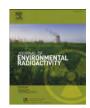
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An analysis of the intake of iodine-131 by a dairy herd post-Fukushima and the subsequent excretion in milk



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ABSTRACT

This study focuses on iodine-131 detected in milk samples from the Dairy Science Unit at Cal Poly, San Luis Obispo, California following events at the Fukushima Dai-ichi Nuclear Power Plant in March of 2011. The milk samples, collected between March 21 and April 11, 2011, were part of the Diablo Canyon Nuclear Power Plant Radiological Environmental Monitoring Program. A correlation is made between the integrated activity of iodine-131 found in milk and the integrated activities of iodine-131 of rainwater, vegetation and air samples that were collected from March 19 to April 18, 2011. A comparison is then made to previous studies conducted on dairy cattle that were administered controlled amounts of iodine-131 through ingestion. The comparison shows good agreement to the model which states that generally 1 percent of the activity of iodine-131 ingested by dairy cattle will be detected in harvested milk. Considering the environmental factors and the uncertainties involved, these data and calculated results derived from a real world situation provide an excellent application and confirmation of studies done under controlled settings.

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1. Introduction

1.1. Objective

The purpose of this study is to account for and make comparisons with the activity of iodine-131 that was detected in the milk samples from the dairy at Cal Poly, San Luis Obispo, California. This will be done by taking a select set of data comprised of the results of air, vegetation and rainwater samples collected through the Diablo Canyon Power Plant Radiological Environmental Monitoring Program and fitting exponential regression curves to each sample set. These functions will then be integrated to calculate a total amount of activity present in the system over a period of selected days. Applicable daily consumption rates of feed, water and air will be multiplied by each integrated activity resulting in amounts in units of becquerel. Then the amount of activity detected in milk will be divided by the sum of the activity detected in feed, water and air. The result will be compared to transfer coefficients and activity percentages derived from previous studies.

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1.2. Background

On March 11, 2011, the Tohoku earthquake with a magnitude of 9.0 (U.S. Geological Survey, June 2013), struck the east coast of Japan, generating a massive tsunami that disabled the backup power and control systems of the Fukushima Dai-ichi Nuclear Power Plant operated by Tokyo Electric Power Company (TEPCO). The plant was rendered virtually helpless and on its way to what was eventually categorized as a Class 7 rated accident on the International Nuclear and Radiological Event Scale (INES). This INES classification for Units 1, 2 and 3 was upgraded from a Class 5 event (International Atomic Energy Agency, May 2012). In the weeks following the event, the fission product iodine-131 was detected in the environment around Fukushima (Shozugawa et al., 2012).

Within days of the disaster, on the west coast of California, traces of iodine-131 were detected in samples acquired as part of the Radiological Environmental Monitoring Program (REMP) at the Diablo Canyon Nuclear Power Plant (DCPP). In response to the events unfolding, DCPP REMP initiated increased frequency of milk sampling from monthly to weekly. Also, supplemental air, vegetation and rainwater samples were collected at DCPP and the surrounding county.

2. Materials and methods

2.1. Collection methods

Milk samples were collected by technicians employed by Pacific Gas and Electric Company tasked with supporting the DCPP REMP. Aside from the increase in sampling frequency, the procedure remained the same and was conducted as follows: Prior to arriving at the Cal Poly Dairy, four 1 gallon sampling containers are prepared. The one container that will be sent to CDPH¹ has with it a measured amount of iodine carrier (5 ml of KI carrier solution in 15 ml of 1 M NaHSO₃) and a 100 ml allotment of 37% formaldehyde solution (Ruberu, May 2011 October 2013). The three remaining containers, to be analyzed by GEL,² are paired with a 40 g allotment of sodium bisulfite.

The milk at the dairy is stored in two 11,356 L (3000 gallon) stainless steel tanks. Upon arrival at the dairy facility, the four sampling containers are arbitrarily divided to where two are filled from one of the tanks and the other two are filled from the other tank. Before filling the sample that is collected for CDPH, the iodine carrier is added to the container. After the CDPH container is filled, the formaldehyde is added. The other three containers, collected for analysis by GEL, are first filled, then the 40 g of sodium bisulfite is added. After collection, the samples are packaged and shipped to their respective laboratories for analysis. The CDPH sample is sent overnight and the GEL samples are sent via two day delivery. Shipping methods are mentioned due to the nature of iodine-131 decay which is taken into consideration by the testing laboratories.

