molecular weight PAHs”* and concluded that *this result could be partly explained by the higher water-solubility of naphthalene favouring its selective concentration during digestion”* (Grova, Laurent et al. 2000). Distribution of PAH congeners in samples of

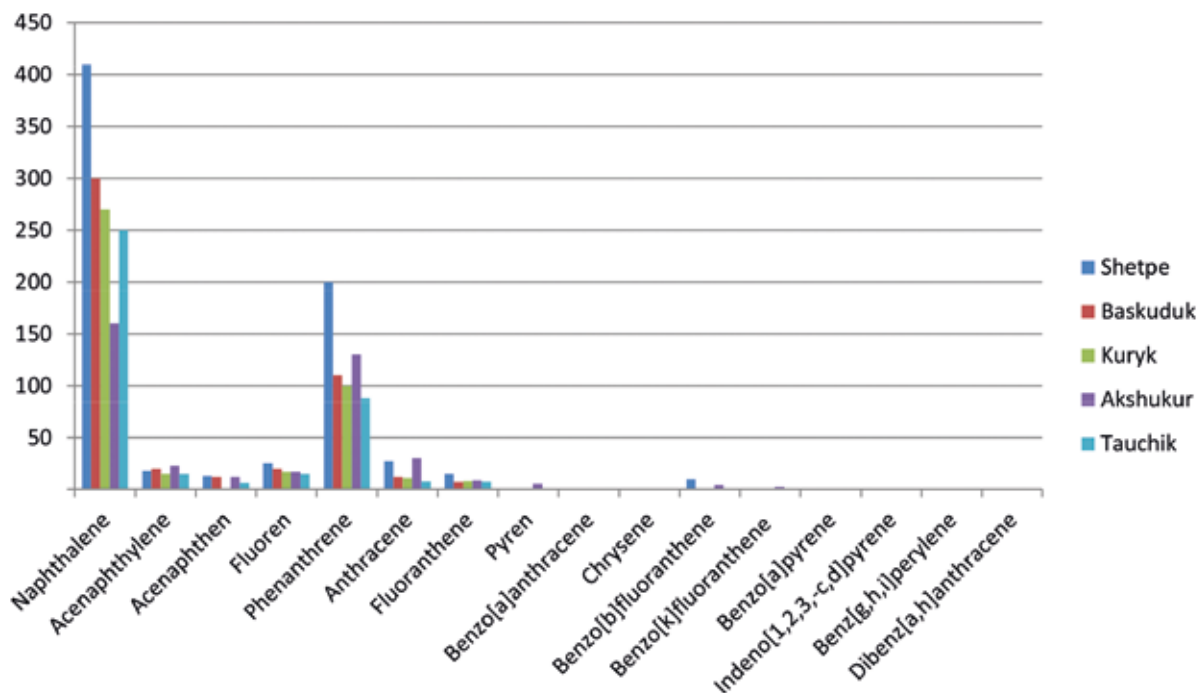
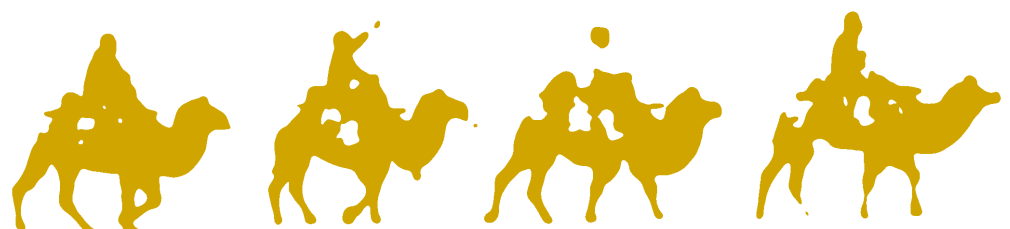


Figure 10: Graph showing 16 PAHs congener patterns in 5 camel milk samples. All PAH congeners were below LOQ in the sixth milk sample from Kyzyl Tube. Data are in ng g^{-1} fat.



ruminants' milk seems to mainly depend on their specific metabolism and less, if at all, on specific PAH congeners patterns of particular sources of pollution. Grova, Rychen et al. (2006), in the study based on experiment with goat's milk, concluded that "(1) benzo(k)fluorene, benzo(a)pyrene and benzo(g,h,i) perylene were not detected in the milk; (2) unexpectedly, the concentration of fluorene, phenanthrene, anthracene, fluoranthene, pyrene, and chrysene did not change with time".

We estimated that the daily intake of naphthalene by consumers of camel milk in Mangystau Region would be between 20 and 80 ng kg⁻¹ body weight per day for adults and possibly double for children¹⁶. This is still well below the TDI set, for example in Canada (Haffield Consultants 2008) or the USA (Haffield Consultants 2008 a), at the level 0.02 mg kg⁻¹ body weight (= 20,000 ng kg⁻¹ body weight), despite the fact that the naphthalene levels in camel milk samples from the Mangystau Region were much higher than in other studies focusing on PAHs content in milk. Nevertheless, high levels of PAHs in camel milk samples indicate the serious contamination of the environment in Mangystau Region by this group of chemicals in comparison with other countries (Grova, Laurent et al. 2000, Grova, Feidt et al. 2002) or regions in Kazakhstan (Konuspayeva, Jurjanz et al. 2011). A very low transfer rate of PAHs into the milk of ruminants – from 0.5 to 8% according to Rychen, Jurjanz et al. (2008) – needs to be taken into account, meaning that the milk of ruminants does not represent a major contributor to overall exposure to PAHs.

