National Atmospheric Release Advisory Center Dispersion Modeling During the Fukushima Daiichi Nuclear Power Plant Accident

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Abstract

The U.S. Department of Energy / National Nuclear Security Administration (DOE / NNSA) deployed personnel to Japan and stood up expert teams to aid in assessing the consequences of releases from the Fukushima Daiichi Nuclear Power Plant. The National Atmospheric Release Advisory Center (NARAC) was activated as the DOE/NNSA's operational plume modeling capability. NARAC provides real-time atmospheric dispersion predictions of air concentrations and ground contamination as well as dose resulting from a radiological incident. This paper briefly summarizes NARAC response activities during the Fukushima emergency and then discusses NARAC source reconstruction efforts. A range of source estimates were found to be consistent with the available data, with estimates varying depending on assumptions about the release rates (e.g., time-varying vs. constant-rate), the radionuclide mix, the meteorology, and/or the radiological data used in the analysis. However, NARAC results were consistent within expected uncertainties and were found to agree with other studies that used different models, source estimation methodologies, and radiological measurement data sets. Results from a preliminary model sensitivity study of the dependence of calculated thyroid dose on iodine partitioning between gas and particulate phases also are presented in this paper.

Keywords: Fukushima Daiichi; reactor accident; atmospheric dispersion modeling; meteorological modeling; source estimation; dose exposures; environmental monitoring

1. Introduction

Following the 2011 Tohoku earthquake and tsunami, the U.S. Department of Energy / National Nuclear Security Administration (DOE/NNSA) deployed personnel to Japan and activated expert teams across the DOE laboratory complex to aid in assessing the consequences of releases from the Fukushima Daiichi Nuclear Power Plant[1)]. DOE/NNSA personnel provided predictive modeling, air and ground monitoring (including the deployment of the Aerial Measuring System to Japan), sample collection, *in situ* field and laboratory sample analysis, dose assessment, and data interpretation. The National Atmospheric Release Advisory Center (NARAC) at Lawrence Livermore National Laboratory (LLNL) was activated by the DOE/NNSA on March 11, 2011. The center remained on active operations through late May when the DOE/NNSA ended its deployment to Japan. Over 32 NARAC staff members, supplemented by other LLNL scientists, invested over 5000 person-hours of time and generated over 300 analyses during the response.

NARAC simultaneously supported a number of Fukushima-related modeling activities in response to a variety of requests for meteorological and dispersion analyses including:

- Daily weather forecasts and hypothetical atmospheric dispersion predictions to provide on-going situational awareness of meteorological conditions and to inform planning for U.S. field data collection and operations

- Estimates of possible dose in Japan resulting from hypothetical scenarios developed by the U.S. Nuclear Regulatory Commission (NRC) that were used to inform U.S. federal government considerations of possible actions that might be needed to protect U.S. citizens in Japan
- Predictions of potential plume arrival times and dose for U.S. locations
- Plume model refinement and source estimation based on meteorological analyses, atmospheric dispersion modeling, and available field data

An overview of NARAC response activities including a description of the first three activities listed above is available in Sugiyama et al., 2012[2)] and will not be replicated here. This paper discusses NARAC source estimation and provides some additional material on potential dose exposures. The paper concludes with results from a preliminary investigation of changes in predicted thyroid dose resulting from different assumptions regarding iodine partitioning between gas and particle phases.

2. Source Estimation

As part of standard response procedures during a U.S. radiological release, NARAC provides preliminary model predictions to guide initial measurement surveys. In turn, field teams conduct air and ground monitoring and collect samples for laboratory analysis. These data are uploaded into DOE databases for quality assurance by the U.S. DOE/NNSA Consequence Management Home Team (CMHT) and transferred electronically to NARAC. NARAC uses specialized software to select, filter, and both graphically and statistically compare measurements and model predictions. Modeling analyses are then refined based on the available data and the new predictions are provided to the field teams. This iterative process continues until the impacts of the release are characterized. Although this standard procedure was altered during the Fukushima response due to the prioritization of other modeling requests, as well as the unique aspects of the DOE/NNSA response to the Fukushima Dai-ichi accident, NARAC conducted an initial set of source reconstruction estimates that are discussed in this paper.