Air samples are normally collected on a weekly basis and it remained so throughout the study period. However, four additional air sampling pumps were deployed after the Fukushima event. These supplemental air sampling pumps were co-located with four of the existing stations. The co-locations were at 0S2, 8S1, 7D1 and 5F1. In addition, the four supplemental air sampling pumps were deployed with an increased flow rate of 3.40 m³/hr. Normally they are set with a flow rate of 2.55 m³/hr.

An air sample station consists of an F&J Specialty Products, 0.25 horsepower oil-less, carbon vane vacuum pump with a flow meter, a vacuum gauge and a constant airflow regulator. This is connected to a filter holder containing a TEDA impregnated charcoal cartridge, for the collection of gaseous radioiodine, and a glass fiber filter, for the collection of particulate radioiodine, positioned in front of the charcoal cartridge. The entire low volume air sampling system is located within a vented metal box stand called a *doghouse*.

The four supplemental air samplers were placed within the same doghouses as the existing samplers. The charcoal cartridges and glass fiber filters are replaced weekly. Charcoal cartridges are placed individually in plastic zip-lock bags, the glass fiber filters are placed in paper envelopes, all of which are placed in a zip-lock bag and they are all shipped overnight to GEL. None of the air samples discussed in this report were sent to CDPH as they operate and maintain similar air sampling stations throughout the region. The results from GEL are reported as an integration of the entire sampling week. The date of sampling will correspond to a mid-point in the week-long sampling period. For example, the results reported as sampled on 3/19/11 would be from charcoal cartridges deployed on 3/16/11 and removed from the field on 3/23/11. For this study, the reported integrated dates from GEL will be used in the integrated activity calculations covered in the Results portion of this paper. Of the four types of samples discussed in this report (milk, air, vegetation and rain water), air samples are the only ones

Table 1Types of vegetation sampled at each location.

Location	Sample type
7C1	Mustard Weed, Coyote Brush
6C1	Garden Crops, Citrus Leaves
5F2	Commercial Crops
ATAS	Milk Thistle, Fescue Grass, Miner's Lettuce
7G1	Commercial Crops

reported in this manner. Furthermore, only the glass fiber filters associated with the four additional co-located air pumps were analyzed for particulate iodine-131 by GEL.

Vegetation sampling consisted of grab samples taken from local crops or annual grasses and plants collected from various locations. The samples were collected, placed in plastic bags and shipped via two-day delivery to GEL. Various types of vegetation were collected depending on location and what was available at the time. Of the five sample types of samples in this study, vegetation had the widest distribution in terms of geographical area covered. Table 1 shows the sample matrix of available vegetation during the study period.

Rainwater was collected as a supplemental sample as it is not part of the normal REMP matrix. However, in the wake of the Fukushima event it was deemed necessary. To accomplish this, plastic paint trays were purchased from a local hardware store and placed at four locations. Three of the sampling locations were near DCPP and the fourth was located in San Luis Obispo. At each location approximately 10 trays were laid out and each day after experiencing rainfall in the area, the contents of the paint trays were poured into 1 gallon bottles. The bottles were then shipped via two-day delivery to GEL. The paint trays remained in the field at their respective sampling locations throughout the study period.

2.2. Analysis methods

GEL conducted analysis on all sample types in this study. Gamma analysis was performed with a high purity germanium detector (HPGe) utilizing methods from the Department of Energy (DOE) EML Procedures Manual, HASL-300, I-01. If necessary, iodine was separated by absorption on AG 1×8 anion resin. For the analysis of water samples, the methods followed were from EPA 600/4-80-032 Prescribed Procedures for Measurements of Radioactivity in Drinking Water, Method 901.1. Minimum Detectable Activity (MDA) for iodine-131 varied due to the short half-life and the length of time between sample collection and counting. Relative Percent Difference (RPD) was 20% or less or 100% or less if the activity was less than five times the MDA. The criteria for the assessment of method bias was \pm 25% of true value on laboratory control samples that were included with each batch (General Engineering Labs, August 2013).

Analysis conducted by CDPH involved transferring the sampled milk to 4 L Marinelli beakers and counting on a 50% relative efficiency HPGe for 16 h. The method was in accordance with EPA Method 901.1 Analysis for Gamma Emitting Radionuclides in Drinking Water. If lower detection limits were desired, the inclusion of the iodine carrier (KI) would provide an avenue for applying Standard Methods 7500-I C, in which radioiodine is converted to iodide and concentrated on an anion resin, then counted on a betagamma coincidence system (Ruberu, May 2011 October 2013). In this case, an HPGe was used for the milk samples. CDPH only

¹ California Department of Public Health, Richmond, California.

