DRAFT protocol for collecting and testing filtered air samples

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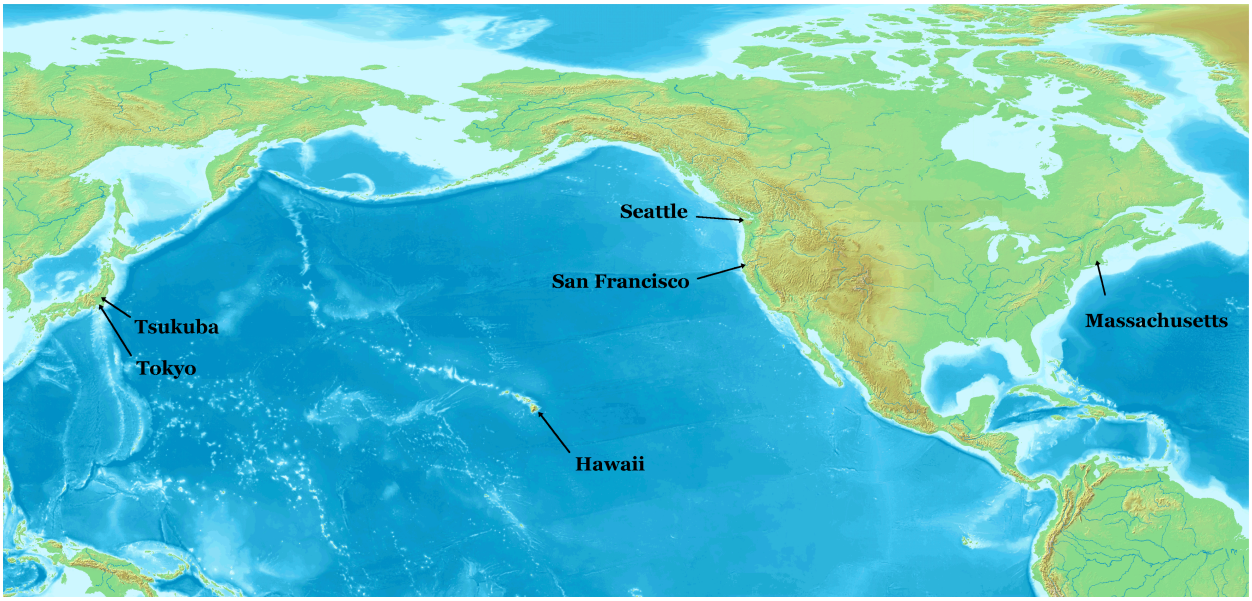
The Fukushima Daiichi nuclear complex released radioactive materials starting on March 11, 2011 in Fukushima, Japan. Much of this radiation is associated with fine particulate matter that can travel long distances by air. In partial fulfillment of the requirements for a dissertation research proposal, samples of environmental airborne fine particulate matter are collected using medium volume air pumps and preassembled filter cartridges. Sampling stations are set up in Tsubuka, Ibaraki Prefecture and Tokyo in Japan, Seattle, WA, San Francisco, CA, and Massachusetts, with a pending sampling station in Hawaii. Filter cartridges with drawn air volumes on the order of 60 cubic meters are shipped and analyzed after a 24 hour holding period. This holding period reduces the effects of natural radon in air. Analysis proceeds by beta and alpha counting, gamma spectrometry, autoradiography, and SEM/EDS analysis of individual particles. Of these analyses, only gamma spectrometry is performed on the WPI campus, primarily upon sealed air filter cartridges. The sampling program as proposed produces a minimum volume of solid wastes, (less than 1 liter total), all of which originates from environmental air samples. The advisor for this project is Prof. John Bergendahl, in the Department of Civil & Environmental Engineering. The dissertation committee also includes, in addition to Prof. Bergendahl, Prof. Paul Mathesin, Department of Civil & Environmental Engineering, and Dean Richard Sisson.

Outline of this procedure

Sampling Objective
Procedure: air sampling station & filter set up
Procedure: alpha and beta counting
Procedure: gamma spectrometry
Sample transport to off-campus locations
Procedure: Autoradiography
Procedure: SEM/EDS analysis
Sample custody
Chain of custody for BCD samples
Chain of custody for WPI samples
Sample tracking
NRC Regulations

Sampling objective

The goal of this project is to support or disprove the hypothesis that particles related to the Fukushima radiation releases can be identified at least two physically distant locations, (in Japan and in the United States, see figure 1), and that the environmental fate and transport of these particles can be related to their physical characteristics.