NARAC analysts reconstructed source estimates by optimizing the overall graphical and statistical agreement of model predictions with dose-rate measurements by comparing data and model values paired in space and time. Source reconstruction for the Fukushima accident was complicated by the long duration of the releases, emissions from multiple reactor units, unknown reactor and spent fuel pool conditions, rapidly-changing meteorological conditions, complicated geography and land-sea interfaces, and the relatively limited measurement data available during critical stages of the releases.

2-1 Meteorological Conditions

Rapidly changing atmospheric conditions presented a significant modeling challenge during the Fukushima response. NARAC meteorological analyses were developed from observational data provided by the Japan government and/or numerical weather predictions generated using the U.S. community Weather Research and Forecast (WRF) model[3)] driven by NOAA Global Forecast System (GFS) model output[4)]. The WRF model was used in both pure forecast mode and in four dimensional data assimilation (FDDA) simulations that incorporated Japanese meteorological observations. The latter simulations used analysis nudging[5)] for the outer model domains (27, 9, and 3 km grid spacing) and observational nudging[6)] for the innermost domain (1 km grid spacing).

NARAC simulations showed that following the earthquake and tsunami, winds remained primarily off-shore until the March 14 – March 16 UTC period, during which the wind direction rotated in a clockwise direction consistent with the movement of a low pressure area. NARAC-simulated wind directions pushed modeled plumes southwards from March 14 13:00 UTC to March 15 02:00 UTC period, rotated towards the northwest around March 15 03:00 UTC, and turned off-shore again on March 16 UTC. Winds then remained off-shore until March 21 UTC when changing conditions again sent radioactive material southward in the general direction of Tokyo.

Precipitation occurred episodically throughout the release period. NARAC investigated a variety of precipitation conditions ranging from uniform grid-wide, time-varying precipitation based on Japanese meteorological observations to WRF FDDA spatially and temporally varying precipitation[2)]. Comparisons of measured and WRF FDDA modeled rates showed good qualitative agreement with precipitation data for the passage of a rain band in the March 15th UTC time frame, as did time-series comparisons of measured and predicted precipitation rates for stations located near Tokyo and Fukushima City.

Initial NARAC forecasts captured the overall pattern of winds and the occurrence of precipitation, but subsequent higher resolution (3-km) WRF FDDA simulations provided increased accuracy in modeling the timing of the wind shifts and precipitation patterns[2)] and were used in the source estimation process. Wet deposition from both in-cloud and below-cloud precipitation scavenging significantly impacted NARAC estimates of downwind plume transport and deposition.

2-2 Radiological Data

The primary data available to NARAC for source estimation during the Fukushima response consisted of dose-rate measurements. The Japanese Ministry of Education, Culture, Sports, Science and Technology[7)] (MEXT) provided data from its radiological monitoring stations, although most data from the prefectures closest to the Daiichi nuclear power plant were available only after March 15 0900 UTC. MEXT also collected dust sample data, but insufficient data of this type were available to NARAC for use during the response. The U.S. DOE Aerial Measuring Survey (AMS) arrived and began taking data on March 17-18 and U.S. personnel collected ground monitoring data as well as samples for laboratory analysis[1)]. The center also received on-site Tokyo Electric Power Company (TEPCO) radiological measurements, although significant data gaps existed in the time period following the earthquake and tsunami and during the March 15 site evacuation. TEPCO measurements were used as qualitative guidance only, as these data were collected from locations very close to the site that were likely to have been heavily influenced by the local wind conditions and the exact location of the mobile monitor relative to the release locations. To supplement the very limited information available regarding reactor and spent fuel conditions, NARAC also drew upon U.S. Nuclear Regulatory Committee (NRC) analyses of possible nuclear reactor scenarios.

2-3 Source Reconstruction Methodology

NARAC’s source estimation efforts concentrated on the critical period from March 14-16 UTC, due to DOE/NNSA interest in the relatively high deposition pattern measured by the Aerial Measuring System (AMS) to the northwest of the Fukushima Daiichi nuclear power plant. The various NARAC source estimates were based on the data and other information that were available at the time the analysis was conducted. As additional or corrected data were received, these estimates often were updated to take into account the new information. As discussed above, NARAC meteorology was derived from both weather observations and WRF FDDA simulations. Releases from all reactor units were treated as one combined source.