² GEL Laboratories, Charleston, South Carolina.

³ Manufactured by Bio-Rad Laboratories, Life Science Group.

analyzed milk in this study and had one instance of a result above the minimum detectable activity within the study period (from 3/19/11 to 4/18/11).

3. Results

The data obtained from GEL and CDPH were submitted by DCPP to the Nuclear Regulatory Commission and are available on the NRC website as a collection of PDF files (U.S. Nuclear Regulatory Commission (NRC), June 2013a). The following Tables 2–6 and Figs. 1–5 display the iodine-131 results from the various sample matrices taken from these data (U.S. Nuclear Regulatory Commission (NRC), June 2013b) (U.S. Nuclear Regulatory Commission (NRC), June 2013c). The following charts (Figs. 1–5) are normalized to Day 0 being 3/19/11.

The average activity concentration graphs are displayed with error bars that are derived from the standard deviation of a group of samples on that particular day. Error bars are not displayed in the average charts for days when only one result was available. All sample results have their respective 2σ TPU or CE values in the data tables.

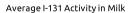
Not included in the charts or data tables are some positive iodine-131 results that fell outside of the study date range. An air charcoal cartridge with an integrated sampling date of March 17, 2011, showed a positive result for iodine-131 at station 7D1-2. The amount detected at this station was 0.0177 Bq/m 3 (\pm 0.00165).

On May 2, 2011, after two weeks of results that were below detection limits from both laboratories, a final instance of iodine-131 detection in Cal Poly milk occurred in samples analyzed by both GEL and CDPH. GEL reported 0.0577 Bq/L (± 0.0193) and CDPH reported 0.153 Bq/L (± 0.0337). There were no other corresponding air, vegetation or water samples that showed positive results for radioiodine.

All of the raw data activities have been converted to SI units from picocuries with the volumes remaining in SI units. The individually charted data points are shown with their respective error. GEL reports error as 1.96 times the Total Propagated Uncertainty (TPU) (Trent, May 2012 October 2013). This is displayed as 2σ TPU.

Table 2Cal Poly milk sampling results (units in Bq/L).

	<u> </u>			
Analytical laboratory	Date collected	Result	MDC/MDA ₉₅	2σTPU/CE
GEL	3/21/11	1.48E-01	1.67E-02	2.06E-02
GEL	3/21/11	2.51E-01	1.22E-01	1.07E-01
GEL	3/28/11	6.55E-02	1.56E-02	1.65E-02
CDPH	3/28/11	1.23E-01	9.69E-02	2.18E-02
GEL	4/4/11	3.41E-02	1.74E-02	1.29E-02
GEL	4/11/11	2.87E-02	1.51E-02	1.57E-02



Error bars are standard deviation (where possible)

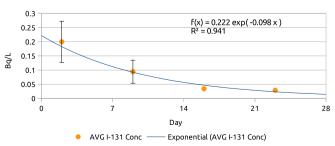


Fig. 1. Averaged milk sampling results from the Cal Poly Dairy.

Table 3Rainwater I-131 results (Bq/L).

Location	Date collected	Result	MDC	2σTPU
0S2	3/19/2011	3.56E + 00	1.02E-01	3.50E-01
1S1	3/21/2011	8.66E + 00	1.11E-01	8.03E-01
5S2	3/21/2011	8.03E + 00	9.92E-02	7.25E-01
5F1	3/21/2011	1.02E + 01	1.14E-01	9.44E-01
1S1	3/23/2011	2.55E + 00	1.10E-01	2.75E-01
5S2	3/23/2011	2.62E + 00	1.23E-01	2.80E-01
5F1	3/23/2011	4.59E + 00	1.20E-01	4.63E-01
1S1	3/24/2011	1.75E + 01	1.26E-01	1.59E + 00
5S2	3/24/2011	1.93E + 01	1.17E-01	1.70E + 00
5F1	3/24/2011	6.77E + 00	1.19E-01	6.36E-01
1S1	3/25/2011	8.40E + 00	1.41E-01	7.81E-01
5S2	3/25/2011	8.70E + 00	1.22E-01	8.33E-01
5F1	3/25/2011	3.63E + 00	1.15E-01	3.60E-01
1S1	3/28/2011	5.77E + 00	9.58E-02	5.66E-01
5S2	3/28/2011	5.18E + 00	1.02E-01	5.07E-01
5F1	3/28/2011	5.81E + 00	9.88E-02	5.51E-01
5F1	4/9/2011	5.14E-01	9.55E-02	1.14E-01

Average Rainwater I-131 Activity Concentration

Error bars are standard deviation (where possible)

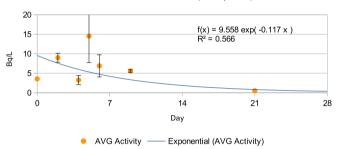


Fig. 2. Average of rainwater sample activities.

