

Development and deployment of an underway radioactive cesium monitor off the Japanese coast near Fukushima Dai'ichi

Caffrey, J.A.*, Higley, K.A., Farsoni, A.T., Smith, S., Menn, S.

*Corresponding Author

Abstract

A custom radiation monitoring system was developed by Oregon State University at the request of the Woods Hole Oceanographic Institute to measure radioactive cesium contaminants in the ocean waters near Fukushima Dai'ichi Nuclear Power Plant. The system was to be used on board the R/V Ka'imikai-O-Kanaloa during a 15 d research cruise to provide real-time approximations of radionuclide concentration and alert researchers to the possible occurrence of highly elevated radionuclide concentrations. A NaI(Tl) scintillation detector was coupled to a custom-built compact digital spectroscopy system and suspended within a sealed tank of continuously flowing seawater. A series of counts were acquired within an energy region corresponding to the main photopeak of ^{137}Cs . The system was calibrated using known quantities of radioactive ^{134}Cs and ^{137}Cs in a ratio equating to that present at the reactors' ocean outlet. The response between net count rate and concentration of ^{137}Cs was then used to generate temporal and geographic plots of ^{137}Cs concentration throughout the research cruise in Japanese coastal waters. The concentration of ^{137}Cs was low but detectable, reaching a peak of 3.8 ± 0.2 Bq/L.

1. Introduction

On March 11, 2011 the Tōhoku Earthquake and resulting tsunami devastated much of the eastern Japanese coastline. The resulting damage and power loss at the Fukushima Dai'ichi Nuclear Power Plant (FDNPP) led to meltdown in three of the six reactors on site (IAEA, 2011). In the massive effort to cool the leaking reactors, a substantial inventory of fission products was released to the environment in the form of contaminated cooling water. Due to the plant's immediate proximity to the Pacific Ocean, much of this inventory was drained directly to the sea. Additional radioactive releases to atmosphere (Bowyer et al, 2011) and the surrounding land occurred due to venting and hydrogen explosions in the reactor containment buildings (IAEA, 2011), providing an additional source of radioactivity to the Pacific Ocean.

Radionuclide concentrations in the waters immediately surrounding Fukushima Dai'ichi were sampled regularly by the Tokyo Electric Power Company (TEPCO) and, later, the Ministry of Education, Culture, Sports, Science and Technology-Japan (MEXT). Concentrations of radioactive ^{137}Cs and ^{134}Cs in seawater effluent at the plant's outlet reached maximum levels in early April and gradually declined thereafter (TEPCO, 2011). Dilution in seawater dramatically decreased radionuclide concentrations at locations further from the power plant and later sampling yielded levels not exceeding detection limits in most locations offshore from FDNPP (MEXT, 2011).

The Woods Hole Oceanographic Institute assembled an oceanographic research team, sponsored by the Gordon and Betty Moore Foundation and the National Science Foundation, in an effort to characterize the migration and concentration of radioactive contaminants in Japanese coastal waters. As part of this effort the Department of Nuclear Engineering and Radiation Health Physics at Oregon State University (OSU) was requested to construct a system capable of analyzing the concentration of radioactive cesium while underway in the Japanese seawaters.

In response to this request, OSU developed a flow-through monitoring tank to house a NaI(Tl) scintillation detector. A portable high voltage power supply, digital multichannel analyzer (MCA), and spectroscopy interface were developed in-house for use in this effort. A graduate student was enlisted to operate the system during the research cruise and to perform health physics surveys to minimize exposure to cruise participants.

Due to the uncertain status of the reactors at the time that this project began, it was imperative to create a system capable of alerting researchers to any sudden changes in radionuclide concentrations that might compromise the safety of the crew. For this reason, a custom spectroscopy interface was employed to monitor the count rate within a targeted region of interest. An audible alarm would alert researchers and crew to the presence of elevated levels of radioactivity in the seawater.

2. Materials and Method

2.1.1. Flow-through Chamber

The flow-through monitor cavity was constructed using readily available PVC materials. The lower portion of the chamber consisted of a large 8" pipe bonded to a solid PVC plate while a large reducing coupler created a top. The sodium iodide detector was placed in a smaller 2.5" diameter sealed PVC tube that had been bonded within the small end of the reducing coupler to provide a sealed housing. Small inlet and outlet pipes on either side of the cavity allowed seawater to flow in through the bottom and out near the top. In this manner, the gamma detector would stay dry while immersed in a constant volume of continuously flowing seawater. (Fig. 1)

2.1.2. Detector

A 2" x 2" NaI(Tl) scintillation detector (802-335, Canberra Industries, Inc. Meriden, CT USA) was selected for gamma counting due to its high efficiency and ready availability. A 3" x 3" NaI(Tl) was preferred, but delays in shipment and the inherent time constraints of the research cruise precluded its use in this design.