Above: Figure 1, location of air sampling stations operating as of April 20, 2011.

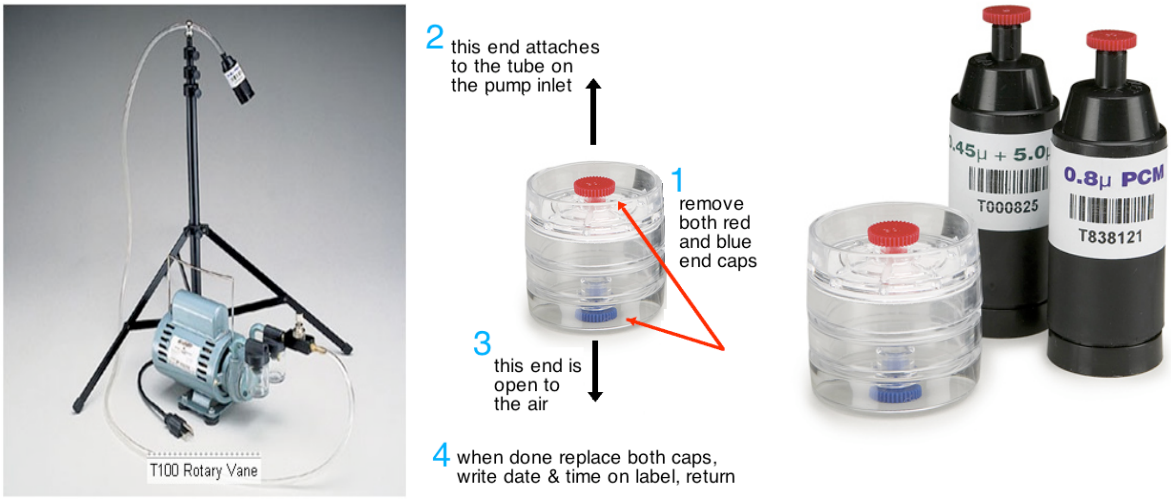
Procedure: air sampling station & filter set up

An Allegro sampling pump drawing 10 to 25 LPM or equivalent is used to draw air sample through sealed polycarbonate filter assemblies. (See figure 2 next page) The sampling period will be as close as is practical to 48 hours per filter. Given that quantities of analytes captured are anticipated to be very low for US sites, flow rates for US stations can be maximized for a given pump set up. Start and end time, date, and flow rates will be recorded in the sampling note book, and transferred to the sample tracking sheet and the chain of custody. The assemblies contain a support pad and a 0.45 micron pore size membrane filter.

The samples will be collected at different rates in order to reduce the possibility of having to store, transport, handle, or dispose of more radioactive materials that exceed the exempt quantity. (See NRC Regulations section, on the amounts of radioactive material that can be

handled without triggering the need for additional permitting.) The two Gillian/MSA-type pumps sent to Japan collect from 1 to 3 LPM, while those used in the USA, where lower concentrations are anticipated, collect 15 to 800 LPM using Allegro rotary vane pumps and a commercial HEPA vacuum from Lab Safety Supply, Inc. The higher rate for these presumed lower exposure-stations provides the requisite lower limits of detection.

Figure 2: L to R, Sampling set-up, sealed air sampling cartridges, 37 mm and 25 mm cartridges



Analyses on campus will be by gamma spectrometry, using the Dept. of Physics NaI and GeLi detectors. While on campus, the filter cartridges will remain sealed, and the integrity of the seal will be ensured with evidence tape. At this point assemblies may also be selected for autoradiographic based on gamma spectrometry results. This work will be done off campus. Autoradiography is the last step to select samples for SEM/EDS analysis, which is also done off campus, at a licensed commercial facility. All samples will originally be shipped to Boston Chemical Data Corp., (BCD), 2 Summer Street, Suite 14, Natick, MA 01760.