Table 4 Vegetation I-131 results (Bq/kg).

Location	Date collected	Result	MDC	2σTPU
ATAS	3/27/2011	1.71E + 01	5.59E-01	1.81E + 00
ATAS	3/27/2011	7.14E + 00	3.81E-01	7.96E-01
ATAS	3/27/2011	1.28E + 01	3.06E-01	1.28E + 00
5F2	3/28/2011	4.22E + 00	3.11E-01	5.40E-01
ATAS	4/3/2011	6.03E + 00	5.77E-01	8.47E-01
ATAS	4/3/2011	1.89E + 00	5.33E-01	6.14E-01
ATAS	4/3/2011	7.70E + 00	4.66E-01	9.25E-01
7G1	4/4/2011	1.41E + 00	4.59E-01	4.37E-01
7C1	4/4/2011	4.26E + 00	3.65E-01	6.03E-01
5F2	4/4/2011	1.76E + 00	5.85E-01	6.25E-01
5F2	4/11/2011	1.07E + 00	5.88E-01	5.11E-01
6C1	4/14/2011	2.50E + 00	5.33E-01	5.00E-01
5F2	4/18/2011	6.99E-01	3.89E-01	4.66E-01

CDPH reports Counting Error (CE) which is defined as counting error at the 95% confidence level from EPA-600/4-80-032 (August 1980), Prescribed Procedures for Measurement of Radioactivity in Drinking Water.

Minimum Detectable Concentration (MDC), the term utilized by GEL, is calculated in Equation (1) (General Engineering Labs, March 2012):

$$MDC = \frac{3.29*\sqrt{\left(Bkgcpm*C_t*\left(1 + \frac{C_t}{BkgC_t}\right)\right)} + 3.0}{2.22*E*C_t*V*decay*A*R*DF*I}$$
(1)

where:

Average I-131 Activity Concentration

From Vegetation Samples. Error bars are standard deviation (where possible).

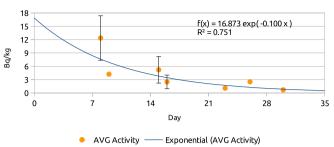


Fig. 3. Average of vegetation sample activities.

- $C_t = \text{Sample count time}$
- $BkgC_t$ = Background count time
- Bkgcpm = Background counts per minute (cpm)
- E = Counting efficiency
- V =Sample volume or weight
- *decay* = isotopic decay
- *A* = Isotopic abundance
- R = Recovery
- *DF* = Dilution factor for liquid scintillation
- I = Additional decay or ingrowth factors

CDPH utilized the MDA₉₅ method, which is the sample specific

Table 5 Air charcoal cartridge I-131 results (Bq/m^3) . Co-located samples designated with "-2" in location name.