One of the key assumptions in NARAC's source estimation process was the selection of an appropriate radionuclide mix. Initial NARAC source estimates used a radionuclide mix of ^{133}Xe, ^{131}I, and ^{137}Cs that was provided by the DOE/NNSA CMHT based on an analysis of data provided by the USS Ronald Reagan from a location approximately 100 miles off the east coast of Japan. Later analyses incorporated three other primary radionuclide contributors to dose: ^{132}I, ^{132}Te (due to its decay into ^{132}I), and ^{134}Cs. Typical release activity ratios for ^{133}Xe:^{131}I: ^{132}I: ^{132}Te:^{137}Cs:^{134}Cs used in NARAC's source estimation process were 100:20:20:20:1:1 or 100:10:10:10:1:1. These relative activity ratios were determined *a priori* from data provided by DOE laboratory analyses, supplemented by U.S. NRC reactor scenario radionuclide mixes. For example, DOE *in situ* field assays, later confirmed by laboratory analyses of soil samples and air filters collected over the March to May, 2011 period [8)], showed that a reasonable choice for the ^{134}Cs:^{137}Cs activity ratio was 1:1, despite considerable scatter in the data.

NARAC source estimates were produced by statistically and graphically comparing data and model results paired in both space and time[9)]. Input assumptions were varied to find the best fit to the data and the average measured-to-predicted value ratio was used to scale the release amounts to best match the measurements. Below-threshold measured and/or predicted values were not used in the comparisons, and outlier values were removed as appropriate. The primary statistics used in the model-data comparisons were the percentage of predicted values that fell within a factor, R, of the measured values (where R = 2, 3, 5, 7, 10, 20, 50, 100, and 1000), supplemented by a bias analysis (i.e., consideration of the relative magnitude and number of values over or under predicted). Ratios of measured and computed values were used for statistical comparison of air concentration and ground deposition values that varied over many orders of magnitude. Additional statistical measures included the (absolute and signed) bias, the normalized mean square error, and the average and standard deviations of the ratios of measurements to calculated values.

3. Results

NARAC conducted a number of source reconstruction analyses using a range of possible release assumptions and meteorological conditions. Both uniform and time-varying release rates were examined and a limited investigation was made of the sensitivity to different radionuclide activity ratios, release heights, and particle-size distributions. In this paper, we will focus on the results of one source reconstruction analysis that will be designated the "baseline" estimate. Comparisons with other NARAC analyses that used different meteorology (e.g., observational data, WRF forecasts, WRF FDDA analyses), radionuclide mixes (i.e., relative activity ratios for iodine, cesium, tellurium, and xenon), and radionuclide measurement data (e.g., AMS, MEXT) are summarized in the results discussion below.

3-1 "Baseline" Case Source Estimate

The "baseline" case[2)] is a constant release rate fit to the data for the March 14-16 UTC, 2011 period derived by optimizing the overall graphical and statistical agreement of model predictions with 451 MEXT dose-rate measurements from 19 stations within Fukushima and the surrounding Prefectures. The MEXT dose-rate measurements were assumed to include both air immersion and ground-shine contributions. The "baseline" case used arguably the best meteorology developed by NARAC during the response – WRF FDDA simulations at 3 km resolution, which incorporated Japanese meteorological observations. The assumed radionuclide mix for ^{133}Xe: ^{131}I: ^{132}I: ^{132}Te: ^{137}Cs: ^{134}Cs was 100:20:20:20:1:1.

Graphical comparisons from the model-data fit for the "baseline" case are shown in Figure 1 for two time example time periods - 0900 UTC and 1200 UTC on March 15. The corresponding total release estimate over the March 14 0000 UTC to March 16 0000 UTC period was 3.7×10^{15} Bq (1×10^{5} Ci) each of ^{137}Cs and ^{134}Cs, 7.4×10^{16} Bq (2×10^{6} Ci) each of ^{131}I, ^{132}I, and ^{132}Te, and 3.7×10^{17} Bq (1×10^{7} Ci) of ^{133}Xe.

