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Trend Test and Change-Point Detection for Concentrations of I-131 in Drinking Water and Dairy Milk in the U.S. Midwest

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Abstract

Radioactive iodine (I-131) is one of the radionuclide byproducts of uranium and plutonium fission or neutron capture by Tellarium-130. I-131 decays to Xenon and releases β-particles and γ-radiation. Due to its release of IR during nuclear decay, I-131 is a potent radioactive hazard to public health. In this study, I-131 data from the EPA database, measured from drinking water and pasteurized dairy milk in the U.S. Midwest, were analyzed using non-parametric trend test and change-point detection techniques. We perform the Gaussian Kernel (G-K) smoothing to separate long-trends from the random noise in the data. The Mann-Kendall (M-K) rank correlation and Theil-Sen's (T-S) tests indicated the existence of positive trends on the data. Furthermore, the Pettitt's Homogeneity (P-H) tests revealed statistically significant increases (α=0.05, $p<0.0001$) in I-131 radiation levels between the periods 1978-1983 and 1984-1990 in pasteurized dairy milk. With regards to drinking water, we noted significant increases between 1978-1995 and 1995-2017. We recommend further studies to determine the possible factors contributing to the periodicities in the data.

Keywords: Dairy milk • Drinking water • Ionizing radiation • Radioactive iodine (I-131) • Non-parametric test • Trend test

Introduction

Understanding the trends of I-131 nuclide contamination levels in the environment, food, and drinking water remains a national priority. Monitoring concentrations of Ionizing Radiation (IR)-producing nuclides in various media started during the nuclear testing era due to nuclear fallout, leakage, and/or accidents. The association between I-131 radiation exposure and human thyroid carcinoma has been reported in several studies, with the risk at its peak during childhood, and decreasing with increasing age, and lowest in adults. Based on the United Nations report (2000), the high-level risk during childhood and early adolescence stage mostly is due to the absorbance and accumulation of I-131 in the thyroid glands during growth. IR exposure can result in malignant and/or papillary thyroid carcinoma, with the later mostly occurring after at least 5 years [1]. I-131 is commonly absorbed by the high iodide-affinity tissues in the thyroid.

The accumulation of I-131 radionuclides in thyroid glands has the potential to damage the thyroid tissue. Exposure to I-131 can occur through different pathways such as airborne, direct radiation, waterborne, and ingestion. For instance, human exposure to I-131 can occur through internal (ingestion, inhalation, absorption through wounds, etc.) and external (direct skin) routes drinking water and pasteurized milk are potentially two major sources of I-131. Due to its short half-life of 8.0545 days ± 0.0063 days, I-131 significantly decays within few weeks after production. However, the time between production and distribution is very short, which makes pasteurized dairy milk one of the sources of high levels of I-131. Several US Midwestern states, particularly those in EPA Region 7 are currently monitoring the levels of I-131 in the environment, rain, food, and drinking water. In EPA Region 7, Missouri, Kansas, and Iowa have old nuclear dump sites that are currently being cleaned-up to prevent human exposure to IR. The need levels and trends of I-131 levels in drinking water and milk remains a high priority in this region [2].

Background

While the high risks of thyroid carcinomas due to exposure to I-131 are documents, the contributions of improved screening procedures to the increasing thyroid cancer incidences is another topic of discussion. This study does not examine that debate, but it reviews the sources, potential risks, and pathways of I-131, which are crucial to the trend analysis of measured concentration levels [3].

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Sources of I-131 in the environment

I-131 is commonly produced by nuclear fission and neutron capture by Tellarium-130. In the former process, Uranium and plutonium nuclear fission are the major sources. I-131 radionuclides decay to two forms of Xenon, a process that produces about 182 keV of β-particle energy and γ- radiation ranging from 80.18 keV to 637.0 keV. Nuclear fallout, leakages from nuclear reactors, and dump sites are the major anthropogenic sources of I-131. According to the ACA (2019), between 1945 and 2017, about 2,056 nuclear test explosions were conducted worldwide, of which 528 were atmospheric. While nuclear plants are highly controlled, in the past, human errors, natural disasters, and technical faults have triggered nuclear disasters. The Three Mile Island accident in 1979, the Chernobyl accident in 1986, and the Fukushima Daiichi accident in 2011 are the three major accidents at civilian nuclear plants recorded to date. For instance, the nuclear radiation from the Fukushima nuclear power plant accident were still detectable five years after the accident. I-131is one of the poisonous isotopes of major concern to the extend it is used as a reference for classifying the impact of nuclear accidents/explosions. The Chenobyl accident alone released about 1,800,000 Tbq of I-131 into the atmosphere, while the Fukushima released an estimated 150,000 TBq of I-131 into the atmosphere. The release airborne I-131 nuclide from the atmosphere through rainy/snowy weather is one of major sources of radioiodine contamination in the environment. Based on the ATSDR (2008), the major sources of I-131in the environment in the U.S. have been leakages from nuclear power plants, dumpsites and/or nuclear weapons testing. The I-131 particles eventually find their way into the earth's biosphere [4].