-2 III location hame.						
Location	Date collected	Result	MDC	2σTPU		
0S2	03/19/11	5.48E-02	5.59E-04	5.03E-03		
0S2	03/26/11	1.33E-02	3.81E-04	1.43E-03		
0S2	04/02/11	2.42E-03	3.41E-04	5.22E-04		
1S1	03/19/11	5.59E-02	5.44E-04	5.62E-03		
1S1	03/26/11	1.47E-02	4.66E-04	1.74E-03		
1S1	04/02/11	2.67E-03	6.36E-04	7.96E-04		
1S1	04/09/11	9.29E-04	8.92E-04	7.88E-04		
MT1	03/19/11	5.88E-02	6.29E-04	5.55E-03		
MT1	03/26/11	1.36E-02	3.42E-04	1.51E-03		
MT1	04/02/11	2.88E-03	4.11E-04	6.36E-04		
MT1	04/09/11	6.14E-04	5.59E-04	5.00E-04		
8S1	03/19/11	5.62E-02	5.51E-04	5.14E-03		
8S1	03/26/11	1.37E-02	4.77E-04	1.68E-03		
8S1	04/02/11	2.28E-03	4.37E-04	6.22E-04		
8S2	03/19/11	5.88E-02	8.40E-04	6.99E-03		
8S2	03/26/11	1.30E-02	3.74E-04	1.48E-03		
8S2	04/02/11	2.50E-03	6.81E-04	7.40E-04		
7D1	03/19/11	6.22E-02	4.48E-04	6.22E-03		
7D1	03/26/11	1.26E-02	3.66E-04	1.49E-03		
7D1	04/02/11	2.25E-03	4.18E-04	4.96E-04		
5F1	03/19/11	5.70E-02	4.22E-04	5.25E-03		
5F1	03/26/11	1.41E-02	4.18E-04	1.51E-03		
5F1	04/02/11	2.83E-03	2.14E-04	5.92E-04		
0S2-2	03/20/11	5.77E-02	3.77E-04	5.66E-03		
0S2-2	03/26/11	1.46E-02	2.58E-04	1.48E-03		
0S2-2	04/02/11	2.32E-03	2.62E-04	4.37E-04		
8S1-2	03/20/11	5.07E-02	3.85E-04	4.66E-03		
8S1-2	03/26/11	1.32E-02	3.10E-04	1.33E-03		
8S1-2	04/02/11	1.96E-03	2.89E-04	3.77E-04		
7D1-2	03/17/11	1.77E-02	2.33E-04	1.65E-03		
7D1-2	03/21/11	6.55E-02	4.77E-04	5.77E-03		
7D1-2	03/26/11	1.16E-02	2.12E-04	1.23E-03		
7D1-2	04/02/11	2.16E-03	2.41E-04	4.88E-04		
5F1-2	03/19/11	6.11E-02	4.88E-04	5.62E-03		
5F1-2	03/26/11	1.21E-02	3.16E-04	1.27E-03		
5F1-2	04/02/11	2.73E-03	3.57E-04	5.40E-04		
5F1-2	04/16/11	3.69E-04	3.69E-04	3.08E-04		

Average I-131 Air Charcoal Activity Concentration

Error bars are standard deviation (where possible)

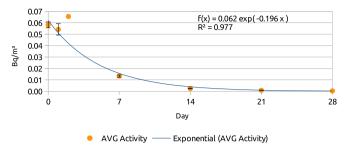


Fig. 4. Average of air charcoal cartridge activities.

Table 6 Air particulate filter I-131 results (Bq/m^3). Co-located samples designated with "-2" in location name.

Location	Date collected	Result	MDC	2σTPU
5F1-2	3/19/2011	3.33E-03	2.09E-04	4.88E-04
0S2-2	3/20/2011	2.97E-03	1.84E-04	4.37E-04
8S1-2	3/20/2011	3.03E-03	1.99E-04	4.37E-04
7D1-2	3/21/2011	2.79E-03	3.50E-04	4.88E-04
0S2-2	3/26/2011	1.34E-03	1.83E-04	2.82E-04
5F1-2	3/26/2011	1.49E-03	1.92E-04	3.03E-04
7D1-2	3/26/2011	1.39E-03	1.82E-04	3.20E-04
8S1-2	3/26/2011	1.99E-03	1.70E-04	3.74E-04
0S2-2	4/2/2011	3.74E-04	2.93E-04	1.85E-04
5F1-2	4/2/2011	4.00E-04	2.24E-04	3.20E-04

minimum delectable activity at the 95% confidence level, or the LLD_{95} , divided by 2.22 (dpm/pCi), the yield and the efficiency. The calculation may also include abundance, decay and ingrowth factors depending on the radionuclide as well as defining S_b as the square root of the instrument background count rate. LLD_{95} is defined in Standard Methods for the Examination of Water and Wastewater, American Water Works Association, 21st Edition, 2005, section 7020C (Equation (2)).

$$LLD_{95} = 3.29\sqrt{S_b} \tag{2}$$

where:

• $S_b = Standard deviation of background counting rate (cpm)$

It must be reiterated that the methods from both GEL and CDPH include conversion from decays per minute (DPM) to picocuries (pCi) by use of the factor 2.22 (dpm/pCi). In this study the values were then multiplied by 0.037 to attain the SI unit of becquerel.

Average I-131 Activity Concentration

From Air Particulate Filters. Error bars are standard deviation (where possible)

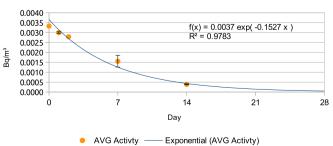


Fig. 5. Average of air particulate activities.