Procedure: alpha and beta counting

While still at BCD, samples will be screened for total alpha and beta counts on a DPM basis, after a 24 hour holding period to reduce the influence of local radon accumulations. The assemblies are analyzed off-campus at BCD for total alpha and beta counts using a Ludlum Model 3030 two channel counter for 60 minute counts. The assembly must be opened for these

analyses. No other preparation of the sample is required. Once received, all sample cassettes must be handled using disposal gloves to prevent contamination. After recording the count results, the sample filters are immediately returned to their respective cassettes. The count results are recorded in a log book, in the sample tracking form, and, using a marker, on the cassette housing.

Procedure: gamma spectrometry

After resealing the assemblies, the filters will be transported to WPI and analyzed at WPI's Dept. of Physics via gamma spectrometry. The samples should be counted for 24 hours each on either the NaI or GeLi spectrometer in room 26 at Olin Hall. The key must be acquired from the departmental office for each use of the spectrometers. Room 26 is to be kept locked at all times unless it is occupied. The entire assembly will fit into the spectrometer well once the sample liner is removed. Each user will be trained and certified by the primary investigator prior to working with the equipment. Analysis proceeds by use of the MAESTRO software package. Total counts, total live time, and an electronic copy of the .chn data file will be retained for every analysis on a floppy disk. Disks are also available from the departmental office. After analysis, samples will be returned to the water quality laboratory isotope storage area in Kaven Hall.

Sample transport to off-campus locations

Samples to be analyzed further will be removed to a commercial off-campus laboratory, which will arrange for proper disposal of completed samples. If samples are analyzed nondestructively, then remaining samples are to be returned to Kaven Hall at WPI for storage, pending final disposal via a certified and prequalified hauler. The chain of custody should be updated to reflect each transport of samples onto and off of the WPI campus. The sample tracking file (see figure 5) should be updated with the current location of the sample. All custody documents are to be maintained by the document custodian, (Marco Kaltofen, Kaltofen@wpi.edu).

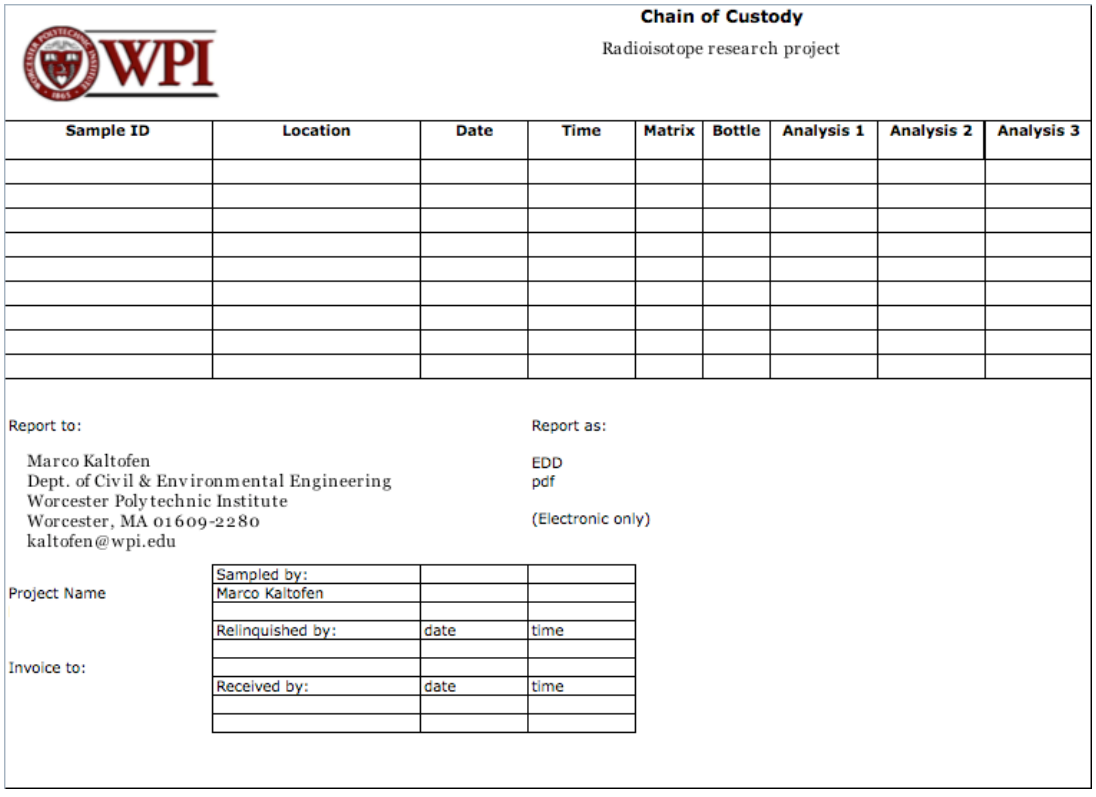
Procedure: autoradiography/SEM-EDS

Autoradiographs provide important information on the number, intensity, and exact filter pad location of radioactively-hot particles. The filter pads which have positive gamma spectrometry