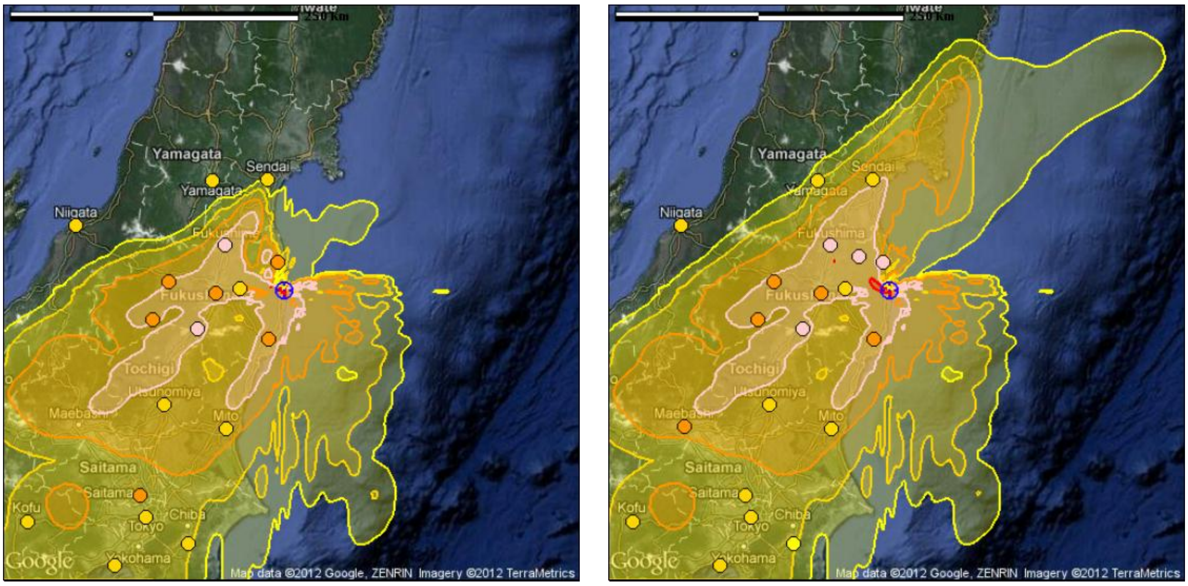

Figure 1. Dose rate results from the NARAC-modeled "baseline" case (color-filled contours) are compared with MEXT data (circles color coded to the same levels as the contours) for March 15 0900 UTC (left panel) and March 15 1200 UTC (right panel). The innermost red contour is the area where the model predicts that 120 µGy h^{-1} (12.0 mrad h^{-1}) is exceeded; pink shows 4-120 µGy h^{-1} (0.4-12.0 mrad h^{-1}), orange 0.4-4 µGy h^{-1} (0.04 – 0.4 mrad h^{-1}), light orange 0.04-0.4µGy h^{-1} (0.004-0.04 mrad h^{-1}), and yellow 0.004-0.04 µGy h^{-1} (0.0004-0.004 mrad h^{-1}). The blue circle indicates the location of the Fukushima Daiichi plant. (Background map courtesy of Google)

Over 35% of the predicted values were with a factor of 2 of the measurements, (i.e., the ratios of measured and predicted values for the same time and location were between 0.5 and 2) and 82% within a factor of 10. The agreement between predicted and measured values was slightly better for the MEXT stations located outside of Fukushima Prefecture (not shown). The NARAC "baseline" case model-predicted values also were found to fit the March 18th AMS data to a similar degree, even though these data were not used in developing the source estimate (see Figure 2).

The "baseline" case provides an interesting comparison to other NARAC analyses in which time-varying release rates were used to match the data. These results showed that the deposition pattern to the northwest of the Fukushima Daiichi nuclear power plant could be matched by different combinations of time-varying emission rates, spatially and temporally-varying precipitation, and precipitation scavenging parameters. It is unclear whether sufficient data is available to distinguish the competing contributions of these two effects, as similar deposition patterns can be derived by either increasing release rates or increasing wet deposition rates over the appropriate time periods. It also should be noted that NARAC analyses did not account for the changes in wet deposition resulting from different types of precipitation (e.g., rain, snow).