2.1.3. Electronics

A user-programmable digital multi-channel analyzer (Avicenna RX-1200 Digital MCA, Avicenna Instruments, LLC. Corvallis, OR USA) was programmed in-house to perform pulse processing and send spectral information to a PC through its USB connection. The small size and light weight of the device were ideally suited for travel or shipment, especially in comparison to NIM-based MCA systems (Fig. 2).

A high voltage supply was also required to provide bias voltage to the detector, but size and weight constraints again prevented the use of a NIM-based system. A portable high voltage power supply was therefore constructed in-house, with the internally adjustable voltage set at 1400 volts to match the ideal operating bias for the detector (Fig. 2).

2.1.4. Software

Construction and analysis of the pulse height spectra required a PC and software designed to integrate with the MCA system. Avicenna's RayPanel graphical user interface is a software package included with purchase of the RX1200 Digital MCA. It is built in the Visual Basic language and links directly to the MCA through a USB connection. Pulses are processed through the MCA and transmitted to the interface along with energy information so that a spectral plot of counts as a function of energy is constructed on-screen. To analyze the activity of a given isotope a specific region of interest (ROI) may be selected. An ROI rate history panel was added to the interface in preparation for the research cruise, allowing the user to monitor and plot the count rate within the specified region of interest over the duration of the count. This modification also included the audible alarm which could be adjusted to activate at a threshold count rate of the user's choice (Fig. 3).

2.2. Initial Calibration

A rough calibration of the detector was performed prior to the research cruise. A bucket and PVC detector enclosure were used for this purpose to prevent contamination of the flow-through monitor prior to field-use. A ^{137}Cs source was placed near the detector to determine energy calibration and region of interest for analysis of seawater. A series of spectra were then taken while sequentially increasing the concentration of ^{137}Cs from 1 Bq/L to 50 Bq/L in 7 L of fresh water. It was known at the time that this calibration would not be sufficient for accurate analysis of radiocesium concentrations, and that a post-expedition calibration would therefore be required. Regardless, this initial calibration provided an opportunity to test the system before collecting data in the field.

2.3. On Board System Preparation

The detection system and computer were secured in the wet laboratory once on board the R/V Kaimikai-O-Kanaloa (KOK) in the port of Yokohama. A nearby sink provided a tap of seawater pulled from 1-2 m below the water surface at the bow of the ship. A hose connected the sink outlet to the lower fitting of the flow-through monitor. A second hose extended from the upper outlet into the sink drain. The seawater tap was fully opened and measured a steady flow of 4.8 L/min. (Fig. 4)

Seawater in the port of Yokohama was assumed to be essentially uncontaminated by radioactive cesium. A long-duration background count was taken using this water with the region of interest around the ^{137}Cs peak. A ^{137}Cs check source was also used to confirm that the 662 keV peak did not move outside of the calibrated region of interest due to temperature effects on the NaI(Tl) system. Measured ambient dose rates of approximately 0.02 $\mu\text{Sv/h}$ suggested that the background reading was not substantially affected by surrounding radioactivity. The presence of ^{208}Tl and ^{214}Bi in seawater with respective gamma energies of 583.2 keV and 609.3 keV likely contributed counts to the ^{137}Cs region of interest, in addition to scattered photons resulting from the abundant ^{40}K (Povinec et al, 1996). It was assumed that the concentration of these nuclides remained constant between the point of background measurement and the point of sampling, so that such contributions are not included in the calculated net count rate.

2.4. Data Collection

Data collection began on the morning of June 5, 2011 as the R/V KOK steamed towards the first sampling station, being the furthest from the Fukushima Dai'ichi site. Spectra were sampled as water flowed continuously, so that all spectra were in effect a composite average of concentration between the beginning and end of the counting period. No activity was noted in the first two measurements of 10 h counting times. Measurements for 2 h counting times were performed through the following days in an attempt to build a series of spectra relative to the ship's position, while measurements of 10 h counting times were taken overnight. This method was later forfeited as the delays between data collection were undesirable. Long counting times of 10, 20, and 24 h were taken as the ship began to encounter increased concentrations of activity. Region of interest counts were then recorded manually in shorter intervals within each long counting period. A clear spectrum was thus

collected based upon a long term average while the manually recorded region of interest counts provided incremental measurement results. These could later be segmented and grouped into appropriate counting times to balance between counting error and temporal resolution.

As the counting time was extended in higher activity regions, it became apparent that the region of interest was accumulating additional counts beyond the calibrated 662 keV peak of ^{137}Cs due to the presence of ^{134}Cs (with peaks at 605 keV and 796 keV) in seawater. It was then known that the system was overestimating the concentration of ^{137}Cs and would require a recalibration that included ^{134}Cs upon return to Oregon State University.

