

ENVIRONMENTAL SAMPLE ANALYSIS BY MEANS OF
NEUTRON ACTIVATION ANALYSIS AND GAMMA-RAY
COUNTING

NE 170 PROJECT PROPOSAL

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1 Introduction

Our team aims to determine the isotopic composition of kelp, fish, and other sea life along the west coast of the Americas, particularly in the San Francisco Bay and Long Beach, though we will be studying samples from Alaska all the way to Chile. In particular, we would like to see if there are any major differences in isotopes found in samples from different parts of the West Coast, and whether the isotopes found in the different samples will present a toxicity or radiological health hazard to the peoples living along the Pacific.

We will be using neutron activation analysis (NAA) to determine the isotopic composition of our samples. NAA involves irradiating test samples with a high neutron flux in order to activate the isotopes present in the sample. These isotopes then decay, emitting characteristic gamma rays that allow us to identify the isotopes that were originally in the sample. Based on the half-life of the isotope, its branching ratio at a particular gamma energy, and the counts registered on the detector, we can also determine the amount of the isotope in the test sample. NAA provides advantages that other techniques do not. NAA is relatively unobtrusive, and will not cause significant damage to the test sample. As neutrons can penetrate deeply into a material, NAA can be used on bulk samples. These samples do not need the careful preparation that is needed for other techniques to determine isotopic composition. Thus, NAA provides a fairly accurate and precise measurement of isotopic compositions without causing physical damage to the sample, and without the hassle of extended preparation time. With these advantages come some significant disadvantages that we will have to work around. One is that materials activated by neutrons remain radioactive for a long time after irradiation took place. Dependent on the isotopes in the sample, and the half-lives of those isotopes, this can make working with activated materials hazardous. This should not be an overwhelming concern for our team, as the samples that we will be working with are not expected to contain hazardous isotopes in the concentration needed to be harmful to us. Nevertheless, presence of toxic and radiologically active materials should be noted. The other concern is that there are only a select few facilities that have the capabilities needed to perform neutron activation on test samples. Fortunately, the McClellan Nuclear Research Center (MNRC), which is able to perform neutron activation, is relatively close, an advantage that will be crucial to the success of our project.

2 Goals

Our ultimate goal in this project is to be able to communicate to the public radiological information about things they regularly interact with. Since the Fukushima incident in 2011, public concern about all things nuclear has dramatically increased. The air people breathe, the fish they eat, and the bodies of water that surround them can be analyzed radiologically. Publishing the analysis for people to see creates a more radiologically informed public. We intend to accomplish this overarching goal by meeting these more specific needs:

- Analyze recently taken NAA data on kelp from all over the West Coast
- Obtain new samples from the bay, carry out NAA on them at MNRC, and analyze those results
- Perform NAA measurements on seaweed samples
- Find new samples to perform NAA and gamma-ray analysis on
- Compile results and write article(s) on the findings
- Take important data and articles and publish them to the Radwatch website

3 Roles

Shreyas:

- Initial data analysis
- Maintain living document(s) as necessary

- Make presentation(s) as necessary
- Compile group work into LATEX
- Lead in publishing to Radwatch site

Zach:

- Initial data analysis
- Lead in communicating with instructor on a weekly basis on behalf of the group
- Lead in communicating with points of contact on behalf of the group
- Generate efficiency curve(s) for detectors being used
- Help with publishing to Radwatch site

Eduardo:

- Initial data analysis
- Find new, useful samples for more analysis
- Help with new sample data analysis
- Lead in writing article(s) to be published
- Help with publishing to Radwatch site

Peter:

- Initial data analysis
- Find new, useful samples for more analysis
- Lead in data analysis for new samples tested
- Help with writing article(s)
- Help with publishing to Radwatch site

4 Timeline

Figure 1 shows a Gantt chart timeline of the various tasks required for this project. The first task revolves around the existing NAA data. Following the procedure described in the introduction, we must identify the composition of the existing samples