4.4 Organochlorinated pesticides (OCPs)

The EU limits for pesticide residues, including OCPs in milk, are set per fresh weight of milk. The comparison of OCPs content in five samples of camel milk from Kazakhstan is in Table 9. Neither Kazakhstani nor the EU limits were exceeded in any of the samples. Lindane (gama-HCH) reached a quarter of the EU limit value in the sample from Kuryk. In general, Lindane also showed the highest levels from all the OCPs analysed in samples, as shown in Table 9.

Table 9: Summarized results of analyses for OCPs in five pooled camel milk/shubat samples from Mangystau Region collected in 2015–2016 (note: the sample from Tauchik came from a single camel). The EU limit values are shown for comparison. These results are expressed in ng g⁻¹ fresh weight because the EU limits for OCPs are set for fresh weight.

Sample number	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	
Fat content in sample	0.62%	0.85%	3.13%	2.21%	1.06%	
OCPs in ng g ⁻¹ fresh weight	Shetpe	Baskuduk	Kuryk	Alkshukur	Tauchik	EU standards/limits
HCB	0.022	0.029	0.016	0.027	0.019	10
alfa-HCH	0.013	0.008	0.016	0.004	0.004	4
beta-HCH	0.008	0.006	0.004	0.009	0.008	3
gama-HCH (lindane)	0.105	0.101	0.244	0.018	0.028	1
sum 4 DDT metabolites	0.011	0.020	0.009	0.013	0.009	40

¹⁶ The method of calculation was the same as demonstrated for other POPs in section 4.6.

High levels of OCPs were not detected also in previous studies (Konuspayeva, Jurjanz et al. 2011). An analysis of pesticides showed the presence of HCHs (beta, delta and, only in the Kyzylorda Region, also gamma HCH). DDT was found in milk from the Kyzylorda Region at the level of 0.8 ng g⁻¹ (Konuspayeva, Jurjanz et al. 2011), which was much higher than in this study.

4.5 Heavy metals

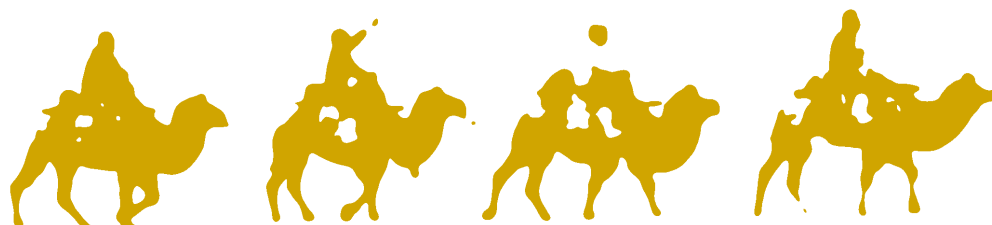
Results of analyses of camel milk/shubat samples from six localities in Mangystau Region are summarized in Table 10. Results for mercury, cadmium, chromium and arsenic were below LOQ in all analysed camel milk samples.

Levels observed for zinc are comparable with the study by Konuspayeva, Jurjanz et al. (2011), with the exception of the sample from Kyzyl Tube with the zinc level almost three times higher. For five samples collected in autumn 2015 (KZ-M-15), these levels were lower compared to the mean level observed for shubat by Konuspayeva, Faye et al. (2009). Two samples, from Baskuduk and Kyzyl Tube, exceeded the maximum acceptable level for milk according to the Russian legislation (see Table 2). Zinc content in the sample from Kyzyl Tube was almost 3 times higher than the limit set up for milk. The most likely source of contamination is the mining activity near Kyzyl Tube (see section 2.1.6).

The levels of lead and cadmium measured in our samples were lower than those found in camel milk from the broader area in Kazakhstan by Konuspayeva, Jurjanz et al. (2011).

Table 10: Summarized results of analyses for heavy metals for six pooled camel milk samples from Mangystau Region collected in 2015–2016 and their comparison with results of analyses presented in Konuspayeva, Faye et al. (2009) and Konuspayeva, Jurjanz et al. (2011). Levels are expressed in mg kg⁻¹ fresh weight.

Sample	KZ-M-15-1	KZ-M-15-2	KZ-M-15-3	KZ-M-15-4	KZ-M-15-5	KZ-M-16-6	Limits	Konuspayeva, Jurjanz et al. (2011); range milk/shubat	Konuspayeva, Faye et al. (2009); mean values milk/shubat
Locality	Shetpe	Baskuduk	Kuryk	Akshukur	Tauchik	Kyzyl Tube			
Mercury	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.005	–	
Lead	<0.004	<0.004	<0.004	<0.004	<0.004	0.008	0.1	<0.01–0.06/ <0.01–0.06	0.025/0.007
Cadmium	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.03	<0.001–0.003/ <0.001–0.003	
Copper	0.03	0.08	0.03	0.02	0.02	0.07	0.4–1.4	<0.05–0.07/ <0.05–0.06	0.07/0.16
Manganese	0.03	0.04	0.04	0.02	0.04	0.07	–	–	0.084/0.088
Chromium	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	–	–	
Zinc	3.06	5.28	4.58	3.11	3.56	14.1	5	4.07–5.31/ 4.16–6.50	5.16/7.212
Arsenic	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	0.05	–	<0.1/<0.1
Aluminium	3.00	0.50	<0.10	<0.10	0.20	0.60	–	–	