2.5. Post-cruise Calibration

To account for the discrepancy in the OSU monitor, a post-analysis calibration was performed at Oregon State University, this time using the same flow-through monitor and both ^{134}Cs and ^{137}Cs isotopes. ^{134}Cs was created in the OSU TRIGA reactor through activation of stable cesium. The activity of ^{134}Cs was confirmed using a calibrated HPGe detector. Given that a 1:1 activity ratio of ^{137}Cs to ^{134}Cs was measured in seawater in the days following the Fukushima meltdown and radioactive release (Buesseler et al, 2011), a solution of the two cesium isotopes in a 1:1 activity ratio was therefore used for the calibration process. Activity in the 662 keV region of interest was measured at concentrations of between 0.1 Bq/L up to 7 Bq/L with respect to ^{137}Cs . The effect of peak interference was then accounted for and a relationship between region of interest count rate and ^{137}Cs concentration was more accurately correlated.

Limits of detection were calculated using methods described in Strom and Stansbury (1992), in which samples are counted for times varying from that of the background measurement. Limits fluctuated with each sample as count times were modified, but a typical minimum count time of 1800 s yielded a critical limit of $1.7\text{E}-2$ cps (95% confidence), equivalent to a concentration of 0.27 Bq/L (Nir-El, 2001). All measurements are presented herein including those with values of less than the critical level, as suggested in Potter and Strzelczyk (2008).

3. Results and discussion

The first several days of sampling yielded no detectable concentration of radioactive cesium. The sampling performed on June 8 presented the first hints of increased activity with the corrected calibration reporting a maximum concentration of 0.8 ± 0.2 Bq/L (Fig. 5). Due to the rough calibration performed prior to deployment, the presented on-board results were known to be inaccurate. It was a confirmation of the ability of the system, however, to detect relatively low levels of radioactivity. This spike in readings occurred on a day of somewhat foul weather with winds blowing from the west. Ambient dose rates also increased slightly during this period (~ 0.02 $\mu\text{Sv/h}$ increased to ~ 0.06 $\mu\text{Sv/h}$). Rain washout of radon daughters

is known to increase ambient dose rates in terrestrial settings (Mercier et al, 2009), but at this time it is unclear to what extent these radionuclides, namely ^{214}Bi , may have contributed to the measured fluctuations of radioactivity throughout the cruise.

Concentrations returned to lower levels in the following days, but a defined increase of concentration was noted as the R/V KOK moved further inland and entered shallower waters west of the Japan Trench. Pockets of further elevated activity were noted in some areas (Fig. 6). Somewhat surprisingly, the day of closest sampling to the Fukushima nuclear power plants (June 15) did not yield the highest measured concentrations of radioactive cesium. This despite the R/V KOK's 30 km proximity to Fukushima Dai'ichi and Fukushima Dai'ni, even as they were partially visible on the horizon.

The highest concentration of activity was in fact noted on June 16, one day following the closest sampling station as the ship traveled to a point south-east of the power plant. Peak concentrations reached a level of 3.8 ± 0.2 Bq/L, and on the announcement of the measured value a series of additional seawater samples were taken within the region. The R/V KOK made two passes over this region and therefore measured a change of concentration in multiple locations.

The flow-through monitor and detection system performed reliably throughout the cruise. The 2" x 2" NaI(Tl) detector provided sufficient detection efficiency to monitor the concentration of radioactive cesium at the levels present in the sampling region. Several logistical challenges arose during data collection, however, and these are addressed herein.

The typically low concentration of radiocesium, and therefore low activity, required counting periods of 2 h or more to achieve reasonable counting error. Temporal resolution of concentration (also relating to spatial resolution as the ship was underway) was therefore limited by this requirement. Resolution of concentration was improved in areas of higher activity, as the counting time could be reduced while maintaining reasonable counting error.

Negative net counting rates were occasionally measured at the lowest concentrations due to the accumulation of counts grossing less than the background count rate. This led to the reporting of unrealistic negative concentrations on a few occasions, though a concentration of zero fell within the margin of error for most of these negative measurements. These could possibly be attributed to a marginally elevated count rate during background measurement, perhaps from washout of radon daughters or other nuclides, but is more likely caused by statistical fluctuation and thermal effects within the NaI(Tl) detector (Moszyński et al, 2006).