Project	Tasks	Start	End	Task Lead	Days till End date	14-Feb	21-Feb	28-Feb	6-Mar	13-Mar	20-Mar	27-Mar	3-Apr	10-Apr	17-Apr	24-Apr	1-May	8-May
Task 1 - Existing NAA Data						1	1	1	1	1	1	1	1	1	1	1	1	1
	Identify Peaks & Isotopes	2/12/2016	2/15/2016	All	-1													
	Develop Detector Efficiency Curve	2/8/2016	2/15/2016	Zach	-1													
	Extract Counting Rates	2/12/2016	2/15/2016	All	-1													
	Correct Monitor Data	2/15/2016	2/19/2016	All (with Keenan's help)	3													
	Analyze Samples	2/19/2016	2/26/2016	All	10													
	Check Data with Flux Analysis	2/19/2016	2/26/2016	All	10													
Task 2 - New Samples						7	7	7	7	7	7	7	7	7	7	7	7	7
	Contact MNRC for March Availability	2/12/2016	2/12/2016	Keenan	-4													
	Identify New Samples	2/12/2016	2/22/2016	Peter/Eduardo	6													
	Estimate Activation Products	2/22/2016	2/29/2016	Peter/Eduardo	13													
	Confirm with MNRC for samples and schedule	2/22/2016	2/29/2016	Keenan	13													
	Procure Samples	2/29/2016	3/7/2016	All	20													
	Prepare Samples	3/7/2016	3/11/2016	All	24													
	Perform Irradiation	3/18/2016	3/25/2016	MNRC	38													
	Receive Samples and start counting	3/28/2016	4/4/2016	Zach/Shreyas	48													
	Data Analysis of new Samples	4/4/2016	4/14/2016	All	58													
	Publish Data and Article to RadWatch website	4/28/2016	Shreyas	72														
Task 3 - NE 170 Project Deliverables						7	7	7	7	7	7	7	7	7	7	7	7	7
	Initial Project Proposal Draft		2/16/2016		0													
	Initial Project Proposal		2/23/2016		7													
	Progress Report and Presentation		3/15/2016		28													
	Draft Final Report		4/28/2016		72													
	Oral Presentation		4/29/2016		73													
	Final Report		5/12/2016		86													

Figure 1: This figure shows a Gantt chart with the expected timeline to finish the various tasks for this project.

5 Critical Equipment

We will be using the following instruments/tools:

- High Purity Germanium (HPGe) Detector in 1110C
- PeakEasy 4.81
- MNRC reactor
- High precision scale
- Plastic vials for sample preparation

5.1 Detector Efficiency Curve

Figure 3 shows the detector efficiency curve for the HPGe detector used in room 1110C. The sources used to generate the efficiency curve were ^{137}Cs , ^{60}Co , ^{152}Eu , ^{22}Na , and ^{228}Th . We used a plethora of sources in order to cover a wide range of gamma ray energies and find the efficiency over the entire energy spectrum we will be measuring. Using this efficiency curve, we will be able to determine the initial elemental concentrations of our samples.

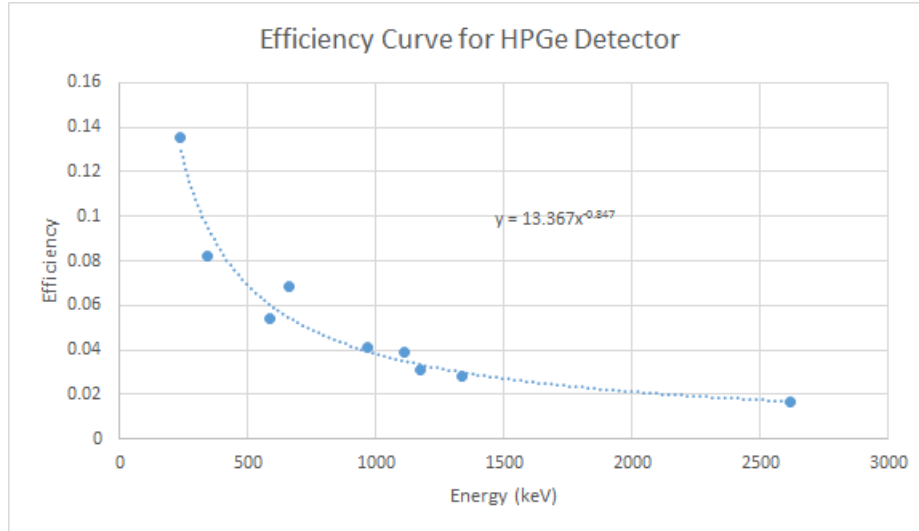


Figure 2: This figure shows the dectector efficiency curve for the HPGe detector in room 1110C.