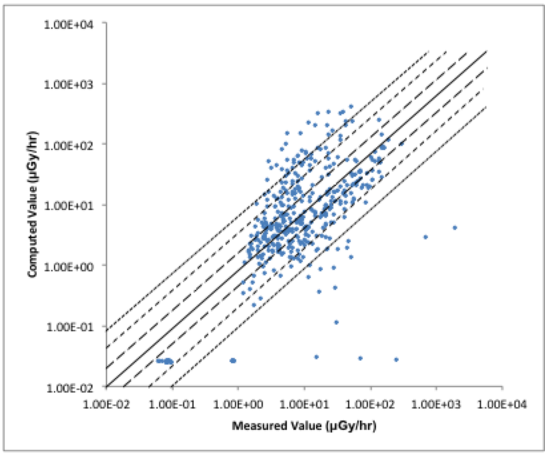

Figure 2. Scatter plot showing a comparison of the computed NARAC "baseline" case predicted values versus AMS data from March 18. The solid, long dash, short dash, and dotted lines delimit factors of 1, 2, 5, and 10 respectively.

3-2 "Baseline" Case Dose Estimates

The Total Effective Dose (TED) is the adult whole body dose resulting from inhalation, air immersion (during both initial plume passage and from resuspension), and ground-shine. Figure 3 shows both the predicted TED and the 50-year Committed Effective Dose (CED) from the "baseline" case. The CED is the adult combined internal dose from inhalation using a weighted sum of doses to various organs and is the internal dose component of the TED. The 50 mSv (5 rem) and 10 mSv (1 rem) levels shown in the plot as orange and yellow contours, respectively, are the early phase TED upper and lower U.S Protective Action Guide (PAG) dose levels for evacuation / sheltering. U.S. PAG levels are known to differ from those used in Japan.

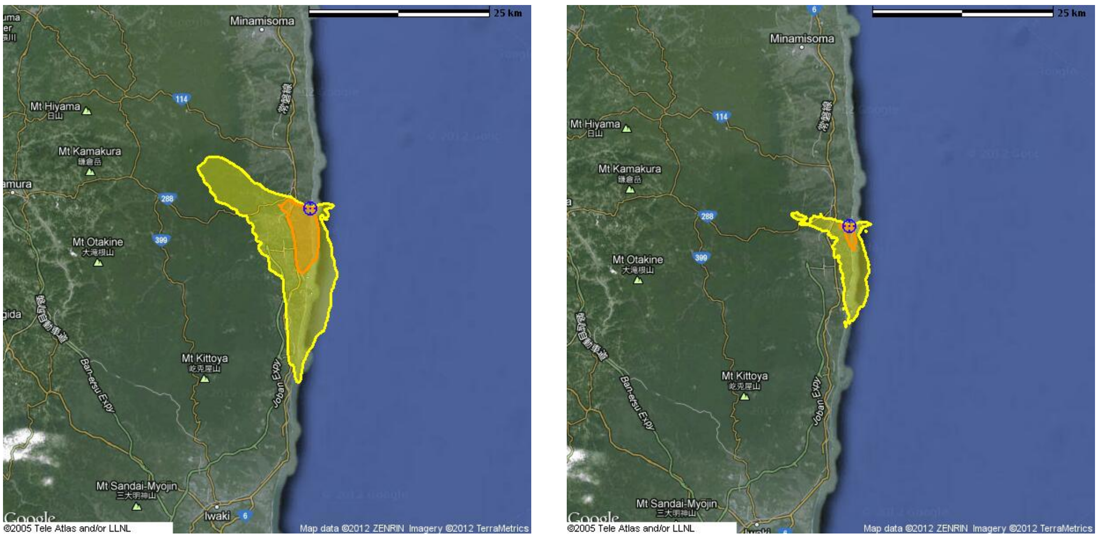

Figure 3. The Total Effective Dose (left panel) and the 50-year Committed Effective Dose (right panel) are shown for the "baseline" case. The contours delimit predicted areas exceeding 50 mSv / 5 rem (orange) and 10 mSv / 1 rem (yellow) for 4 days of exposure based on the "baseline" simulation for March 14-16 UTC. (Background map courtesy of Google)