Toxicological implications of I-131 exposure

Increased incidence of thyroid cancer has been attributed to improved diagnostic procedures and exposure to radiation and other carcinogens. While the actual mechanisms which determine the high sensitivity of thyroid glands to IR are not well known, the probable biological mechanisms of carcinogenic effect of IR on the thyroid glands have been comprehensively discussed. Nevertheless, concerns about the carcinogenic risks of exposure to I-131 have grown following the discovery of increased thyroid carcinoma incidences among nuclear explosion survivors [5].

The ATSDR (2008) reported a linear relationship between exposure of the thyroid gland and moderate to high doses of I-131 to increasing risk of thyroid cancer. In addition, Shibata et al. (2001) reported a strong relationship between exposure to I-131 and increased incidences of thyroid cancer among children in Belarus, Russia, and Ukraine following Chernobyl accident. The pathophysiology of thyroid carcinoma has shown that high incidences of papillary thyroid carcinomas were commonly observed following exposure to IR. Similarly, an increase in thyroid cancers among people clustering around nuclear power plants have been reported. The sensitivity of thyroid glands to radiation may be due to its ability to concentrate iodine, including I-131. In the U.S., the concentrations of some nuclides have been higher in some Midwestern than Western states although the levels are low based on the EPA standards. Thyroid cancer is the common form of cancer in the U.S. and is the sixth most common cancer among women aged 20 to 34. The CDC report on cancer mortality by state indicated significantly high rates in the U.S. Midwest [6].

Pasteurized dairy milk contaminated with I-131 poses greater risks of IR exposure to children than adults. Children frequently drink milk than most adults, and that their small thyroids are likely to sustain greater IR damage than adults'. Thyroid hormones, which are needed for growth in babies and children require iodine. The impact of IR on children is not immediately recognizable but can manifest as cancer in several years after exposure. For instance, an increase in thyroid cancer with age, peaking at 20 years-35 years followed by a decline thereafter. Thus, the adverse toxicological effects of IR to the human health are well documented, but the magnitude of the impacts of long-term exposure to very low levels of I-131 remains a topic of intense study.

This study has two major goals: 1) to determine the variations and trends of I-131 using time series analyses; and 2) determine the changes in the trends and their statistical significance. With increasing incidences of thyroid cancer diagnosis and mortality in the US, which are currently estimated to be about 3.6% and 1.1%annually, respectively, there is increasing concern about the causes of surging thyroid cancer cases. It is unclear if the rising cases are either primarily due to improved diagnostic and detection techniques or an indication that the disease is on the rise. Given that thyroid cancers have been linked to I-131 exposure, there is need to determine the trends in the pollution levels of I-131 in drinking water and pasteurized milk [7].

Materials and Methods

While there are 36 radioactive iodine isotopes, I-131 is one of the elements known to have the greatest environmental and human effects. Due to its high potential for human health risks, I-131 is considered in this study. I-131 is commonly found in particle form, which can be ingested along with food, milk or water. The analyses in this study involves only I-131 monthly concentration levels in pasteurized dairy milk and annually measured concentrations in drinking water measured in four states in the US Midwest. The pasteurized milk I-131 concentration data were for the following cities: Des Moines (Iowa), Omaha (Nebraska), Saint Louis (Missouri), and Wichita (Kansas). The radioiodine data in drinking water were for Cedar Rapids (Iowa), Lincoln (Nebraska), Jefferson (Missouri) and Topeka (Kansas). These data, sampled between 1978 and 2017 were taken from an open source EPA database. These cities had the most comprehensive data in EPA Region [8].

To reduce the noise in the data, a 4-point Gaussian-kernel (G-k) smoothing was used to remove the fluctuations in the I-131 data for both pasteurized milk and drinking water. For this study, non-parametric statistical tests were used to analyze the trends in the time series. The Mann-Kendall (M-K) rank correction test and the Theil–Sen slope (T-S) were applied to the G-K filtered data to test for the existence and types of trends in the time series, respectively. The M-K test is robust against nonnormality and is widely used to test monotonicity in time series. Thus, its application was suited for the longitudinal data used in this study. Based on the T-S technique, a non-parametric median-based linear model that estimates the strength of trend determined by the M-K test (Table 1) [9].