6 Irradiation Procedure

6.1 Sample Preparation

Before preparing our samples, we decided to determine what new samples to activate. We decided to choose various samples of fish, seaweed, and seashells for analysis. The following list shows the elements expected to be seen in the different samples.

Seaweed:

- Potassium, Sodium, Calcium, Magnesium and Sulfur
- Trace amounts of Zinc, Copper, Chlorine, Cobalt, Manganese, Selenium, Bromine, Iron, and Arsenic

Seashells:

- Calcium, Carbon, Oxygen

Fish:

- Carbon, Oxygen, Hydrogen, Sulfur, and Nitrogen
- Trace amounts of various metals, such as Sodium, Aluminum, and Iron
- Looking for Mercury-203 at 279 keV

Preparation of the samples for irradiation required two individuals, both wearing latex gloves. One individual handled the weighing of the vial using the high precision scale before and after a sample was placed inside. The other individual used tongs to carefully place each sample into the vial, making sure to not contaminate the outside of the vial to prevent radioactive contamination from forming during irradiation. After all the samples were placed in vials, the vials were soldered shut to ensure that the vials would not open inside the reactor. The vials were then transported to MNRC where the irradiation occurred.

6.2 Long Run Procedure

For the long run, we used samples of fish, seaweed, and seashells in order to diversify our sample data and findings. Again, after receiving approval from MNRC to perform long run irradiation on our samples, we took the samples to MNRC. The samples were placed in the NTD position, which has a lower flux, but allows for irradiation at a power level of 1 MW for many hours, as opposed to the PTS. After waiting a few days for the samples to decay to safe levels, we received the samples in Etcheverry and counted them using the HPGe detector in the 1110C laboratory.

6.3 Short Run Procedure

For the short run, we used four samples from the initial set of kelp samples. After receiving clearance from MNRC to run these samples for a short-lived isotope test, we brought the samples to MNRC. We put our sample vials inside rabbit tubes, which eventually went in the pneumatic transfer system (PTS). The PTS is used for the short run, whereas the neutron transmutation doping position (NTD) is used for the long run. See Figure *insert figure number based on report* for the locations of the PTS and NTD. We ran the reactor at a power level of 20 kW, for a total of 100 seconds of irradiation. Immediately after the samples decayed to a level safe to handle, MNRC ran counts on the samples up to an hour in length.

7 Spectra Analysis

7.1 Kelp Long Run Spectra

Figure 2 shows a sample spectra that we have already analyzed. Our initial analysis found the characteristic gamma rays of ^{82}Br and ^{24}Na . This shows that bromine and sodium were most likely present in our original sample, and using the neutron flux, neutron capture cross section, counts in peak, and detector efficiency, we can determine the initial composition of bromine and sodium in our sample.

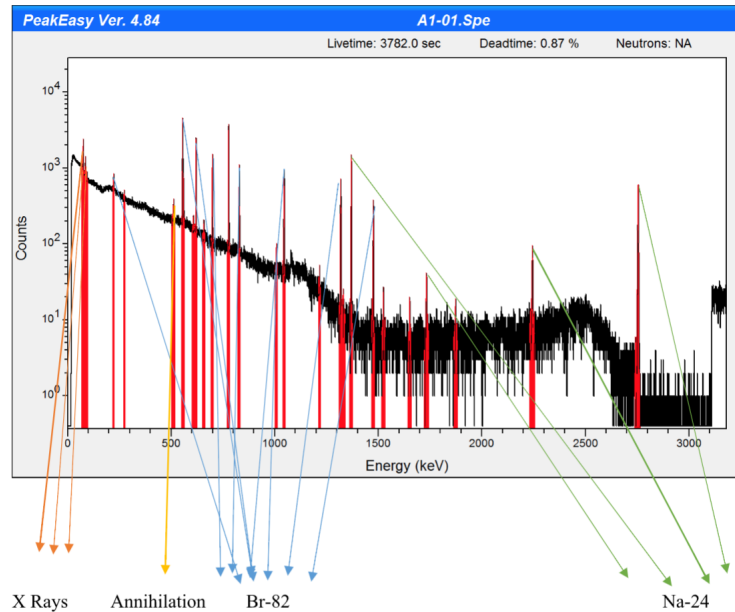


Figure 3: This figure shows a NAA spectra for sample 1. ^{82}Br and ^{24}Na are very prominent in the spectra.