Values reported from both GEL and CDPH were reported to the level of three significant figures. GEL reported MDC and 2σ TPU while CDPH reported MDA₉₅ and CE (Table 2).

4. Discussion

4.1. Previous studies

In a study conducted by Lengemann and Comar (1964), cows were fed encapsulated iodine-131 under controlled conditions and the levels of iodine-131 were measured in milk, thyroid and whole body. An important factor in this study was that the batch of iodine-131 was prepared and then allowed to decay throughout the study period while being administered in feed (Lengemann and Comar, 1964). This matched the conditions, to a degree, of the plume experienced at the Cal Poly Dairy which, with the help of precipitation, deposited radioiodine over quantified portions of the feed and water supply of the herd. The bulk of the precipitation and deposition occurred over a short period of time, thus mimicking a single contamination event. The outcome of the Lengemann study showed that generally 1% of the iodine-131 ingested was detected per liter of milk.

In another study by Bustad et al., cows were fed iodine-131 and it was observed that 0.4–1.0% of the daily activity ingested was detected in milk. A simulated single contamination event was also attempted by allowing the initial batch of radioiodine to decay throughout the study. Furthermore, part of the study included the feeding of radioiodine labeled grain in addition to other feed sources (Bustad et al., 1963).

In a later study on dairy cows, transfer coefficients ranging from 0.2% to 2.7% per liter of milk, with 90% of the results lying between 0.5% and 2.0% per liter were determined. In this study, cows were also given encapsulated radioiodine. The overall determination was that the transfer coefficient for iodine-131 was 0.88% per liter of milk (Sam et al., 1980). The concept of the transfer coefficient is similar to that of stating a percentage in that it represents the fraction of the daily intake of radionuclide that is detected in milk.

Considering air inhalation contribution to the milk activity, transfer coefficients for the inhalation pathway are generally regarded as orders of magnitude lower than the ingestion pathway. One study suggests that there is a possibility that the inhalation pathway could be underestimated; however, this considers dust loads nearer to ground level that could contain higher

concentrations of radionuclides and inhalation while feeding (Zach, 1985). Though, in this case, with the confinement of the herd (not on pasture), feeding in troughs or on a cement passageway through feeding slots in a fence and presumably lower dust loads during periods of seasonal precipitation, the inhalation pathway will prove to be a small contributor to the activity found in milk in this study.

In all, a preponderance of studies have been conducted that consider a two compartment model whereby the pathway is pasture vegetation through cows to milk. Shaeffer (Shaeffer, 1981) offered least-squares fits to data acquired after pulses from nuclear weapons testing by the Peoples Republic of China in the 1970's. That approach is similar to what is shown in this report. However, in this study it will be shown that amount of exposed feed consumed and the activity detected in milk would not agree with the models mentioned previously without the consideration of water intake from exposed troughs. Not considering water exposed to rain washout of iodine-131 could explain the higher milk concentration to vegetation concentration equilibrium values (C_m/K_v) calculated by Shaeffer.

4.2. Weather & sampling time-line

Fig. 6 shows the distribution of rain that fell throughout the region during the study period. The rainfall amounts are averaged from five weather stations across the region. Four weather stations are part of the California Irrigation Management Information System (CIMIS) (California Irrigation Management Information System, August 2013). The CIMIS data is from stations 163, 160, 52 and 202. The last set of data is from the DCPP meteorological station (Lindsey, June 2012). The error bars are the standard deviation of the rainfall averages. Also pictured are the positive result time-lines from the samples collected during the study period.

4.3. Cal Poly Dairy

At the time of the Fukushima event, the Cal Poly Dairy was operating with approximately 200 cows in production. The cows produce between 30 and 32 L of milk per day (Silacci, April 2011 October 2013) which is stored in two 11,356 L tanks (3000 gallons) and periodically transferred to a truck that transports the milk to various dairy product facilities. The samples were collected from both tanks and sent for analysis to GEL Laboratories, LLC (GEL) in Charleston, South Carolina or to the California Department of Public

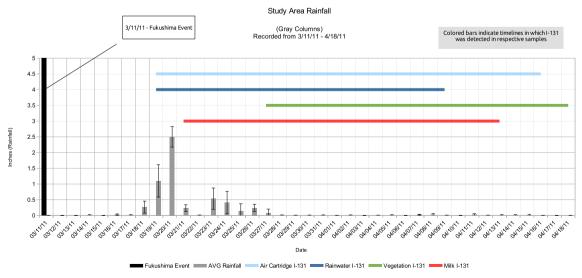


Fig. 6. Rainfall averages displayed with I-131 result time-lines.