	City	N	Mean	Study	M-K Test			T-S slope	P-H test	
					p-value	Tau	S		t	p-value
Dairy milk	DM	176	2.113	2.428	<0.0001	0.31	338	0.267	114	<0.0001
	SL	165	1.344	2.832	0.0277	0.135	126	0.163	89	<0.0001
	OM	165	1.867	3.233	<0.0001	0.256	240	0.219	90	<0.0001
	WI	166	2.508	3.128	0.0003	0.22	206	0.28	97	<0.0001
Drinking water	CR	44	0.017	0.028	0.8802	0.056	2	0.001	32	<0.0001
	LI	43	0.024	0.04	<0.0001	0.888	32	0.045	21	<0.0001
	JF	38	0.088	0.07	0.0500*	0.389	14	0.039	10	<0.0001
	TO	38	0.045	0.04	0.0237	0.444	16	0.016	20	<0.0001

Table 1. Descriptive statistics for the M-K rank correlation and P-H tests based on G-K smoothed. In both tests, the significance level α=0.05 (two-tailed) was used. The cities DM, SL, OM, WI, CR, LI, JF,and TO stand for Des Moines, Saint Louis, Omaha, Wichita, Cedar Rapids, Lincoln, Jefferson, and Topeka, respectively. The null hypothesis (Ho) is rejected for all dairy milk time series.

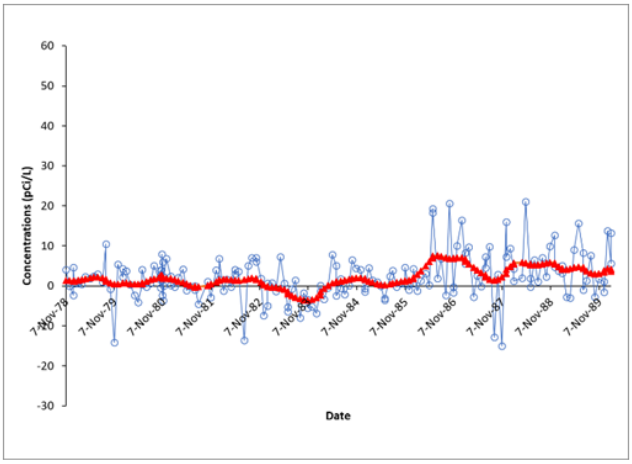
The Pettitt's Homogeneity (P-H) nonparametric test was used to detect the time of shifts in time series at 95% Confidence Level (CI). The P-H technique does not require the knowledge of the distribution of the data. The null hypothesis (Ho) assumed the data to be homogeneous, and the alternative hypothesis (Ha) was that the data were non-homogeneous. Homogeneity is an indication that all the data in the series are not significantly different. Data analyses for the changes in the trends based on the P-H test for pasteurized dairy milk and drinking water are presented [10].

Results

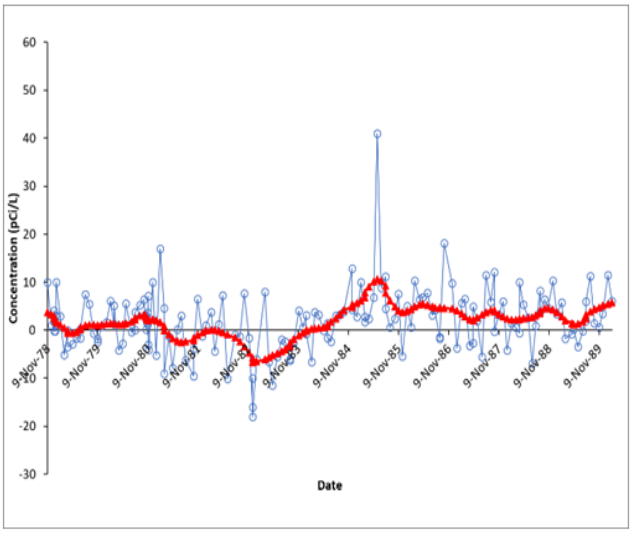
The descriptive statistics and non-parametric test for the existence of trends and homogeneity of I-131 concentrations are presented in the sections below.