7.2 Kelp Short Run Spectra

7.3 Fish Long Run Spectra

7.4 Seaweed Long Run Spectra

7.5 Seashell Long Run Spectra

8 Data Analysis and Results

8.1 Concentration Calculation

$$\frac{\text{element weight}}{\text{sample weight}} = \left(\frac{d}{s}\right) \frac{A}{N_0 W(I.A.)} \frac{t_{1/2}}{\ln 2} \frac{\exp\left(\frac{(t-t_{irrad}) \ln 2}{t_{1/2}}\right)}{\phi \sigma \Delta T_{irrad}} \quad (3)$$

Figure 4: This figure shows the orthodox equation used to determine the relative abundance a given element in an unknown sample.

The orthodox way to determine relative elemental composition in an unknown sample is given by the equation in figure 4. However, by using standard pottery, we are able to determine the elemental composition of an unknown sample without detector efficiency, irradiation flux, and cross section. Standard pottery is clay pottery which contains a wide range of chemical elements. Using NAA, we can activate the pottery and use it as a chemical fingerprint for many different elements. Many minor components of the pottery are activated via NAA and these component compositions are known to high accuracy, making the standard pottery a valuable tool in determining the composition of an unknown sample. Figure 5 shows the elemental composition of the standard pottery used as part of this project.