The concentration of ^{134}Cs was not accounted for in the initial calibration of the monitor system (and was in fact not available for use at that time). Data provided by TEPCO regarding concentrations at the release point showed a nearly equal concentration of ^{134}Cs activity to that of ^{137}Cs . The decay of ^{134}Cs emits photons at energies of 604.7 and 795.8 keV. These interfere with the region of interest for ^{137}Cs about 661.6 keV, due to the poor resolution of NaI(Tl) in the case of the 604.7 keV, and due to the contribution of a Compton

continuum in the case of the 795.8 keV peak. It was known that the prior calibration was not suited for estimation of concentration, and on-board analysis showed that the OSU monitor overestimated ^{137}Cs concentration. A post-analysis calibration that included OSU-synthesized ^{134}Cs in equal activity concentration served to reproduce the experimental conditions and yielded an accurate measurement of ^{137}Cs concentration from the data collected on the cruise.

The RayPanel software, modified in-house at OSU, functioned reliably throughout the cruise. The inclusion of an audible high-rate alarm and ROI rate history offered some level of comfort to both the crew and to researchers who were in contact with the water and biological specimens so near the exclusion zone. Some additional features that were not considered prior to the cruise may be helpful in future deployments. The ability to save raw data files for later study could be used to determine the contributions from ^{134}Cs peaks strictly through spectral analysis, but the present calibration method performs just as well. The next deployment of the monitor would also benefit by including multiple regions of interest and by further automating the process to record region of interest measurements at regular intervals. Ideally, the system would perform a continuous count and save a series of spectra at regular intervals. The user could then analyze a long-interval spectrum for a given time period created by adding the series of short-interval spectra as desired.

4. Conclusions

The detection system performed reliably throughout the cruise and provided a great deal of pertinent data. The capability for this system to estimate radioactive cesium concentrations in real time assisted the crew and science personnel in determining the relative necessity for sampling in any given region. The inclusion of an audible alert to indicate elevated concentrations of radionuclides provided a measure of protection for personnel and could be explicitly used to search for regions of higher activity for the purpose of sampling in future deployments.

The short time frame for development of this system provided some difficulty, but the resulting product met or exceeded the expectations of the research team. Beyond providing the only continuous measurement of radionuclide concentration during this research expedition, the system performed remarkably as a general proof of concept. The digital spectroscopy hardware is unparalleled in its convenience and reliability. Overall costs for construction of the device were also very modest.

The difficulty of balancing counting error against temporal resolution resulted in some logistical problems, but these were mitigated and overcome as the expedition progressed toward the Fukushima nuclear power plants. Future deployments of the system will include some improvements to the detector to improve efficiency and to the spectroscopy interface to improve data collection.

The maximum ^{137}Cs concentration of 3.8 ± 0.2 Bq/L was noted at a location 138 km southeast of Fukushima Dai'ichi. The mean and median concentrations of ^{137}Cs throughout the research cruise were 0.78 and 0.47 Bq/L, respectively. Lower concentrations were generally noted at distances furthest from the source, especially to the east of the Japan Trench where sea depth increased dramatically. Pockets of elevated activity exceeded the concentrations noted at the point of closest sampling, however, indicating the significance of ocean currents in the distribution of contamination.

Acknowledgements

The Fukushima research cruise and related work was funded through sponsorship from the Gordon and Betty Moore Foundation and the National Science Foundation. Special thanks are extended the Woods Hole Oceanographic Institute for inclusion of Oregon State University in this project and to Dr. Ken Buesseler for his impressive management of the R/V Ka'imikai-O-Kanaloa research cruise.

References

Bowyer, T.W., et al., 2011. Elevated radioxenon detected remotely following the Fukushima nuclear accident. *Journal of Environmental Radioactivity* 102 681-687.

Buesseler, K., Aoyama, M., Fukasawa, M., 2011. Impacts of the Fukushima nuclear power plant on marine radioactivity. *Environ. Sci. Technol.*, Web Published 20 Oct, 2011. Accessed 27 Oct, 2011.

Moszyński, M., et al., 2006. Temperature dependences of LaBr₃(Ce), LaCl₃(Ce) and NaI(Tl) scintillators. *Nuclear Instruments and Methods in Physics Research A*, 568(2) 739-751.

Nir-El, Y., 2001. Minimum detectable activity in gamma-ray spectrometry – statistical properties and limits of applicability. *Health Physics* 80(suppl.1) S22-S25.

Potter, W.E., Strzelczyk, J., 2008. Measurements should not be recorded as less than MDA: an extension. *Health Physics* 94(suppl. 2) S62-S63.

Povinec, P.P., Osvath, I., Baxter, M.S., 1996. Underwater Gamma-spectrometry with HPGe and NaI(Tl) detectors. *Applied Radiation Isotopes* 47(9) 1127-1133.

Strom, D.J., Stansbury, P.S., 1992. Minimum detectable activity when background is counted longer than the sample. *Health Physics* 63(3) 360-361.