4.6 Discussion on the levels of camel milk contamination and its potential sources in Mangystau Region

Our study followed a number of previous studies analyzing the content of toxic chemicals in camel milk from several regions of Kazakhstan (Diacono, Faye et al. 2008, Meldebekova, Konuspayeva et al. 2008, Konuspayeva, Faye et al. 2009, Konuspayeva, Faye et al. 2011, Konuspayeva, Jurjanz et al. 2011, Konuspayeva, Faye et al. 2011 a) including some controlled experiments focused on transfer of certain POPs into camel milk (Nurseitova, G. et al. 2014). None of these studies included the Mangystau Region but some covered the Atyrau Region, which carries the comparable burden of industrial and toxic legacy sites. In spite of that, we found higher levels of contamination of camel milk by PCBs and PAHs in particular. The comparison of summarized results for PCBs and PCDD/Fs presented by Konuspayeva, Faye et al. (2011) and (2011 a) with levels observed in this study is in Table 11. We found significantly higher levels of both indicator PCBs and DL PCBs in camel milk/shubat samples than the above-mentioned studies. This could reflect a specific situation in the Mangystau Region, particularly at certain sites. Prevalence of DL PCBs in total WHO-TEQ levels and lower levels of PCDD/Fs in comparison with previous studies seem to point to the environmental legacy of obsolete PCBs sources used in old transformers and capacitors as oils. There is an indication that such equipment exists in the Mangystau Region (Astaniina 2006, Republic of Kazakhstan 2009).

Table 11: Konuspayeva, Faye et al. (2011) and (2011 a)

Group of chemicals	PCDD/Fs (pg WHO-TEQ g ⁻¹ fat)			DL PCBs (pg WHO-TEQ g ⁻¹ fat)		
	Mean ± st. deviation	Median	Min – Max	Mean ± st. deviation	Median	Min – Max
Konuspayeva, Faye et al. (2011) and (2011 a)	0.80 ± 0.15	0.73	0.53–1.49	2.18 ± 1.27	1.66	0.77–5.53
This report	0.61 ± 0.57	0.38	0.01–1.33	12.64 ± 17.63	4.25	2.07–47.30
Group of chemicals	Total PCDD/F + DL PCBs (pg WHO-TEQ g ⁻¹ fat)			6 indicator PCB congeners (ng g ⁻¹ fat)		
	Mean ± st. deviation	Median	Min – Max	Mean ± st. Deviation	Median	Min – Max
Konuspayeva, Faye et al. (2011) and (2011 a)	2.98 ± 1.28	2.48	1.31–6.88	6.3 ± 2.7	5.1	0.6–17.4
This report	13.24 ± 17.61	5.02	2.08–47.61	15.81 ± 16.17	11.84	0.82–44.61

We looked at the total levels of six indicator PCBs, DL PCBs expressed in WHO-TEQ, PAHs and fat content in all six samples analysed in this study. The comparison expressed as a percentage of total values for all six samples is shown in the graph in Figure 11. None of the analysed groups of chemicals followed the curve for the fat content. It means that they depend on other components. Additionally, the curves for PAHs and both groups of PCBs differed. It means that, in the studied area, PAHs and DL PCBs have different sources of pollution. It also shows that oil or gas fires are not potential sources of DL PCBs contamination in the Mangystau Region, otherwise, they should be accompanied by similar levels of pollution by PAHs (see notice in Konuspayeva, Faye et al. 2011 a). The curves for both groups of PCBs are very similar. It seems that they have same source(s) of contamination.

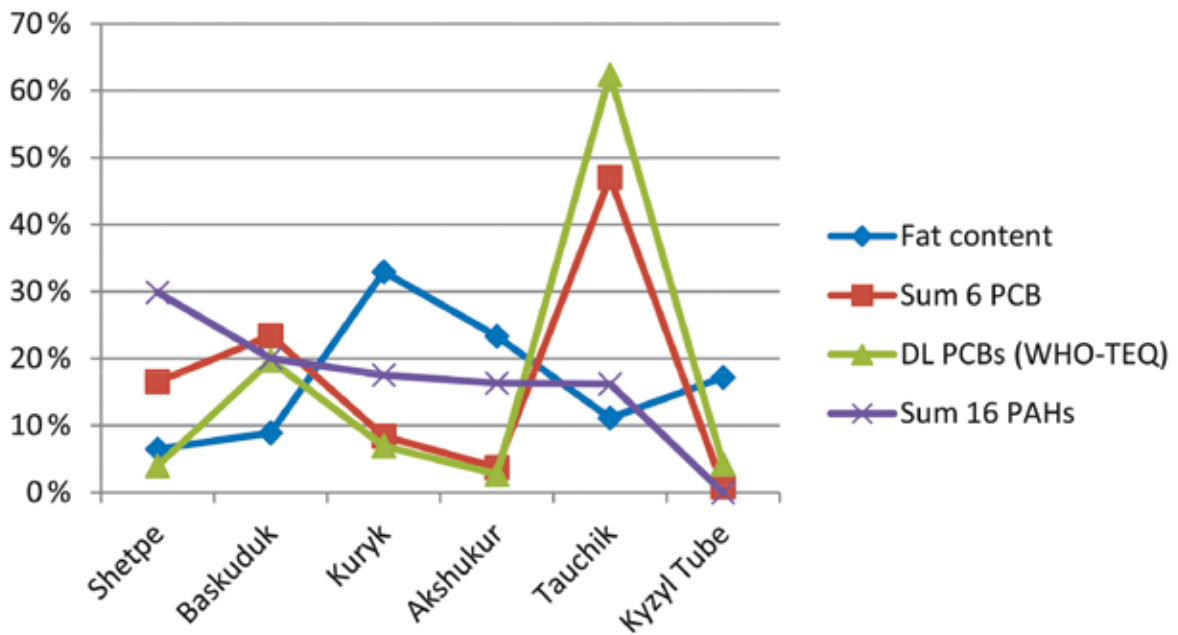


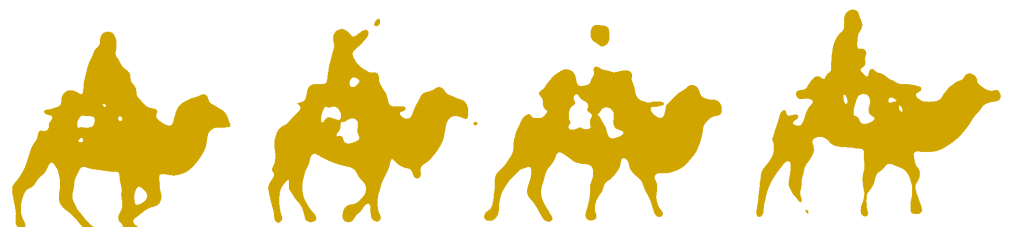
Figure 11: Percentage of fat content levels, sum of 6 PCB congeners, DL PCBs (WHO-TEQ), and sum of 16 PAH congeners. Proportion of the total sum for all 6 locations.