Fig. 7. DCPP, Cal Poly Dairy and sampling locations.

Health, Drinking Water and Radiation Laboratory Branch (CDPH). GEL is a large quality certified environmental solutions company that processes and analyzes environmental samples for many other power plants, Department of Energy sites and industrial manufacturers. CDPH performs parallel analysis of many of the same samples that are collected through the REMP.

Fig. 7 shows a map of the region, the various sampling locations and the geographical locations of DCPP and the Cal Poly Dairy. In this region, the prevailing winds and storms (during the Spring) commonly originate from the northwest. Geographically, the mountain ranges adjacent to DCPP and the Cal Poly Dairy are aligned at similar angles, thus reducing the effects that topography might have on the plume that arrived from across the ocean. Later in this study it will be seen that the coastal mountains had little effect on the levels of iodine-131 seen in rainwater and vegetation because of the similar levels found in samples collected in Atascadero, California which is located in a region beyond the direct influence of coastal winds.

Initially it was assumed that the presence of iodine-131 in milk was primarily the result of the consumption of contaminated vegetation by cows while on pasture. This exposure was assumed to be correlated with the iodine-131 detected in REMP vegetation samples deposited via the airborne-precipitation pathway. During the time of increased sampling in response to the events occurring in Japan during March and April of 2011, observations were made and discussions held with the dairy herd manager that indicated this was not an accurate assumption. The main issue was that the herd had no access to pasture with exposed vegetation.

Fig. 8 shows the layout of the Cal Poly Dairy. The dry lot area marked D on the aerial photo is where the cows are kept while in production. Twice daily they are brought to the milking facility (C)

where they produce between 30.3 and 32.2 L (8–8.5 gallons) of milk per cow per day. The dairy cow feeding regime consists of four parts (Table 7) (Silacci, April 2011 October 2013) which are fed under the shelter of the main barns in area D.

The two grain allotments were not exposed to deposition via the airborne-precipitation pathway. It was also observed that the alfalfa hay supply was located under a covered pole-barn structure (B) and would be exposed to very limited deposition from precipitation, if at all. However, it was observed that the silage pile (A) was left exposed on the end that was being utilized for daily feeding (Fig. 9). It is therefore assumed in this study that the silage used for feed was exposed to airborne and precipitation deposition of iodine-131 at the same general level that was observed in the REMP vegetation samples.

Upon further inspection of the dairy facility it became apparent that of the seven water troughs that the production cows had access to, three of the seven were located at the edge of the barn structures (Fig. 10). In these locations the water troughs were effectively exposed to rain and roof runoff. Therefore, another assumption that is made in this study is that 43% of the water that the herd had access to was exposed to similar levels of rainwater activity as the REMP rainwater samples that were collected. This assumption is supported by the timing of the peak activity of the air plume detected on March 19th, 2011 and the beginning of the largest precipitation event which spanned from the 19th to the 20th of March as well as the subsequent rainfall that followed. All water troughs hold approximately 190 L (50 gallons) and are operated with float valves. For the purpose of this study, iodine-131 infiltration in the three exposed water troughs will be considered equal to the rainwater collected in the paint trays, bringing them within the range of activity concentrations seen in the REMP samples.



Fig. 8. Aerial view of the Cal Poly Dairy [Google, 2013].

Table 7Cal Poly Dairy herd feed ration (Silacci, April 2011 October 2013).

Feed type	Amount/day (kg) per cow	Percentage by mass
Alfalfa Hay	9.1	23.6
Silage	9.1	23.6
Mixed Dry grains	13.6	35.2
Wet Brewer's grain	6.8	17.6

In this study the herd was considered a single entity and both milk storage tanks were considered as a single compartment with an outflow rate that is slightly greater than the inflow rate. By considering the herd as a single entity, any instances of fluctuation of feed or water intake caused by sickness, such as mastitis (Sam et al., 1980), or unforeseen animal issues would be smoothed by the large number of cows. Therefore, it was assumed that the uncertainties in the feed regime were negligible. The outflow/inflow assumption is evidenced by the effective half-life of the iodine-131 in the sampled milk of 7.1 days as calculated from the exponential regression formula in Fig. 1 and the fact that the milking rate did not exceed the rate of transference to milk facility trucks, thus halting production. The activity detected in the milk holding tanks is considered an equilibrium activity concentration in the herd as a whole, affording the opportunity to apply the daily intake rates of feed, water and air for a single cow in the total activity calculations.