Descriptive statistics for I-131 concentrations

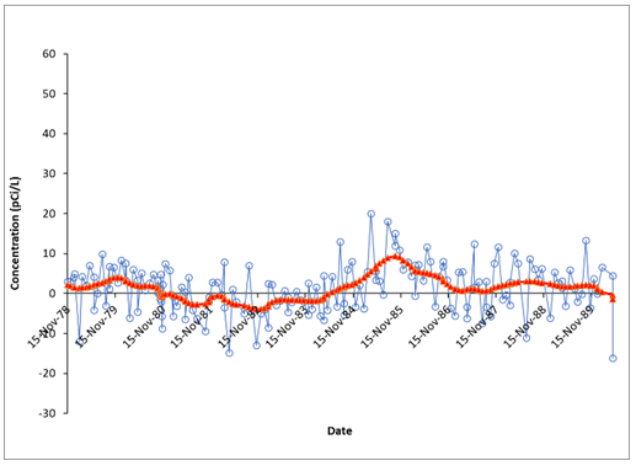
In pasteurized dairy milk, the concentration levels (mean ± stdev) of I-131 data (before G-K smoothing) were 2.0224 ± 6.1970, 1.3307± 5.9868, 1.8970 ± 6.4588, and 2.5676 ± 6.7820 pCi/L for Des Moines, Saint Louis, Omaha and Wichita, respectively. The respective number of samples in each dataset. Analysis of the G-K-smoothed time series of I-131 concentration data indicated decreasing trends for the period 1978-1983. The highest peaks occurred between 1984 and 1988. However, the I-131 contamination data for milk were not available for the period 1991-2017 (Figure 1) [11].



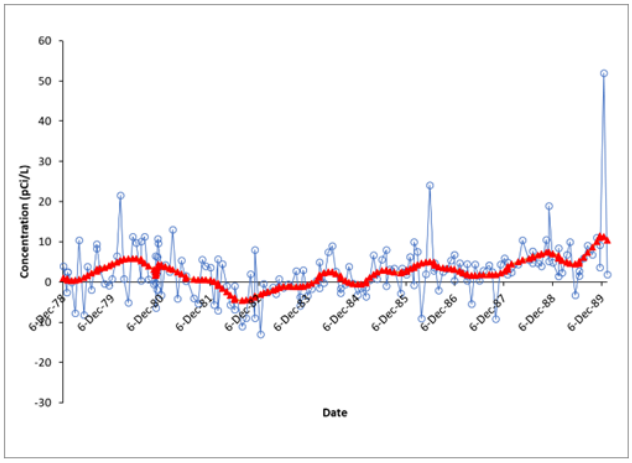
a. DES MOINES, IA. (**Note:** DM-Normal DM Gaussian-Smoothed)



b. OMAHA, NE. (**Note:** OM–Normal OM Gaussian-Smoothed)



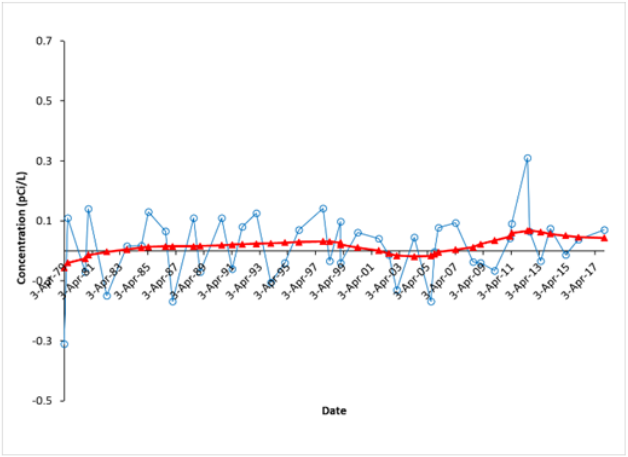
c. SAINT LOUIS, MO. (Note: SL–Normal SL Gaussian-Smoothed)



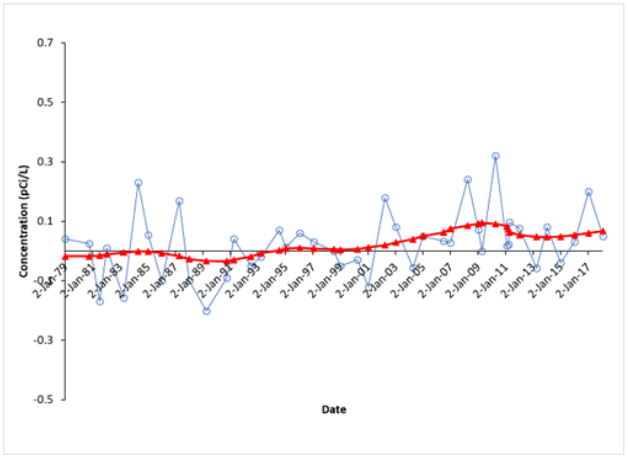
d. WICHITA, KS (Note: WI–Normal WI Gaussian-Smoothed)

Figure 1. The normal and G-K smoothed times series of monthly pasteurized dairy milk data have almost similar fluctuation characteristics. The time series for all the four cities were closely similar with peaks and lows occurring during the same time periods. The lowest contamination levels occurred during 1982-1983.