The highest level of PAHs was detected in the sample from Shetpe, while the highest concentration of both groups of PCBs was found in the sample from Tauchik. An increased level of PCBs was also observed in the sample from Baskuduk. Sample from Shetpe only had an increased content of indicator PCBs.

PCBs and zinc contamination of camel milk/shubat are probably the most serious threats to human health in relation to contaminants found in camel milk in the Mangystau Region. This is not the problem only of the Mangystau Region. For example, for the Kyzylorda Region, UNECE concluded that *“the population in the area has been exposed to organochlorine pesticides ... as a result of the use of defoliants, and to PCB and heavy metals as a result of industrial contamination. Toxic contaminants have accumulated in the water, the soil and the food chain. The main source of organochlorine compounds is probably the consumption of contaminated animal fat from beef, goat, chicken and milk products (butter). For infants, the main source after birth is breast milk. Hospitalized schoolchildren have remarkably higher levels of PCB, DDT, DDE and γ -HCH in their blood than Swedish children.”* (UNECE 2000).

In 2000, more than 15,000 seals died in the Caspian Sea, most of them in the Northern Caspian Sea near Kazakhstan (more than 10,000). A large level of contamination resulting from the continuous long-term impact of the polluted environment on the animals was identified as a cause of these mass deaths in combination with the virus of “canine distemper”. Highly concentrated PCB, DDT, chlordane, HCB and some heavy metals (such as zinc) found in the bodies of dead seals influenced their fertility and other physiological functions (Kajiwara, Niimi et al. 2002, Republic of Kazakhstan and Ministry of Environmental Protection 2003, Kuiken, Kennedy et al. 2006). The levels of PCBs and pesticides in the Caspian seals, however, were comparable to those in other aquatic mammals that have suffered from epizootics, and might pose a risk of immunosuppression (Kajiwara, Watanabe et al. 2008).

The potential sources of contamination by PCBs are listed in the basic document for UNDP/GEF project on addressing the issue of obsolete PCB oils in Kazakhstan, but none is located in the Mangystau Region. However, as the document states, the PCBs voluntary based inventory was not yet complete in 2009 (UNDP and Government of Kazakhstan 2010). Obligatory based PCBs inventory was started in 2012



and should be completed in 2014 but in 2016 it has not been completed yet, and continued. Based on the results of analyses of camel milk/shubat samples from the Mangystau Region, we suggest focusing particularly on potential obsolete PCB oils usage in mining and/or settlement areas in this region. This is the most urgent task in the areas surrounding the Tauchik, Baskuduk and Shetpe sites (see graph in Figure 9). In Tauchik, the source of pollution by PCBs could be the old equipment in an abandoned mine. Other potentially contaminated sites include oil sludge fields and/or mixed waste in Koshkar Ata near Baskuduk which can contain some level of PCBs. However, we did not find any research focusing on a potential content of PCBs in Koshkar Ata.

The high levels of zinc were found in sediments of the Caspian Sea in the mouth of Ural River (de Mora, Sheikholeslami et al. 2004). Researchers linked them to mining activities in the region. The same may apply to the Mangystau Region; increased levels of zinc in camel milk found in this study may relate to its content in surface waters drunk by camels, particularly in Kyzyl Tube and Baskuduk localities. Dust can also be a major source of zinc contamination as the wind is a more important pathway in natural conditions of Kazakhstan. However, more research is needed in order to confirm this explanation.

In section 2.1.3., we have noted that in Kuryk, when the feed for animals is scarce, it is supplemented with cartons. No study specifically focused on assessing the potential toxic contamination of the milk of ruminants which were fed with carton. However, there is an Irish study on using shredded newspapers to feed cows. It concluded that *“the heavy metal/trace element levels in both milk and blood samples were within normal ranges. There was no detectable level of any of the 16 PCB congeners or of naphthalene in any of the samples analysed. This indicates that newspaper in its present form is safe to use as a bedding material for dairy cows”* (O'Connell and Meaney 1997). It seems that feeding animals with carton is not a likely source of increased levels of PCBs or PAHs in a pooled camel milk/shubat sample from Kuryk.

4.7 Discussion on the potential solutions of contamination by PCBs

Results of our study support conclusions of some other studies and reports, that the contamination of the environment in Kazakhstan by PCBs is a serious issue (UNDP and Government of Kazakhstan 2010, Novikov, Simonett et al. 2013). In the Mangystau Region too, it represents a serious threat to human health and the environment.