IAEA, 2011. Mission report: IAEA international fact finding expert mission of the Fukushima Dai-ichi NPP accident following the Great East Japan earthquake and tsunami. May 24 – June 2, 2011. Accessed Oct 6, 2011.

http://www-pub.iaea.org/MTCD/Meetings/PDFplus/2011/cn200/documentation/cn200_Final-Fukushima-Mission_Report.pdf

MEXT, 2011. Readings of Sea Area Monitoring at Post Out of Fukushima Dai-ichi NPP. May 1, 2011. Press release. Ministry of Education Culture, Sports, Science and Technology – Japan (MEXT). Accessed August 30, 2011.

http://radioactivity.mext.go.jp/en/monitoring_around_FukushimaNPP_sea_sea_area/2011/05/index.html

TEPCO, 2011. Detection of Radioactive Materials from Seawater near Fukushima Daiichi Nuclear Power Station (31st release). April 23, 2011. Press release. Tokyo Electric Power Company (TEPCO). Accessed August 30, 2011. <http://www.tepco.co.jp/en/press/corp-com/release/11042309-e.html>

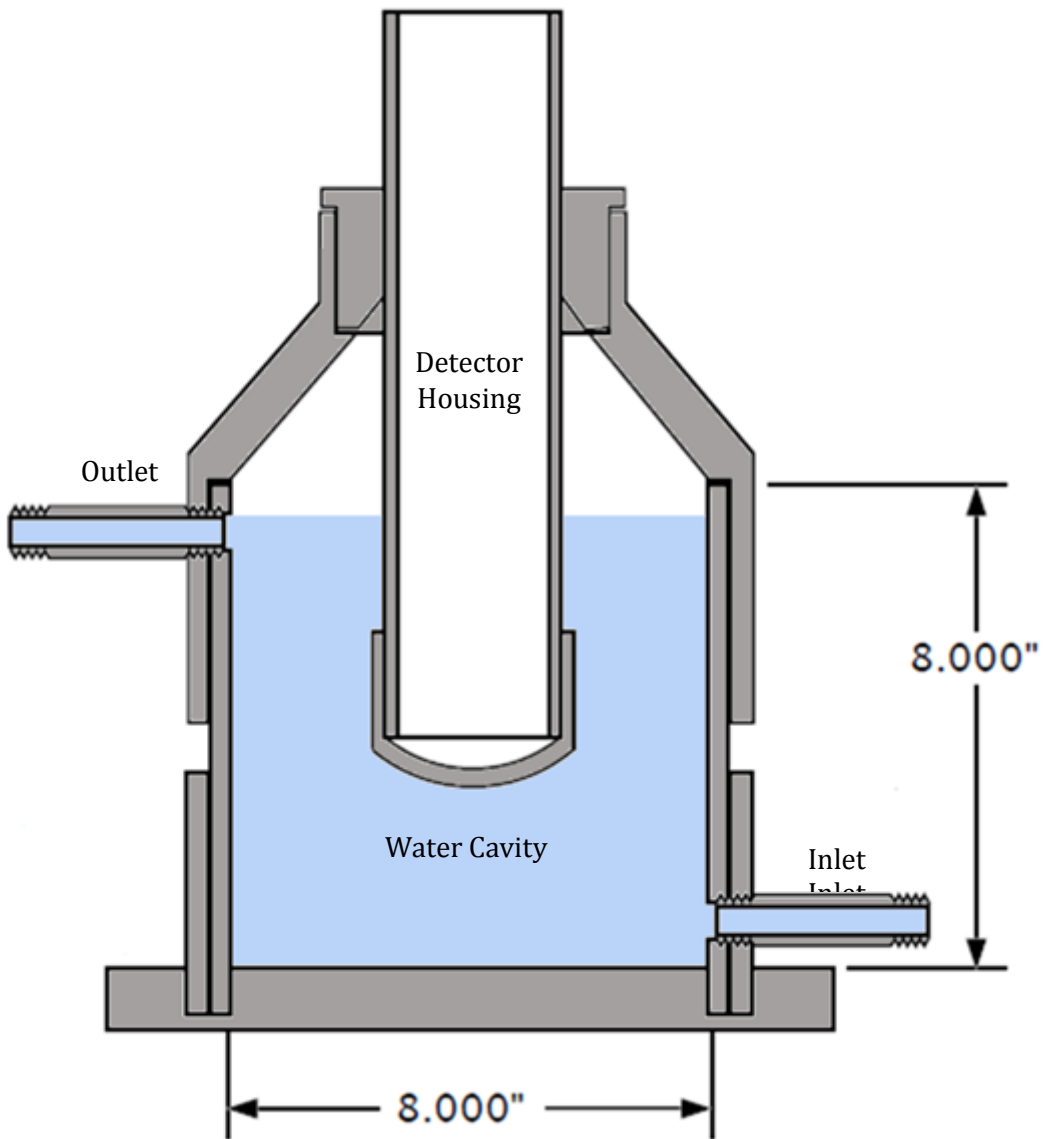


Figure 1: Cutaway of flow-through monitor



Figure 2: Avicenna RX1200 Digital Multichannel Analyzer (left) and high voltage power supply (right)