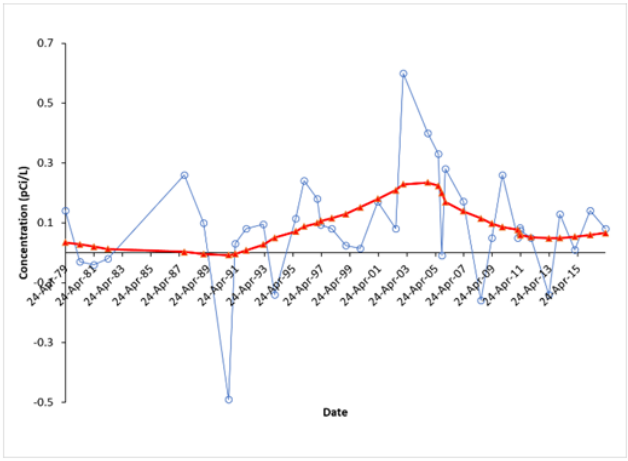
The contamination levels of I-131 in drinking water (mean ± stdev) were 0.015 ± 0.108 (N=44), 0.089 ± 0.176 (N=38), 0.025 ± 0.109 (N=44), and 0.040 ± 0.131 (N=44) pCi/L for Cedar Rapids (IA), Jefferson (MO), Topeka (KS), and Lincoln (NE), respectively. The highest concentration levels were recorded during the period 2007-2017 in Cedar rapids (IA) and in Lincoln (NE). In Saint Louis (MO), the peak occurred during the period 1999-2013. Overall, the I-131 concentrations in drinking water were consistently lower than the levels in milk (Figure 2) [12].



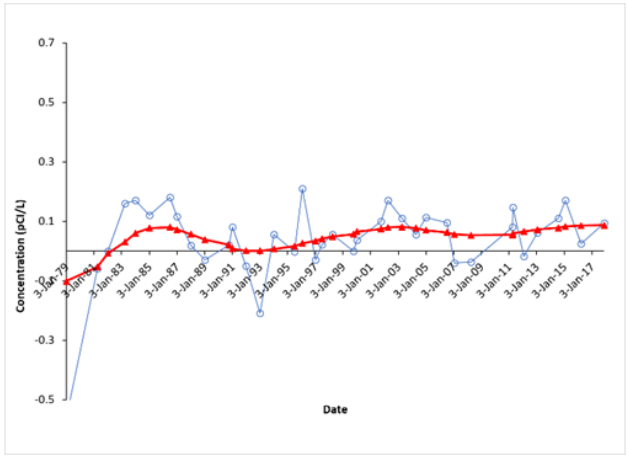
a. CEDAR RAPIDS, IA (Note: CR Normal CR_Gaussian Smoothed)



b. LINCOLN, NE (Note: LI Normal LI_Gaussian Smoothed)



c. JEFFERSON, MO (Note: JF Normal JF_Gaussian Smoothed)



d. TOPEKA, KS (Note: TO Normal TO_Gaussian Smoothed)

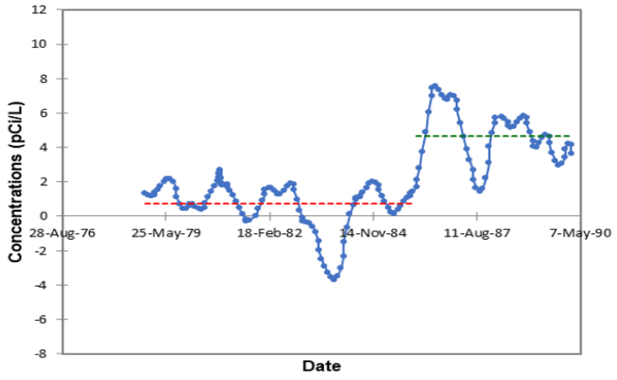
Figure 2. The normal and G-K smoothed times series of drinking water data have almost similar fluctuation characteristics.

Trend and homogeneity tests

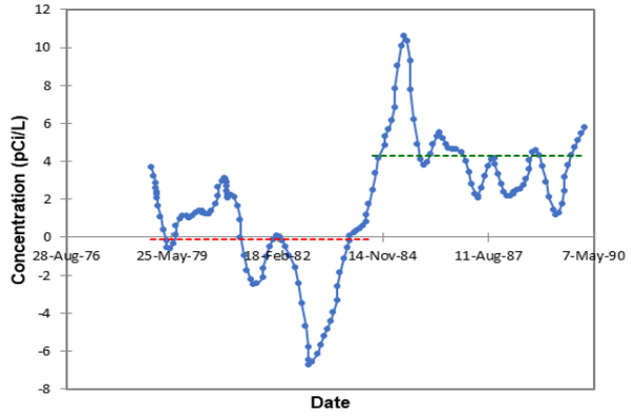
Using the pasteurized dairy milk, the assumption of homogeneity was rejected in the P-H test (95% CI), an indication of abrupt changes in the data. The assumption of the non-existence of the trend in the data was rejected based on the M-K rank correlation tests. Moreover, for all the dairy pasteurized milk, the successive monthly I-131 data were positively related. All the p-value were all less than α=0.05. The rejection of the assumption of the non-existence of trends (Ho) implied that statistically the chances of the existence of meaningful trends was significant (CI: 0.95). The MK test statistic (S) and Theil-Sen's (T-S) slope were consistently positive, indicating that there were general increases in the of trends I-131 data of all the cities [13].