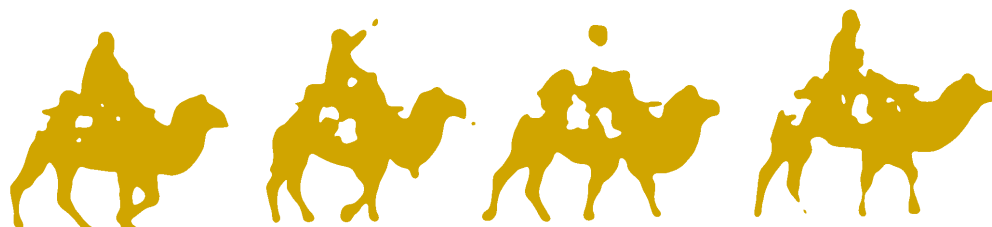
We are aware that there is a project which aims at *“implementing a comprehensive management plan, with the overall objective of significantly cutting emissions of PCBs and reducing their impact on health and the environment through developing sound management across the country. The project seeks to ensure that there is a modern, fully enforceable PCB regulatory system in place. It includes strengthening administrative functions, capacity-building for sound management, dismantling 850 tonnes of PCB transformers and disposing of them, and regionally organized secure storage and disposal of PCB capacitors”* (Novikov, Simonett et al. 2013). However, to our knowledge, there is no complete inventory of PCBs and PCBs contaminated sites in Kazakhstan or in the Mangystau Region. This would be the first necessary step to address the issue of contamination by PCBs in this part of the country. The basic document of the above-mentioned project also states that *“while the initial inventory (of PCBs) is far from complete it reveals significant stockpiles of PCBs in Kazakhstan”* (UNDP and Government of Kazakhstan 2010).

To our knowledge, the construction of a new hazardous waste incinerator in Pavlodar was suggested as part of the project, but the plan was opposed by the public. Waste incineration, plasma technologies, cement kilns and metallurgical technologies are listed among major sources releasing unintentionally produced POPs in Annex C to Stockholm Convention (Stockholm Convention 2010). The use of these technologies for destruction of PCBs and/or combustion of soils contaminated by PCBs may lead to the formation of PCDD/Fs or chlorobenzenes and increase currently low contamination of food sources in the Mangystau Region by this group of POPs.

An expert group of the Stockholm Convention evaluated in 2006 U-POPs releases of several non-combustion technologies and found them very low (UNEP – EG BAT/BEP 2006). Releases data of PCDD/Fs and dioxin-like PCBs (DL PCBs) from physico-chemical treatment of POPs waste technologies is summarized in Table 12. Some of these technologies were also successfully used to destroy the PCB oils and/or for decontamination of soils contaminated by PCBs.

Table 12: Releases of U-POPs by various alternative technologies of waste incineration as potential alternatives for PCBs containing wastes destruction.

Technology	Specific facility	Operation period	Air releases (ng TEQ m ⁻³)	Water discharge (ng TEQ l ⁻¹)	Source
PCDD/Fs and dioxin-like PCBs					
Alkali metal reduction	Japan Environmental Safety Corporation, Kitakyushu facility	Dec 2004– March 2015	0.0000013– 0.0000530		(UNEP – EG BAT/BEP 2006)
	Japan Environmental Safety Corporation, Toyota facility	Sep 2005– March 2015	0.0000842– 0.0024947	NA	
Catalytic-hydro dechlorination	Japan Environmental Safety Corporation, Osaka facility	2006– March 2015	0.000079– 0.00010	NA	(UNEP – EG BAT/BEP 2006)
Photochemical dechlorination and catalytic dechlorination reaction	Japan		0.00007	NA	(UNEP – EG BAT/BEP 2006)
Supercritical Water Oxidation	Japan		0.001–0.002	0.0000005	(UNEP – EG BAT/BEP 2006)
Subcritical Water Oxidation	Japan		0.00009	NA	(UNEP – EG BAT/BEP 2006)
PCDD/Fs only					
Alkali metal reduction	Japan Environmental Safety Corporation, Kitakyushu facility	Dec 2004– March 2015	0		
	Japan Environmental Safety Corporation, Toyota facility	Sep 2005– March 2015	0–0.0015834	0.0000003500 – 0.0000077500	
Catalytic-hydro dechlorination	Japan Environmental Safety Corporation, Osaka facility	2006– March 2015	0.00000066– 0.00000076	NA	
BCD (Australia)	BCD Technologies, Queensland		0.0119–0.05	NA	(UNEP – EG BAT/BEP 2006)



Technology	Specific facility	Operation period	Air releases (ng TEQ m ⁻³)	Water discharge (ng TEQ l ⁻¹)	Source
BCD (Japan)	Japan		<0.01	NA	(UNEP – EG BAT/BEP 2006)
BCD (Spolana Neratovice – pilot)	BCD CZ, s.r.o., Prague		0.013–0.031	NA	(UNEP – EG BAT/BEP 2006)
BCD (Spolana Neratovice – full)	BCD CZ, s.r.o., Prague		0.0017–0.0424	0–1.4	(Veverka, Čtvrtníčková et al. 2004)
Gas Phase Chemical Reduction	Australia		0.0000028–0.00027 (<0.016)	0.00000061–0.00084	(Vijgen and McDowall 2008)
DL PCBs only					
Alkali metal reduction	Japan Environmental Safety Corporation, Kitakyushu facility	Dec 2004–March 2015	0.0000013–0.0000530		
	Japan Environmental Safety Corporation, Toyota facility	Sep 2005–March 2015	0.0000842–0.0024947	0.0000372590–0.0001289250	
Catalytic-hydro dechlorination	Japan Environmental Safety Corporation, Osaka facility	2006–March 2015	0.000078–0.00010	0.0000372590–0.0001289250	
PCBs only					
Alkali metal reduction	Japan Environmental Safety Corporation, Kitakyushu facility	Dec 2004–March 2015	<0.000010 – 0.000600 mg m ⁻³	ND (0.003 mg l ⁻¹)	
	Japan Environmental Safety Corporation, Toyota facility	Sep 2005–March 2015	<0.001 mg m ⁻³	<0.0005 mg l ⁻¹	
Catalytic-hydro dechlorination	Japan Environmental Safety Corporation, Osaka facility	2006–March 2015	0.00067–0.0024 mg m ⁻³	NA	

Technologies like Alkali Metal Reduction (called also the Sodium Reduction), Base Catalysed Decomposition (BCD), Catalytic Hydrodechlorination (CHD) or Gas Phase Chemical Reduction (GPCR) were successfully used for destruction of PCBs wastes as well as some other kinds of waste. These technologies are described, for example, in a document prepared by Arnika – Toxics and Waste Programme (2016).