TABLE III					
COMPOSITION* OF STANDARD POTTERY					
Element	Species studied	Technique†	Diff. techniques	Composition Best value	Chemical symbol
Aluminium	²⁷ Al	neut act 1		(15.9 ± 0.2) × 10 ⁻²	Al
Antimony	¹²³ Sb	neut act 3	(1.66 ± 0.12) × 10 ⁻⁶		
	¹²⁴ Sb	neut act 4	(1.73 ± 0.06) × 10 ⁻⁶	(1.71 ± 0.05) × 10 ⁻⁶	Sb
Arsenic	⁷⁶ As	neut act 3		(3.08 ± 0.22) × 10 ⁻²	As
Barium	¹³⁰ Ba	neut act 2	(7.13 ± 0.32) × 10 ⁻⁴		
	¹³¹ Ba	neut act 4	(7.0 ± 1.1) × 10 ⁻⁴	(7.12 ± 0.32) × 10 ⁻⁴	Ba
Bromine	⁸¹ Br	neut act 3		(2.3 ± 0.9) × 10 ⁻⁶	Br
Calcium	⁴⁷ Ca	neut act 3	< 1 × 10 ⁻²		
		opt spec	< 1 × 10 ⁻³		
		wet chem	< 2 × 10 ⁻⁴		Ca
Carbon	CO ₂	C-H anal		(2 ± 3) × 10 ⁻⁴	C
Cerium	¹⁴⁰ Ce	neut act 4		(8.03 ± 0.39) × 10 ⁻⁵	Ce
Caesium	¹³⁵ Cs	neut act 4		(8.31 ± 0.55) × 10 ⁻⁵	Cs
Chlorine	³⁶ Cl	neut act 1		< 1.3 × 10 ⁻⁴	Cl
Chromium	⁵² Cr	neut act 4		(1.151 ± 0.038) × 10 ⁻⁴	Cr
Cobalt	⁶⁰ Co	neut act 4		(1.406 ± 0.015) × 10 ⁻³	Co
Copper	⁶⁴ Cu	neut act 2	(6.0 ± 0.8) × 10 ⁻⁵		
	⁶⁵ Cu	wet chem	(5.8 ± 0.5) × 10 ⁻⁵	(5.9 ± 0.5) × 10 ⁻⁵	Cu
Dysprosium	¹⁶³ Dy	neut act 2		(4.79 ± 0.19) × 10 ⁻⁶	Dy
Europium	^{152m2} Eu	neut act 2	(1.418 ± 0.048) × 10 ⁻⁶		
	¹⁵³ Eu	neut act 4	(1.477 ± 0.047) × 10 ⁻⁶	(1.448 ± 0.034) × 10 ⁻⁶	Eu
Gallium	⁷² Ga	neut act 2		(4.44 ± 0.46) × 10 ⁻⁵	Ga
Gold	¹⁹⁸ Au	neut act 3		< 1 × 10 ⁻³	Au
Hafnium	¹⁸¹ Hf	neut act 4		(6.23 ± 0.44) × 10 ⁻⁶	Hf
Hydrogen	H ₂ O	C-H anal		(5.4 ± 0.5) × 10 ⁻³	H
Iron	⁵⁹ Fe	neut act 4		(1.017 ± 0.012) × 10 ⁻²	Fe
Lanthanum	¹⁴⁰ La	neut act 3		(4.490 ± 0.045) × 10 ⁻⁵	La
Lutetium	¹⁷⁵ Lu	neut act 3		(4.02 ± 0.36) × 10 ⁻⁷	Lu
Magnesium	²⁵ Mg	neut act 1	(5 ± 2) × 10 ⁻³		
		opt spec	(7 ± 2) × 10 ⁻³	(5 ± 2) × 10 ⁻³	Mg
Manganese	⁵⁵ Mn	neut act 2		(4.09 ± 0.05) × 10 ⁻⁵	Mn
Nickel	⁵⁸ Ni	neut act 4		(2.79 ± 0.20) × 10 ⁻⁴	Ni
Potassium	⁴¹ K	neut act 2		(1.45 ± 0.04) × 10 ⁻²	K
Rubidium	⁸⁶ Rb	neut act 4		(7.00 ± 0.63) × 10 ⁻⁵	Rb
Samarium	¹⁵² Sm	neut act 3		(5.78 ± 0.12) × 10 ⁻⁶	Sm
Scandium	⁴⁶ Sc	neut act 4		(2.055 ± 0.033) × 10 ⁻⁵	Sc
Silicon dioxide	SiO ₂	wet chem		(6.04 ± 0.03) × 10 ⁻³	SiO ₂
Sodium	²³ Na	neut act 2		(2.61 ± 0.04) × 10 ⁻³	Na
Strontium	^{87m} Sr	neut act 2		(1.45 ± 0.22) × 10 ⁻⁴	Sr
Tantalum	¹⁸² Ta	neut act 4		(1.550 ± 0.044) × 10 ⁻⁶	Ta
Thorium	²³² Th	neut act 4		(1.396 ± 0.039) × 10 ⁻⁵	Th
Titanium	⁴⁸ Ti	neut act 3		(7.82 ± 0.34) × 10 ⁻³	Ti
Uranium	²³⁸ U	neut act 3		(4.82 ± 0.44) × 10 ⁻⁶	U
Ytterbium	¹⁷³ Yb	neut act 3		(2.80 ± 0.36) × 10 ⁻⁶	Yb
Zinc	⁶⁵ Zn	neut act 5		(1.26 ± 0.08) × 10 ⁻⁴	Zn

Figure 5: This figure shows the orthodox equation used to determine the relative abundance a given element in an unknown sample.

The counts per second per gram, or $\frac{cps}{g}$ for a given sample is given by

$$\frac{cps}{g} = \frac{N\Phi\sigma(1 - e^{-\lambda t})\epsilon\beta_\gamma}{m * LT}$$

where N is number of atoms, Φ is irradiation flux, σ is cross section, ϵ is detector efficiency, β_γ is branching ratio, LT is live time, and m is sample mass. We can write the activity A of a given sample as

$$A = N\Phi\sigma(1 - e^{-\lambda t})$$

, allowing us to rewrite $\frac{cps}{g}$ of a given sample as

$$\frac{cps}{g} = \frac{A\epsilon\beta_\gamma}{m * LT}$$

Using a simple ratio between level of sample activation and elemental concentration $\frac{A}{C}$, we can say that

$$\frac{\frac{cps}{g} \text{ sample} |_{t=0}}{C_{\text{sample}}} = \frac{\frac{cps}{g} \text{ pottery} |_{t=0}}{C_{\text{pottery}}}$$