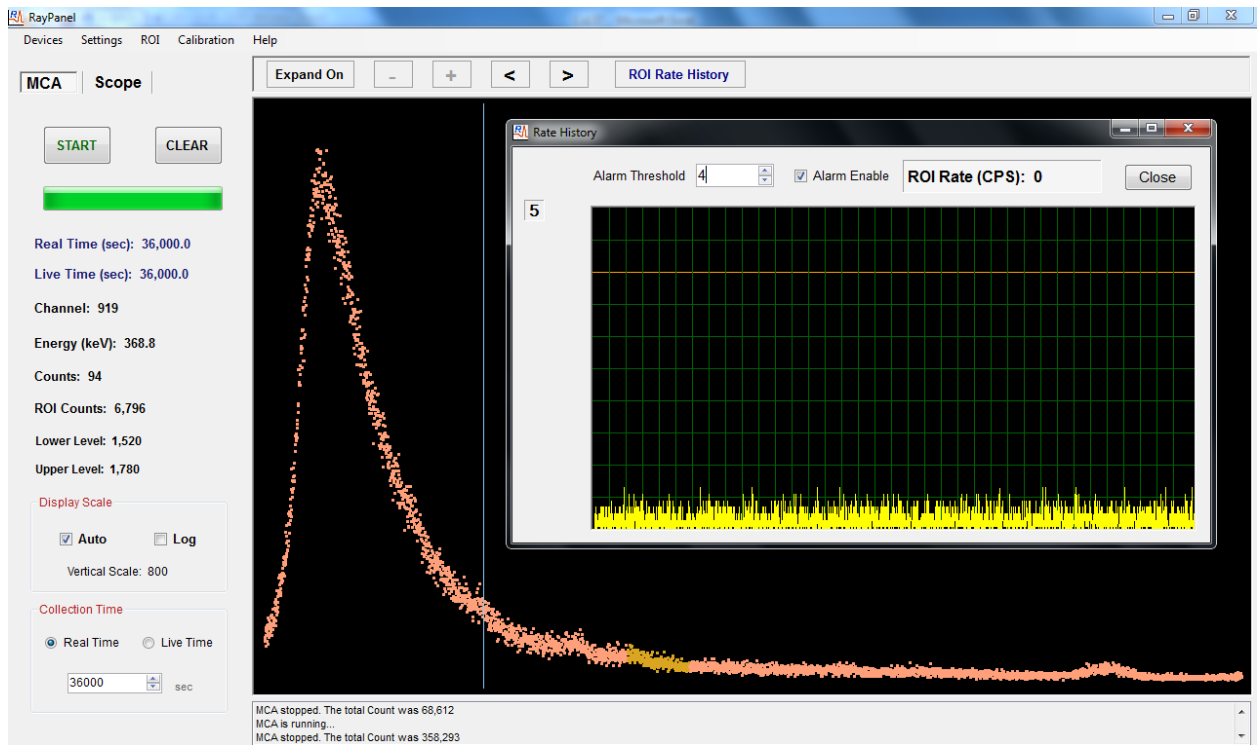


Figure 3: Screenshot of OSU-modified RayPanel multichannel analyzer interface

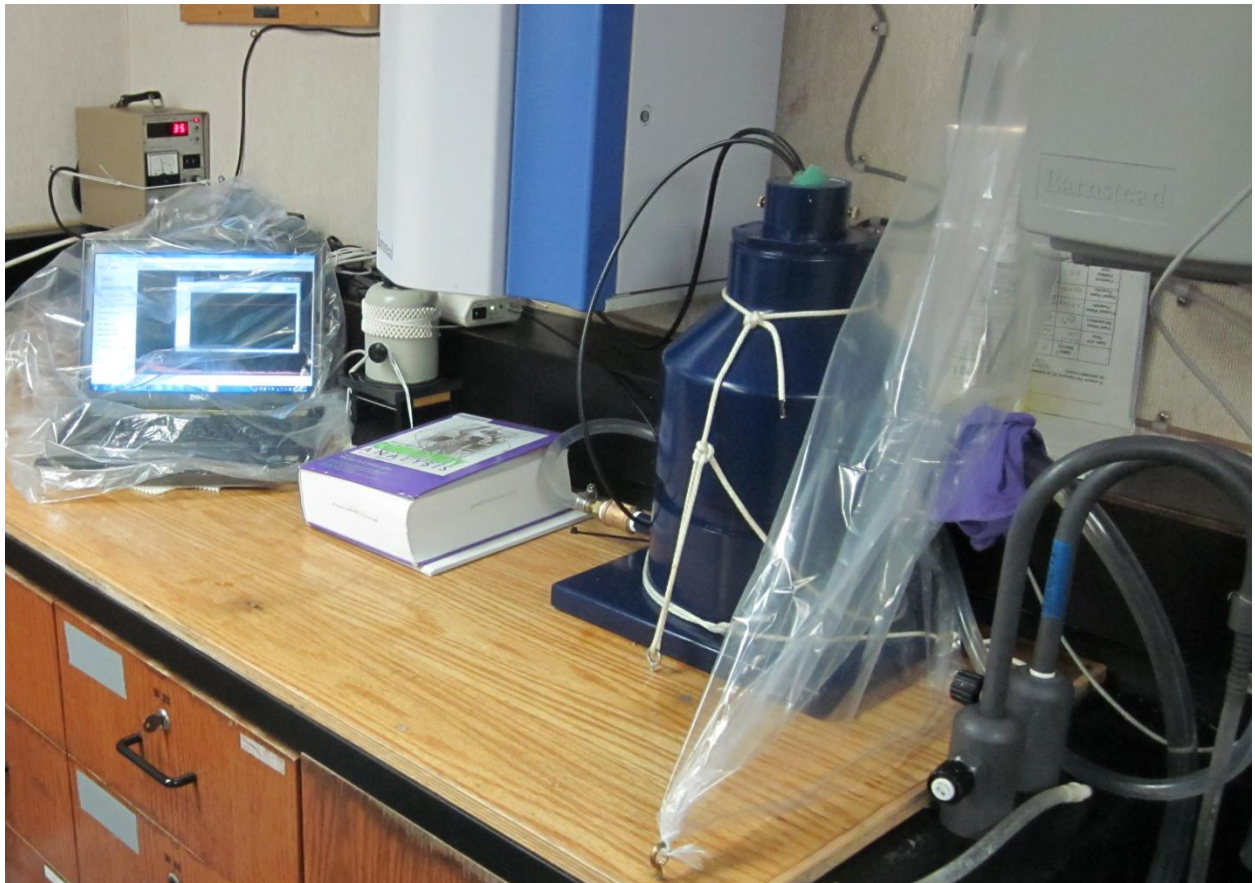


Figure 4: Lab setup on board the R/V KOK. Shown: seawater tap (right), flow-through monitor with rigging (center), laptop computer used for data processing (left). Also shown: shielded GM and scalar used for surface contamination surveys (middle and far left).