3-3 Effects of Gas-Particle Partitioning on Thyroid Dose

The NARAC “baseline” case and other source term estimates conducted during the response modeled the emissions as respirable size particles, as little data were available on activity size distributions or the physical and chemical forms of the released material. Specifically, most NARAC analyses assumed a log-normal activity-size distribution with a median diameter of 1 μm, geometric standard deviation of 2, and minimum and maximum cut-offs at 0.1 μm and 10 μm respectively. However, U.S. Nuclear Regulatory Commission reactor analyses[10)] have shown that iodine can be produced as both non-reactive and reactive gases. In addition, LLNL laboratory analysis of combined paper and charcoal air filter samples at a few locations, including Yakota Air Base and the U.S. Embassy in Japan, are indicative of possible gas and particulate partitioning[8)]. Cesium was observed almost exclusively on the paper filters that are expected to collect most of the particulate material. In contrast, only 30% of the ^{131}I was found on the paper filter and 70% passed through to the charcoal filter that is designed to collect gaseous iodine.

NARAC therefore performed a preliminary sensitivity study on the effects of different assumptions about the chemical/physical form of iodine on calculations of thyroid dose. Four different gas-particle phase partitioning assumptions were simulated:

- 100% respirable particles (the “baseline” case)
- 100% non-reactive gas
- 100% reactive gas
- 25% particles, 30% reactive gas, 45% non-reactive gas

The last mixture is a default partitioning used in NRC RASCAL modeling of nuclear reactor scenarios[10)].

The same “baseline” case inputs described above were used in the thyroid dose simulations, apart from the use of different deposition and dose conversion factors. The former were derived from values found in the U.S. NRC’s RASCAL 4.0 model documentation[10)]. Gravitational settling was applied only to the particulate form. Reactive iodine was modeled using a dry deposition velocity value that was more than twice the value used for particulates (based on a typical RASCAL value for neutral stability and 3 m/s winds), while non-reactive gases were assumed to exhibit no dry deposition. Non-reactive iodine gas was modeled as not affected by wet deposition and the same conservative assumption was used for the reactive gas case. While the latter assumption may not be strictly true, wet deposition for reactive iodine gas is generally presumed to be much less than dry deposition, so this approximation may not have had a significant effect on the final results.

Thyroid dose was calculated from inhalation using dose conversion factors (DCFs) for 1-year old children and a breathing rate consistent with light physical activity levels. Dose conversion factors were derived from DCFPAK 1.8[11] (which in turn is based on ICRP Publications 56, 60, 66, 67, 69, 71, and 72). Specifically, NARAC used the DCFPAK I_2 “vapor” (V) and the CH_3I dose conversion factors for reactive and non-reactive gases, respectively. Gas-phase DCFs are approximately twice the particle DCF, with the reactive iodine DCF 20-30% higher than that for the non-reactive gas. Since the ^{132}I DCF for 1-year old child thyroid exposures is two orders of magnitude less than for ^{131}I, ^{132}I (including ^{132}I activity produced from ^{132}Te) played a minor role in the dose estimates.

Both non-reactive and reactive iodine gas simulations predicted a higher dose at any given distance than that resulting from particulate releases, or equivalently a greater downwind extent for any given dose level. This is apparent in Figure 4, which compares the results of the original “baseline” 100% respirable particle simulation with the calculation using a mixture of 25% particles, 30% reactive gas, and 45% non-reactive gas. Both reactive and non-reactive iodine gas thyroid dose estimates were predicted to be similar in extent. It should be noted that these simulation represent the first steps of a sensitivity study only. The source term was not re-estimated based on the differences in predicted dose rates. Additional data is needed in order to develop more accurate estimates of the iodine gas-particle phase partitioning and thyroid dose exposures resulting from the Fukushima Daiichi releases.