or alpha/beta count results are mounted on a dark acrylic sheet using double stick adhesive tape. In a commercial darkroom, a sheet of Xray sensitive film is sandwiched with the mounted filters, and exposed for approximately 72 hrs. After exposure in the dark, the films are developed using the D76 process. The mounted filter assembly is transported to Microvision Laboratories of N. Billerica, MA. Areas of filters which cause exposures to the Xray film are removed from the mounts in a fume hood, and analyzed by SEM/EDS at Microvision. After SEM-EDS analysis, samples will be returned to WPI for storage and proper disposal.

Sample custody

A chain of custody form will be filled out for each sample or set of samples collected. A blank form is shown below. The information in the chain of custody is used to complete the help complete the sample tracking file. This form is to be used for samples collected, or received from other researchers, such as the filters received from private air sampling stations in Japan and San Francisco, CA. Below: Figure 3 - Chain of custody form, WPI.



Chain of Custody

Radioisotope research project

Sample ID	Location	Date	Time	Matrix	Bottle	Analysis 1	Analysis 2	Analysis 3

Report to:

Marco Kaltofen
Dept. of Civil & Environmental Engineering
Worcester Polytechnic Institute
Worcester, MA 01609-2280
kaltofen@wpi.edu

Report as:

EDD
pdf

(Electronic only)

Project Name

Invoice to:

Sampled by:		
Marco Kaltofen		
Relinquished by:	date	time
Received by:	date	time

Sample tracking

Sample tracking is done on a Microsoft Excel spreadsheet. Currently the two researchers generating original samples, (Marco Kaltofen, kaltofen@wpi.edu and Suzanne LePage, slepage@wpi.edu), maintain their own files. These files will be available to the group via a sharepoint group at:
https://student.sharepoint.wpi.edu/research/radioisotope/default.aspx

Below: Figure 4 – Example sample tracking form

A	B	C	D	E	F	G	H	I	J	K	L	M	N	O	P	Q	R
ID	current location	sample custodian	State of Origin	start date	start time	end date	end time	total min.	LPM	sample size	filter size mm	alpha dpm	beta dpm	NaI counts / 1200 sec.	location sampled	total NaI cts	total NaI time
FKD008W rainwater worcester, ma	Kaven WQL	MK	MA	4/12/11	NA	NA	NA	NA	NA	1 Kg	NA				Natick, MA		
			CA														
SEA010B - moss sample	Olin 26	MK	WA							3 g	NA	1	61				
			HI														
NATICK017F	Kaven WQL	MK	MA	4/6/11							37	1	29	9701		4885930	604376
NATICK018F		MK	MA	4/8/11													
NATICK019F		MK	MA	4/11/11													
NATICK020F		MK	MA														
NATICK021F		MK	MA														

To facilitate sample storage and disposal, minimum sample volumes will be collected so that only the quantity actually needed for analysis is on hand. Analytical work is nondestructive, so arrangements for sample disposal are necessary. One function of the sample tracking spreadsheet is to identify samples which are hazardous or which contain hazardous constituents. Any samples loaned from other researchers will be logged in using this form, until the time of their final return. (It is anticipated that air filters from a private research project in Japan will be handled in this fashion.) This form will also be used to ensure that the project remains well below the exempt isotope quantities. (See sec. NRC Regulations)

NRC Regulations

§ 30.18, (a), (e) Exempt quantities.

(a) Except as provided in paragraphs (c) through (e) of this section, any person is exempt from the requirements for a license set forth in section 81 of the Act and from the regulations in parts 30 through 34, 36, and 39 of this chapter to the extent that such person receives, possesses, uses, transfers, owns, or acquires byproduct material in individual quantities, each of which does not exceed the applicable quantity set forth in § 30.71, Schedule B.