As the problem of PCBs contamination is widespread in different parts of the country, a non-combustion technology which can be easily moved from one site to another and/or can be built as smaller units according to the amount of material which needs to be processed is also more suitable than the waste incineration and/or co-incineration as a method for destruction of PCBs in both liquid and solid phase (soils contaminated by PCBs). Of course, there are more criteria for choosing the right technology for destruction of POPs (or PCBs) wastes that can play a significant role. Some basic steps were proposed by IPEN (IPEN Dioxin PCBs and Waste Working Group 2010) and Costner, Luscombe et al. (1998).

Furthermore, the inventory of sites contaminated by POPs in Kazakhstan needs to be completed before the final decision on their remediation is made. The basic information about the number and size of contaminated sites is needed before choosing the proper way for their remediation and the treatment technology for the clean-up of soils contaminated by PCBs, and potentially by other POPs and toxic chemicals. A basic guidance on how to prepare such inventory and how to manage contaminated sites was summarized by Bell (2015) as a part of broader reports prepared by Arnika, EcoMuseum and CINEST (Arnika, AWHHE et al. 2015, Arnika, CINEST et al. 2015).

5. Conclusions and recommendations

Levels of PCDD/Fs and heavy metals, with exception of zinc level in the sample from Kyzyl Tube, we have found were lower than the levels reported by the previous broader studies led by Ghaukar Konuspayeva in cooperation with other scientists (Konuspayeva, Faye et al. 2009, Konuspayeva, Faye et al. 2011, Konuspayeva, Jurjanz et al. 2011, Konuspayeva, Faye et al. 2011 a). Levels of both DL and NDL PCBs were much higher in our samples than the results published for samples collected in the wider area of Kazakhstan (Konuspayeva, Faye et al. 2011, Konuspayeva, Faye et al. 2011 a), the same applying for PAHs (Konuspayeva, Jurjanz et al. 2011). From the point of view of associated health risks assessed through the dietary intake of discussed groups of chemicals, most significantly they come from exposure to DL PCBs followed by the indicator PCBs (see section 4.2.5), while the health risk from exposure to PAHs contained in camel milk is low as most dangerous PAH congeners do not accumulate in the milk of ruminants due to their specific metabolism. Attention should also be paid to the zinc levels in camel milk from Kyzyl Tube and Baskuduk. Exposure to other heavy metals, OCPs, and PCDD/Fs from the consumption of camel milk in the Mangystau Region found by this study was low due to the relatively low levels of these chemicals in camel milk/shubat samples collected at the six selected localities. However, limitations of this study should be taken into account in the overall evaluation of the situation in Mangystau Region. Worrying levels of PCBs also raise questions regarding the full inventory of PCBs sources in Kazakhstan followed by their destruction and remediation of contaminated sites, given that the most likely sources of contamination of camel milk are the obsolete stockpiles of PCBs and sites contaminated by PCBs, e.g. capacitors with PCB oils.¹⁷

5.1 Unintentionally produced POPs (PCDD/Fs, DL PCBs)

Toxicity of DL PCBs has prevailed over PCDD/Fs congeners' toxicity in all camel milk samples collected for this study. Three out of six samples in this study¹⁸ exceeded the EU limit value for the total TEQ of PCDD/Fs and DL PCBs in milk (European Commission 2011), while none of them exceeded either the Kazakhstani (3 pg WHO-TEQ g⁻¹ fat) or the EU limit value (2.5 pg WHO-TEQ g⁻¹ fat) set just for PCDD/Fs content in milk.

Daily consumption of camel milk/shubat from Tauchik can lead to the exceeding of the TDI set by WHO experts for PCDD/Fs and DL PCBs within the range of 1–4 pg WHO-TEQ kg⁻¹ of body weight per day (van Leeuwen, Feeley et al. 2000) (see Table 7 and section 4.2.5). Furthermore, the consumption of

¹⁷ These conclusions are discussed more specifically in following subsections. More details on the PCBs destruction and remediation of contaminated soils are provided in subsection 5.6.

¹⁸ These samples were from Tauchik, Baskuduk, and Kuryk.



camel milk/shubat from Baskuduk and Kuryk can significantly contribute to the total intake of PCDD/Fs and DL PCBs up to the level exceeding the lower limit of TDI.

5.2 PCBs

Sums of 6 indicator PCB congeners were in the range of 3.54–44.61 ng g⁻¹ fat, and in general significantly higher than those found in previous studies (Konuspayeva, Faye et al. 2011, Konuspayeva, Faye et al. 2011 a). The median of 11.84 ng g⁻¹ fat obtained for the six samples in this study is almost 2.5 times higher than 5.1 ng g⁻¹ fat reported by Konuspayeva, Faye et al. (2011 a). Consumption of shubat/camel milk from three¹⁹ out of six sampled locations presented in this study can result in exceeding the total daily intake of PCBs by an average American in 1990; the mean value for the levels of PCBs in milk from these locations can reach half of the total dietary intake of PCBs in Netherlands (see section 4.2.5). High levels of indicator PCBs support our assumption about the potential source(s) of contamination being the obsolete PCB oils used in equipment in the industrial or mining areas and/or the presence of sites heavily contaminated by PCB oils in the Mangystau Region.

