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

Richard M. Treinen

Oregon State University, USA

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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.

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.

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.

E-mail address: ricktreinen@gmail.com.

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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 CDPH[1] has with it a measured amount of iodine carrier (5 ml of KI carrier solution in 15 ml of 1 M $NaHSO_3$) 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 m^3/hr. Normally they are set with a flow rate of 2.55 m^3/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 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.

Table 1
Types 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

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.[3] 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 beta-gamma 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.