Using this ratio, we can calculate the elemental concentration in a given sample with the standard pottery.

$$C_{\text{sample}} = \frac{\frac{cps}{g} \text{ sample} |_{t=0} C_{\text{pottery}}}{\frac{cps}{g} \text{ pottery} |_{t=0}}$$

The error associated ratio method for calculating sample concentration can be calculated using the error propagation formula

$$\sigma_F^2 = \sum_{i=1}^N \left(\frac{dF}{dx_i} \right)^2 \sigma_{x_i}^2$$

. Expanding this out for the formula given above, we get that the error in concentration can be given by

$$\sigma_{C_{\text{sample}}}^2 = \left(\frac{C_{\text{pottery}}}{\frac{cps}{g} \text{ pottery} |_{t=0}} \right)^2 \sigma_{\frac{cps}{g} \text{ sample}}^2 + \left(\frac{\frac{cps}{g} \text{ sample} C_{\text{pottery}}}{\left(\frac{cps}{g} \text{ pottery} \right)^2} \right)^2 |_{t=0} \sigma_{\frac{cps}{g} \text{ pottery}}^2 + \left(\frac{\frac{cps}{g} \text{ sample} |_{t=0}}{\frac{cps}{g} \text{ pottery} |_{t=0}} \right)^2 \sigma_{C_{\text{pottery}}}^2$$

8.2 Kelp Sample Long Run Results

Table 1: This table shows the elemental compositions of the initial kelp NAA and error.

Kelp ID	Sample Location	% ^{42}K	% ^{24}Na	% ^{82}Br	% ^{76}As
Sitka-1	Sitka, AK	7.30 ± 0.30	4.70 ± 0.08	0.036 ± 0.015	
RES-1	Tofino, BC				
BML-1	Van Damme State Park, CA	3.81 ± 0.46	3.69 ± 0.07	0.064 ± 0.025	
UCSCTER-1	Santa Cruz, CA				
CAM-1a	Cambria, San Luis Obispo County, CA				
REED-1c	Santa Barbara, CA				
Oxy-1	Rancho Palos Verdes, Los Angeles County, CA	6.77 ± 0.61	3.22 ± 0.63	0.08 ± 0.03	
LB-1	Long Beach, Los Angeles County, CA	Not Seen		0.11 ± 0.04	
OI-1	San Clemente, Orange County, CA	2.73 ± 0.13	2.45 ± 0.04		
PTL-1	Point Loma (San Diego), CA	6.49 ± 0.73	3.10 ± 0.06	0.061 ± 0.024	
UABC-1	Baja California, Mexico		3.43 ± 0.06	0.074 ± 0.029	
UCN-1	Chile				
LB-2B	Long Beach, Los Angeles County, CA Not Found	5.43 ± 0.24	4.10 ± 0.07	0.14 ± 0.06	
NS6					
NS1	Tomales Point, Marin County, CA		3.03 ± 0.05	0.031 ± 0.012	

8.3 Kelp Sample Short Run Results

Similar tables and bar graphs will be generated to show results for this section.

8.4 Fish Long Run Results

Similar tables and bar graphs will be generated to show results for this section.

8.5 Seaweed Long Run Results

Similar tables and bar graphs will be generated to show results for this section.

8.6 Seashell Long Run Results

Similar tables and bar graphs will be generated to show results for this section.

9 Conclusion

This section will include a wrap-up discussion of our results

10 Future Work

The important findings from these experiments will be posted on the RadWatch website to assist with their research into radioisotope activity in the oceans. Hopefully, these findings will give some insight into what affects the concentrations of isotopes in ocean going samples, which will allow later teams to devise plans on how to best predict and combat buildup of hazardous materials along the coast.

Future work that can be done for this project would be to survey other areas that are highly populated and near large harbors. Areas such as New York, Boston would be good locations to perform NAA on sea samples to compare elemental compositions across

11 Acknowledgements

We would like to thank Keenan Thomas and Professor Kai Vetter for helping us determine the scope of this project, and for the assistance they give us during the course of completing this project. Furthermore, we would like to thank Wesley Frey and the rest of the MNRC staff for assisting us with the irradiation. Finally, we would like to thank Max Fratoni for his guidance and assistance during the NE170 course and the completion of this project.