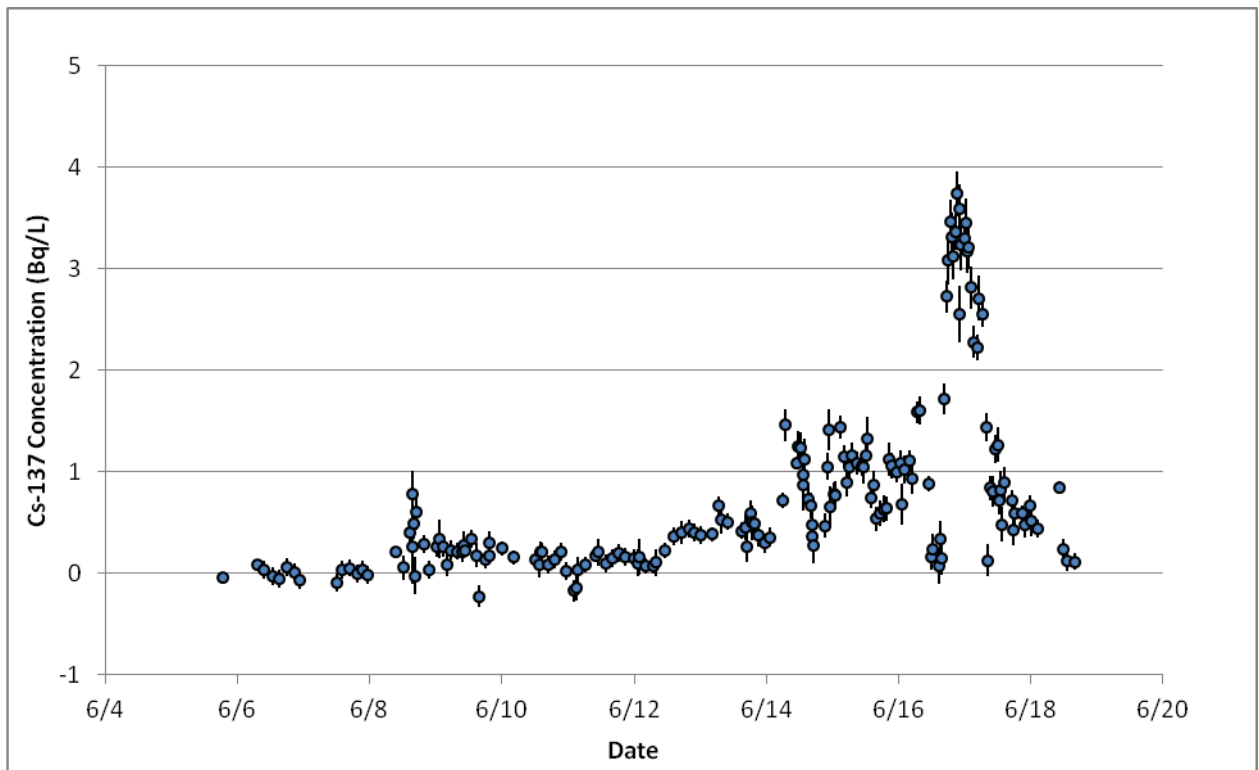


Figure 5: Measured concentration of ^{137}Cs based upon corrected calibration, shown with associated counting error.

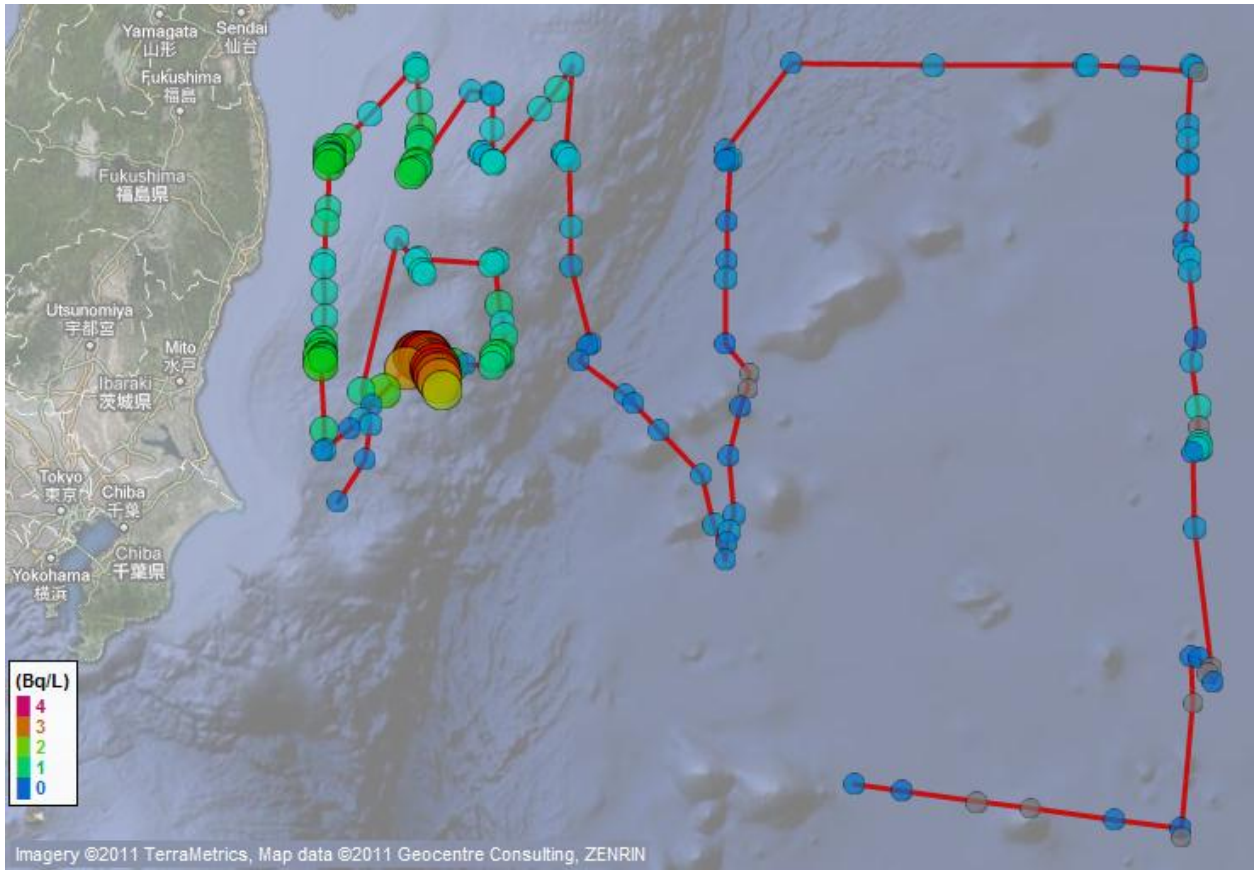


Figure 6: Geographic distribution of ^{137}Cs concentration¹

¹ Map created from web utility at <http://www.GPSVisualizer.com> based upon Google Maps® (2011 Google)