Conclusions for DL PCBs are presented in section 5.1.

5.3 PAHs

Levels of PAHs in camel milk samples from the Mangystau Region are higher than those observed in a previous study from Kazakhstan by Konuspayeva, Jurjanz et al. (2011). Of the 16 PAH congeners, the highest levels were observed for naphthalene, followed by phenanthrene.

The highest level of PAHs per gram of fat was observed in camel milk sample from Shetpe. We assume that the most likely source of this high contamination is the use of oil as a fuel in the near cement kiln. It is difficult to verify this assumption by an analysis of PAH congeners' pattern because the milk of ruminants demonstrates specific metabolism of some POPs, including PAHs, making such analysis impossible (see sections 4.3 and 4.2.3). Sources of PAHs should be addressed in order to decrease contamination of food by PAHs; the contribution of camel milk consumption to the overall dietary intake of PAHs, however, is not high, and definitely does not represent as a serious issue as the contamination by PCBs.

5.4 OCPs

Neither Kazakhstani nor the EU limits were exceeded in any of the camel milk samples in this study. Levels of OCPs were lower in our camel milk samples than those found in the study focusing on the contamination of camel milk in other regions of Kazakhstan (Konuspayeva, Jurjanz et al. 2011).

5.5 Heavy metals

Levels of heavy metals in milk samples collected for this study were in general lower than those observed in other regions of Kazakhstan in previous studies (Konuspayeva, Faye et al. 2009, Konuspayeva, Jurjanz et al. 2011), with the exception of zinc content in the sample from Kyzyl Tube.

Two samples, from Baskuduk and Kyzyl Tube, exceeded the maximum acceptable level according to the Russian legislation for the zinc content in milk (see Table 2). The zinc content in the sample from Kyzyl Tube was almost 3 times the limit value set for milk. The mining activity nearby Kyzyl Tube is the most likely source of contamination (see section 2.1.6).

5.6 Recommendations for addressing the contamination by PCBs

Certain basic steps need to be taken to address the problem of the overall contamination by PCBs, as it was observed in this study in samples of camel milk from the Mangystau Region:

1. Completing the inventory of PCBs
2. Inventory of waste contaminated with PCBs

¹⁹ These locations are Tauchik, Kuryk, and Baskuduk.

3. Completing the inventory of sites contaminated by PCBs (Arnika, CINEST et al. 2015, Bell 2015)
4. Preparing the plan for the remediation of contaminated sites and the destruction of PCBs and/or more complex destruction of POPs waste (Bell 2015)
5. Setting criteria for choosing the best technology to destroy PCBs (POPs); (Costner, Luscombe et al. 1998, IPEN Dioxin PCBs and Waste Working Group 2010, Arnika, CINEST et al. 2015)

Based on the experience with available technologies and the requirements of the Stockholm Convention to prevent the formation of U-POPs when destroying POPs wastes, we suggest preferably using non-combustion technologies for disposal of remaining PCB oils and wastes contaminated with PCBs (UNEP – EG BAT/BEP 2006, IPEN Dioxin PCBs and Waste Working Group 2010).²⁰

6. Limitations of the study

The major limitations of this study include the limited financial and personal resources, and the limited time available. Therefore, only a limited number of camel milk and/or shubat samples could be obtained. Furthermore, it was not possible within our resource limits to follow the same sampling plan as described in previous studies on the contamination of camel milk in Kazakhstan conducted by Ghaukar Konuspayeva in cooperation with other scientists (Konuspayeva, Faye et al. 2009, Konuspayeva, Faye et al. 2011, Konuspayeva, Jurjanz et al. 2011, Konuspayeva, Faye et al. 2011 a). This allows only a limited comparison of the results with those presented in the previous studies. Different circumstances of the studies should be taken into account.

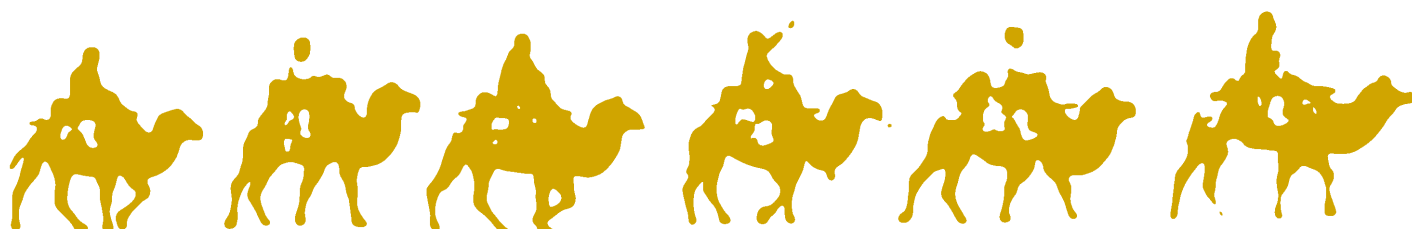
We also lack the data on the level of the total dietary intake of different contaminants in Kazakhstan, as well as the information on a typical diet of the Kazakhstani population. We worked instead with the limited information available to us. Nevertheless, a general idea of the situation including the identification of major issues in relation to the contamination of camel milk as a significant food source in the Mangystau Region was obtained. However, further research in this field is still necessary. The presented results cannot be considered exhaustive, rather expressing the need for an extended study in the future.

The evaluation of pollutant concentration levels found in our samples with regards to the existing regulations has also its limitations. Various regulations are based on different definitions serving different purposes. In addition, there is currently no legislation for some of the pollutants and some existing legal limits or TDI levels might be outdated. Also, to our surprise, there appears to be no sufficient data on the PAHs concentrations in the environment in Kazakhstan.