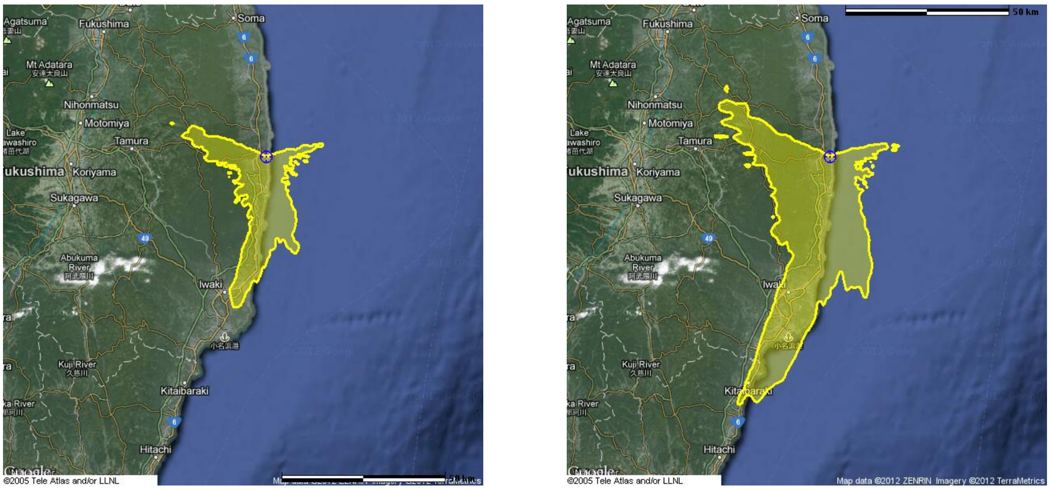

Figure 4. The two figures show the 70-year committed 1-year old child thyroid dose for iodine inhalation using the “baseline” source estimate over the March 14-16 UTC period for 100% particulate iodine (left panel) and a mixture of 25% particles, 30% reactive gas, and 45% non-reactive gas (right panel). The yellow contour is the 50 mSv / 5 rem level that corresponds to the early phase U.S. Protection Action Guide level for KI administration to children. (Background map courtesy of Google)

4. Discussion

NARAC source reconstruction analyses resulted in a range of estimates for emission rates that were consistent with the available dose-rate data, within model and measurement uncertainties. Total release estimates for the two-day period of interest (March 14-16 UTC, 2011) varied within approximately a factor of three of the “baseline” case for the same radionuclide mix[2)]. NARAC source estimates were found to be sensitive to a number of factors including:

- Source term assumptions (e.g., time-varying vs. constant emission rates, radionuclide mix and relative activity ratios, particle / activity size distributions, iodine gas / particle phase partitioning, height of release, reactor conditions)
- Meteorology (e.g., observational data, WRF analyses, WRF FDDA, or GFS global data)
- Model physics, including dry deposition, precipitation rates, type of precipitation (e.g., rain, snow), and precipitation scavenging parameters for both in-cloud and below cloud processes
- Selection of the radiological data to preferentially match in the source estimation process (e.g., MEXT data, AMS surveys)

Source term estimates are significantly more speculative during periods of off-shore wind flow, for which there is little to no regional radiological monitoring data.

NARAC source reconstruction estimates were also compared to other values published in the literature that were documented in sufficient detail that comparisons could be made for the same

March 14-16 UTC period (e.g., Chino et al. 2011[13]; GOJ 2011a[14], 2011b[15], and 2011c[16]; Stohl et al. 2011[17]). Despite the use of different radiological data (MEXT dose rate; MEXT dust data, Comprehensive Test Ban Treaty Organization [CTBTO] global monitoring data), meteorological models, source estimation methodologies, and assumptions regarding reactor conditions, these estimates agreed with a factor of approximately six[2].

4. Conclusion

The Fukushima Daiicihi accident generated a unique and voluminous data set, including both local and global radiological measurements from MEXT, TEPCO, CTBTO global monitoring data, U.S. DOE aerial and ground surveys, and U.S. EPA RadNet[12] monitors. To date, most atmospheric dispersion source estimation efforts have used only a fraction of these data. Model physics improvements are needed to more accurately simulate complex meteorological conditions and dispersion on both regional and global scales, including the use of data assimilation and ensemble techniques to develop probabilistic dose estimates. Atmospheric dispersion modeling could be significantly informed by incorporating on-going nuclear reactor modeling and analyses; the results of radiochemical and spectral analyses that provide insight into radionuclide mixes and gas-particle partitioning of iodine; and/or internal dose monitoring data. Integration of data from multiple sources may allow different release events to be distinguished and may better constrain possible release rates during off-shore flow periods. The combined use of modeling and monitoring data has the potential to fill in key gaps in source and exposure estimates. In addition, such efforts will lead to improved capabilities for responding to future events of a similar scale and complexity.

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