While the cities were different, the M-K test for drinking water yielded mixed results from those observed in pasteurized milk. Two cities, Cedar Rapids (IA) and Jefferson (MO), did not statistically support the existence of meaningful trends in the data (CI: 0.95). The p-values for these cities were 0.8802 and 0.0500, respectively [14].

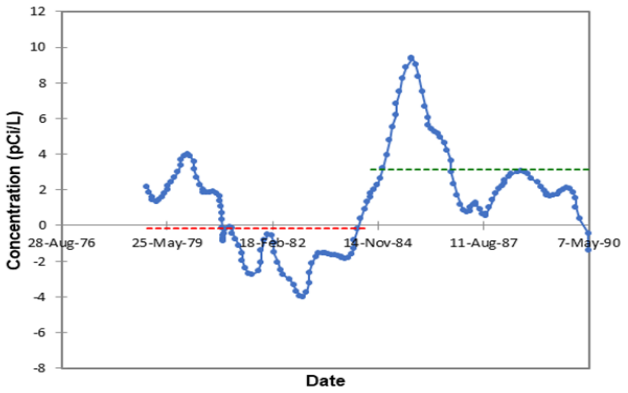
The results from the P-H homogeneity test (α=0.05) indicated that time series were not homogeneous for both the milk and water I-131 data. There were statistically significant differences in I-131 levels in pasteurized milk between the periods 1978-1984 and 1984-1990, respectively (Figure 3) [15].



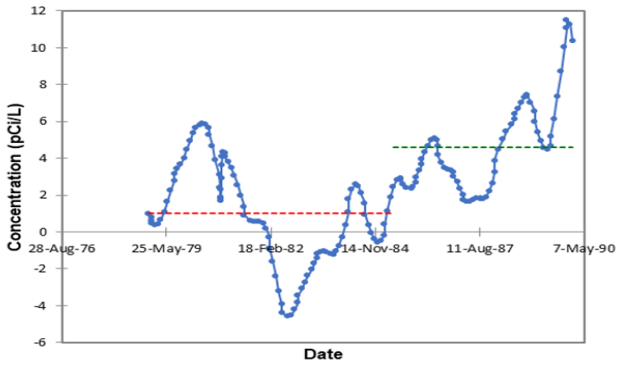
a. DES MOINES, IA (Note: DM_Gaussian Smoothed mu1= 0.721 mu2=4.682)



b. OMAHA, NE(Note: OM_Gaussian Smoothed mu1=-0.119 mu2=4.302)



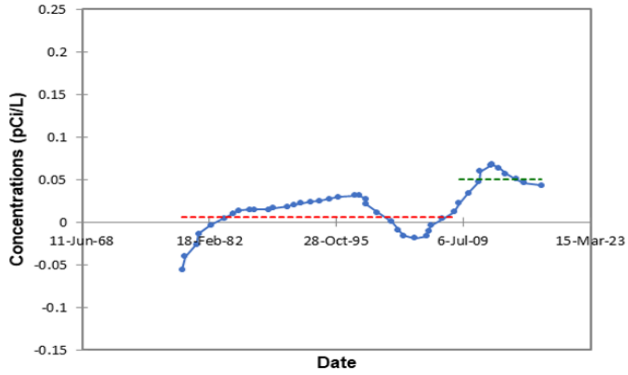
c. SAINT LOUIS, MO (Note: SL_Gaussian Smoothed mu1=-0.168 mu2=3.145)



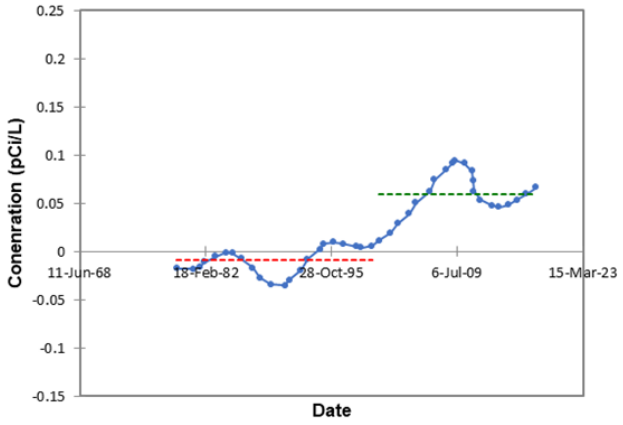
d. WICHITA, KS (Note: WI_Gaussian Smoothed mu1= 1.004 mu2=4.572)

Figure 3. Trends for P-H tests for the two cities indicate abrupt increase in I-131 levels in pasteurized milk around 1984.