The estimation of potential risks to humans and the environment cannot be based solely on the valid legislation; an extensive risk analysis based on a sufficient number of samples and detailed description of the state of the area and the potential risk receivers is crucial. We tried to provide a basic evaluation of health risks expressed as the daily intake of some crucial pollutants to give at least some basic idea about the level of human exposure to different pollutants.

The study limitations and the obvious need for further investigation notwithstanding, we believe that the most important task is to start addressing the overall pollution by such contaminants as PCBs or zinc in the Mangystau Region.

²⁰ Some of them are described in our report: Arnika – Toxics and Waste Programme (2016). Brief Review of Potential Technologies for Addressing the Issue of Obsolete Polychlorinated Biphenyls (PCBs) and PCB Wastes in Kazakhstan. Prague – Karaganda: 12. Releases of U-POPs from these technologies are summarized in Table 12.



7. Photographic supplement



Above: Kumys and shubat – bottled horse and camel milk is sold in supermarkets. However, many people buy fresh milk from small farms, where it is not checked for potential contamination.
Below: Camels feeding on waste from open waste containers in Kuryk.



*Above: Old Aktau city waste landfill is not being reclaimed and neither is it fenced off.
Below: Koshkar Ata, uranium processing tailing pond at Aktau is freely accessible to domestic animals.
They come here because of lack of water in surrounding semi-desert area.*



*Above: Abandoned uranium processing plant in Aktau still represents a threat to the environment.
Below: Malaya Oymasha, a heavily polluted lake surrounded by family houses in Aktau.*



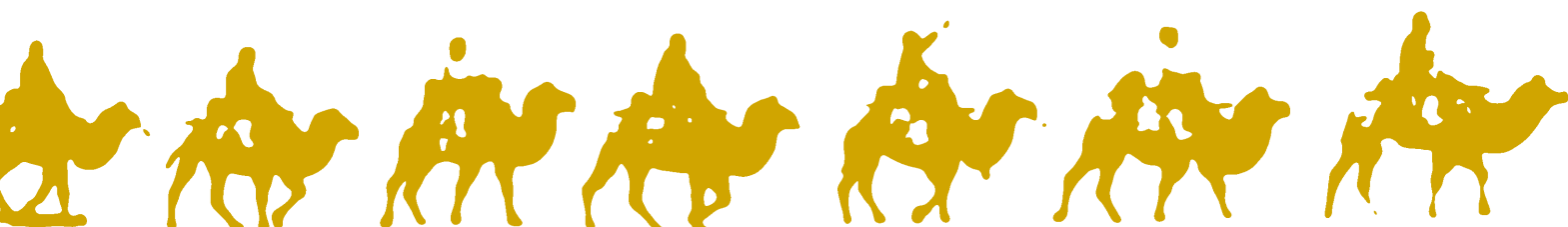
Above: Family camel farm in the steppe.

Below: Collection of camel milk samples.

8. References

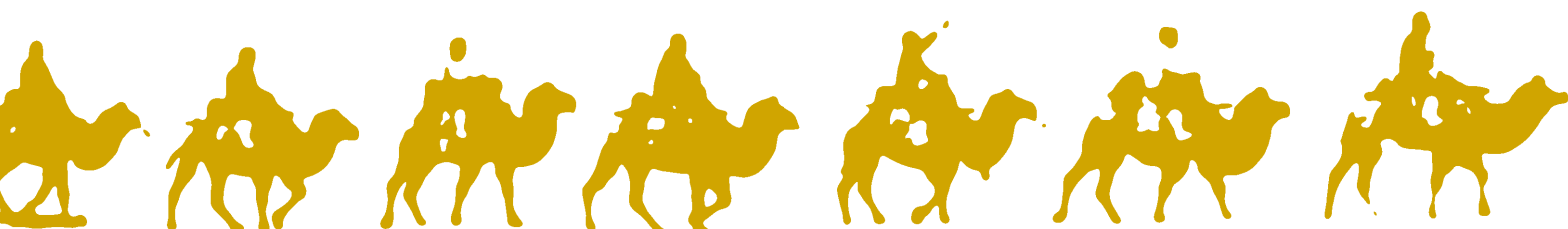
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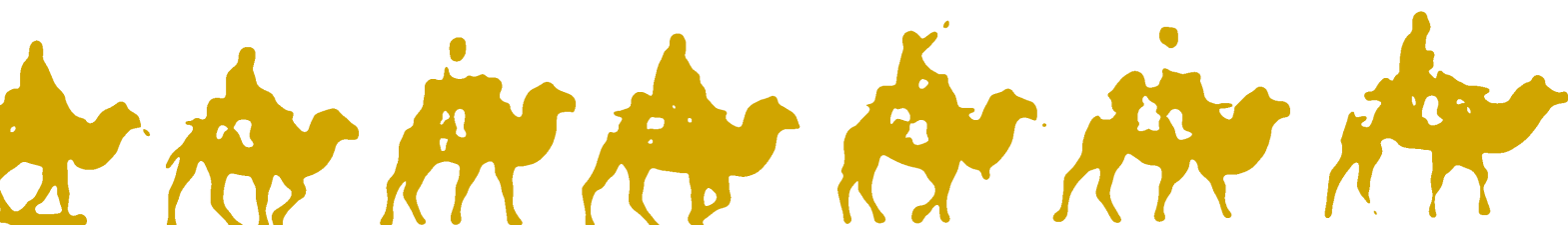
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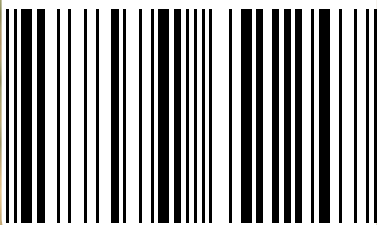
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