Iodine-131 detected in air charcoal cartridges and air particulate filters was shown to be lower by orders of magnitude. Nevertheless, the integrated activities of the two will be considered and calculated based on the average inhalation rate from Zach (1985). Table 8 describes intake rates of feed, water and air that will be used in the calculated total activities.

The amount of water consumed per day has been adjusted by multiplying 0.43 by the 170 L/day average intake (Silacci, April 2011 October 2013) of producing dairy cows. This is to account for the amount of water troughs (3 of 7) exposed to rain and roof runoff.

Utilizing the regression equations calculated from the plotted data, the next step is to integrate each equation over the range of days in the study period. Here, the regression equations are



Fig. 9. Exposed silage.

reformatted to familiar notation.

$$A(t) = A_0 e^{(-\lambda t)} \tag{3}$$

Then Equation (3) is integrated to become:

$$\tilde{C} = \int_0^t A(t)dt = \frac{A_0}{\lambda} \left(1 - e^{(-\lambda t)} \right) \tag{4}$$

where:

- \bullet \tilde{C} = Integrated Activity Concentration (Bq-days per mass or volume)
- A(t) = original exponential function
- A_0 = the peak activity at day 0 (Bq per mass or volume)
- t = 30 days
- λ = removal constant derived from regressions (day⁻¹)



Fig. 10. Water trough exposed to rain and runoff.

Then by multiplying \tilde{C} by the daily intake rates in units of volume or mass per day (Table 8), the volume (or mass) and days cancel and a total integrated activity is derived in units of becquerel. The same calculation is performed on the on the milk activity results. This is shown in Table 9 and Table 10 where the Total Activity column values are calculated by multiplying the result of Equation (4) (\tilde{C}) by the respective intake or output rate.

Finally, the milk sample total activity (66.9 Bq) is divided by the sum of the intake sample total activities (7280.7 Bq) and it is calculated that the amount of activity in milk is 0.92% of the total intake. This percentage can be considered in agreement with or within the range of results attained by Lengemann and Comar (1964), Bustad et al. (1963) and Sam et al. (1980). Furthermore, by examining the contribution of each intake sample to the total amount of intake activity, it is shown that the rainwater influence

Table 8Dairy cow intake rates

Туре	Amount per day	
Feed (silage)	9.1 kg	
Water	72.9 L	
Air	130 m ³	

Table 9Calculated total activities for intake samples.

Sample	A_o^a	$\lambda^{\mathbf{b}}$	\tilde{C}^{c}	Intake rated	Total activity (Bq)
Air charcoal	0.062	0.196	0.315	130	41.0
Air particulate	0.004	0.153	0.026	130	3.4
Feed (vegetation)	16.873	0.100	160.329	9.1	1459.0
Water	9.558	0.117	79.250	72.9	5777.3
Sum total					7280.7

- ^a Units are Bq per mass or volume.
- b Units are day⁻¹
- ^c Units are Bq-day per mass or volume.
- d Units are mass or volume per day.

Table 10Calculated total activity for milk samples.

Sample	A _o (Bq/L)	λ (day ⁻¹)	Č (Bq-day/L)	Output rate (L/day)	Total activity (Bq)
Milk	0.222	0.098	2.146	31.2	66.9

made up 79.35% of the activity detected, the silage made up 20.04%. The air charcoal and air particulate activities made up 0.56% and 0.05% respectively.

5. Conclusion

Presented here has been a review of the DCPP REMP sampling methods and the specific results related to the iodine-131 detected in milk at the Cal Poly Dairy in March and April of 2011. It has been shown that with the help of field observations and inquiries of those with actual knowledge of the conditions under which the dairy cows attain feed and water, integrated activities can be determined that sufficiently account for the amount of activity detected in the milk. Also, the total integrated activity in milk compared to the total integrated activity taken in through ingestion of water, feed and to some extent air, is at a percentage (0.92%) that agrees with previous studies conducted on fewer animals and under controlled laboratory conditions. However, in order to have better characterized the radiological model, samples could have also been collected directly from the dairy feed and water sources.

Finally, it is important to acknowledge that the levels of iodine-131 found in milk were well below the derived intervention level (DIL) for iodine-131 of 170 Bq/kg as deemed by the U.S. Food and Drug Administration (U.S. Food and Drug Administration (FDA), September 2013). Nevertheless, with coefficients of determination ranging from 0.566 to 0.978, the regression curves and their respective integrals can be considered a good representation of the actual radiological conditions that were present at the time.

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