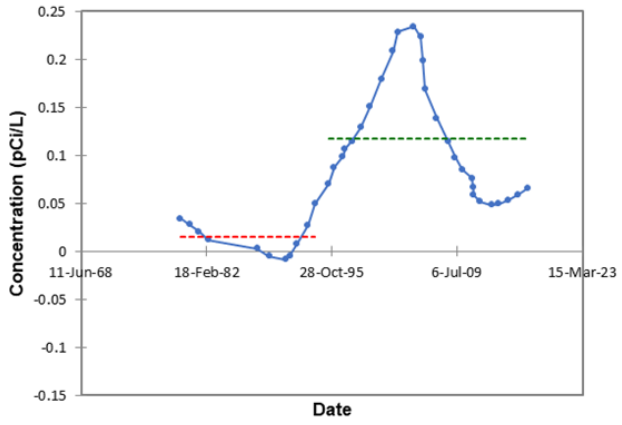
The P-H test of the G-K smoothed annual I-131 concentrations in drinking water yielded similar results in terms of the statistical non-homogeneity of the trends. In all the cities, two segment of time series were observed. Among the four cities, only Jefferson (MO) had a different pattern and the highest level of I-131 pollution level in drinking water (Figure 4).



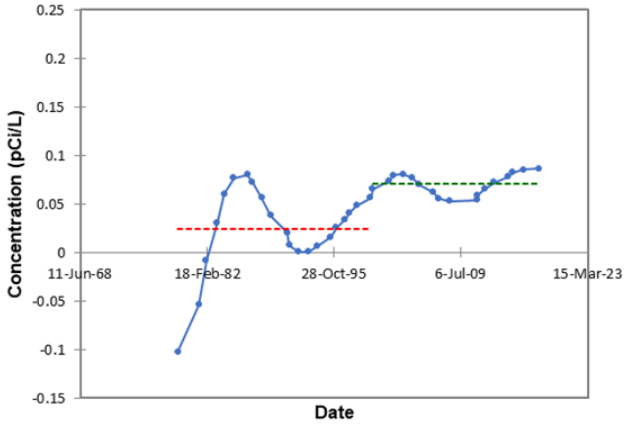
a. CEDAR RAPIDS, IA (Note: CR_Gaussian Smoothed mu1= 0.006 mu2=0.051)



b. LINCOLN, NE (Note: LI_Gaussian Smoothed mu1= -0.009 mu2=0.059)



c. JEFFERSON, M (Note: JF_Gaussian Smoothed mu1= -0.015 mu2=0.117)



d. TOPEKA, KS (Note: TO_Gaussian Smoothed mu1= -0.024 mu2=0.070)

Figure 4. Trends for P-H tests for the two cities indicate abrupt increase in I-131 levels in drinking water.

Discussion

This study had two major aims, namely, understanding how I-131 varied between 1979 and 1990, which was also the time during which several nuclear tests were extensively conducted in the atmosphere world-wide, and determining the existence of the trends in the times series [16].

I-131 concentrations in milk

The findings from this study indicate iodine radioactive isotope levels were not stable between 1978 and 1990. Studies have shown the presence of I-131 in the soils, trees, grass, water, and other living organisms. This radioactive contamination follows a cyclic path in which the I-131 falls to the earth and combines with anything in the earth's ecosystem. For instance, due to the Chernobyl accident (1986), studies showed that in the dairy farms that received higher doses of I-131 radiation, dairy products were also heavily contaminated. Based on the water, plants, animals, and air samples taken at several different times, the results showed that the milk samples from the cows that were stalled had less concentrations of I-131 radiation than the milk from the grazing cows. Since the radiation was mostly dispersed by the wind, most soils were contaminated as well. While some of the I-131 particles may have fallen directly onto the grass, the short half-life would have caused the radioiodine particles to decay within a few days after deposition. There is widespread agreement that most of the I-131 found its way into grass through water and nutrient uptake. For instance, after the reactor meltdown in Fukushima, vegetation and grazing lands thousands of kilometers away from the plant were affected, such that the sale and consumption of pasteurized milk was banned in the affected areas. The author also reported that the tests of I-131 on some spinach samples exceeded the recommended radiation exposure levels by as high as 27 times [17,18].

The P-H test of the G-K smoothed annual I-131 concentrations in drinking water yielded similar results in terms of the statistical non-homogeneity of the trends (5% level of significance). In all the cities, two segments of time series were observed. Among the four cities, only Jefferson (MO) had a different pattern and the highest level of I-131 pollution level in drinking water [19].