(e) No person may, for purposes of producing an increased radiation level, combine quantities of byproduct material covered by this exemption so that the aggregate quantity exceeds the limits set forth in § 30.71, Schedule B, except for byproduct material combined within a device placed in use before May 3, 1999, or as otherwise permitted by the regulations in this part.

This NRC exemption schedule is at URL:

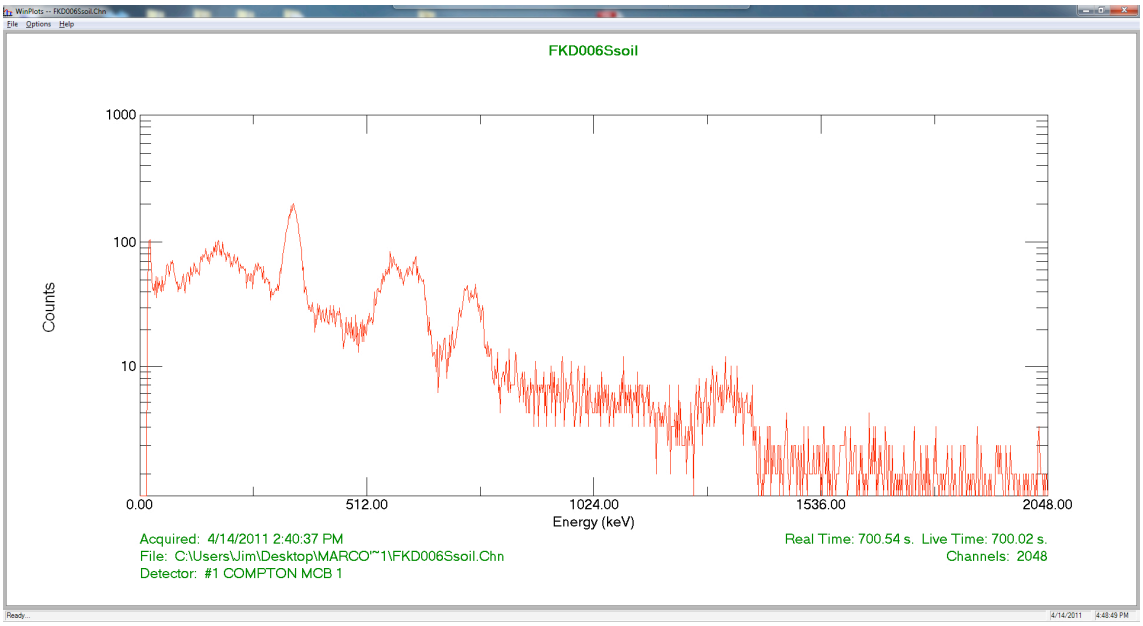
http://www.nrc.gov/reading-rm/doc-collections/cfr/part030/part030-0071.html

The isotopes under study, and the respective schedule B, sec. 30.71 exempt quantity limits are radioiodine, (131I - 1,000 nCi), and radiocesium, (137Cs - 10,000 nCi, 134Cs - 1,000 nCi).

This project will result in the collection of air filtering media. The media are 37 mm in diameter (maximum, some are 25 mm in diameter) and 0.5 mm in thickness. If the project results in the collection of 200 samples, then the total sample volume should be about the same as a deck of playing cards. These membranes will contain whatever particles were filtered from the approximately 60 cubic meters of air drawn through each one.

A limited number of standard materials will need to be analyzed to standardize the gamma spectrometers. It is proposed that a 137Cs source will be loaned to the project. This source would have to be on campus for less than one day, and would be under the direct supervision of the primary investigator while on campus. A set of samples from areas in Japan outside of the danger zone will also be required. These samples will not exceed the limits above, and will be stored off campus. These samples are required to provide source term information for environmental fate and transport investigations of airborne particulate matter. (See figure 6, gamma spectrum of Tsubuka, Ibaraki Prefecture surface soil, collected April 10, 2011.)

Figure 6: NaI gamma spectrum of soil sample, Tsukuba, Japan, showing target isotopes 131I, 134Cs, and 137Cs related to Fukushima nuclear fallout.



References

Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA, J. Diaz Leon,1 D. A. Jaffe,2 J. Kaspar,1 A. Knecht,1 , ∗ M. L. Miller,1 R. G. H. Robertson,1 and A. G. Schubert1

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