Our data showed that the levels of I-131 in pasteurized milk spiked between the periods 1978-1984 and 1984-1990. We note that the uptake of I-131 and the nuclides that produce it is a long-term process. Thus, it is worth to note that there was likely a time lag between radioactive fallout and milk production and distribution.

Therefore, the peaks found in the data may not coincide with the time during which the grass and water sources for dairy animals were contaminated. The wind direction during nuclear fallout also determined the extent of radionuclide pollution. Thus, to understand the dynamics of the I-131 and the level changes in the ecosystem, a systematic review of the rainfall patterns, wind direction, leakages from nuclear reactors and dumpsites is required.

Also, since I-131 is a common byproduct of Uranium-235 nuclear fission and electron capture in Tellurium-130 targets, monitoring their levels may provide additional information about the observed levels of I-131 in dairy milk. The I-131 contamination levels for all the cities sharply decreased towards 1990 in two cities, Des Moines and Saint Louis. The absence of data after that period made it difficult to understand the levels after 1990. While most data entries were recorded as not detected, more sensitive instruments and continuous measurement would help to understand the patterns of I-131 concentrations in dairy milk. Wichita recorded the highest average contamination levels compared to the other cities [20,21].

I-131 concentrations in drinking water

The concentrations of I-131 in drinking water were generally low during 1978-1981 for all cities, except Jefferson City. Moreover, comparatively, Jefferson (MO) and Lincoln (NE) had relatively higher average concentrations of I-131 than Cedar Rapids and Topeka. The sources of I-131 detected in lakes, rivers, and reservoirs include direct deposition on the ground and water surfaces. In several states, rainwater has been reported as a major source of I-131. The dispersed radionuclide particles from nuclear testing sites or reactor accidents often fall to the ground with rainwater. For instance, in 2011, several states elsewhere in the United States found elevated levels of I-131 in air following the Japan nuclear accident. A study indicated that the US Midwest was on the pathway of nuclear winds from nuclear testing centers in Western United States. The National Cancer Institute (1997) also reported that while large radionuclide particles are mostly deposited on the ground, the small particles can remain in the atmosphere/stratosphere for several years, and precipitation is one of the ways the small particles can be deposited to the ground. Based on the data analyzed in this study, the highest average concentration of I-131 was 0.089 pCi/L ± 0.176 pCi/L. Also, floods are common in the Midwest, therefore, any radioactive material from the nuclear dumpsites, leaking nuclear reactors could contaminate ground water and grazing lands [22].

I-131 levels in dairy milk *vs.* drinking water

The radioiodine concentrations were consistently higher in pasteurized dairy milk than in drinking water in all the cities. Time lag between production and distribution is very small to allow the nuclear decay of I-131 in the pasteurized dairy milk. Due to the short half-life of I-131, drinking water is mostly kept storage tanks allowing the radio iodine to decay. Even when the level was below the federal standard, the concentration of I-131 in drinking water increased after 2009.

However, the FDA detection limit for pasteurized dairy milk was set higher than that for drinking water. Moreover, states that had high levels of I-131 in milk did not have high I-131 concentrations in drinking water. In addition, milk consistently contained higher concentrations of I-131 than drinking water.

Our study shows that people that regularly drink pasteurized milk (mostly children) have higher risk of being affected by I-131 than those (adults) who drink water. Moreover, there is need to review how the contamination levels would vary with time during the production to distribution period. Also, causes for the seasonal variations in the milk data require further investigation. Considering that I-131 is more likely to affect children with developing thyroid glands, there is need to reduce the detection limit to that of drinking water [23].

Conclusion

The scope of this study was limited by missing I-131 data, particularly on pasteurized dairy milk. Moreover, few cities have been consistently monitoring the level of radioactive iodine, making it more difficulty to statistically analyze the data. While these constraints the generalization of these results to other nearby cities, the study revealed a statistically significant spike in the level of radioactive iodine between 1984 and 1986. Thereafter, the concentrations decreased until 1990. Therefore, it will be more informative to perform a similar study for the period between 1990 to present if the data were available for the same cities, considering that the state has been a domestic supplier of I-131 used in the diagnosis and treatment thyroid cancer and hyperthyroidism for the past 30 years. Finally, while the I-131 concentrations in both drinking water and pasteurized milk were relatively low compared to the EPA's Maximum Contaminant Level (MCL) of 4 millirems per year (equivalent to an average of 3 pCi/L) for beta particle and photon radioactivity from manmade radionuclides in drinking water (EPA, nd), there is need to understand the sources of I-131 causing the heterogeneity in the time series. Moreover, further analysis is needed to understand the causes for the seasonal variations in the data.

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