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Atmospheric Dispersion Modeling: Challenges of the Fukushima Dai-ichi Response

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ATMOSPHERIC DISPERSION MODELING:

CHALLENGES OF THE FUKUSHIMA DAI-ICHI RESPONSE

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ABSTRACT

This paper describes the support provided by the Department of Energy's (DOE) National Atmospheric Release Advisory Center (NARAC) during the Fukushima Dai-ichi nuclear power plant accident. As part of DOE's Consequence Management Home Team, NARAC produced a wide range of predictions and analyses including:

- Daily Japanese weather forecasts and atmospheric transport predictions to inform planning for field monitoring operations and to provide U.S. government agencies with on-going situational awareness of meteorological conditions
- Estimates of possible dose in Japan based on hypothetical U.S. Nuclear Regulatory Commission scenarios of potential radionuclide releases to support protective action planning for U.S. citizens
- Predictions of possible plume arrival times and possible dose levels at U.S. locations
- Source estimation and plume model refinement based on atmospheric dispersion modeling and available monitoring data

NARAC performed a number of source reconstruction analyses during the response by optimizing the overall agreement of model predictions to dose-rate measurements, using statistical comparisons of measurements and model values paired in space and time. NARAC-estimated emission rates varied by as much as a factor of three from a selected baseline case when different release assumptions (e.g., time-varying vs. constant rates), meteorology, and/or radiological data were used. The results of this and previous studies were found to be generally consistent within expected uncertainties, despite the

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application of different source estimation methodologies and significantly different radiological measurement data. The paper concludes with a discussion of some of the operational and scientific challenges encountered during the response and recommendations for future work.

Keywords: Fukushima-Dai-ichi, atmospheric dispersion modeling, radiological emergencies, reactor accidents, meteorological modeling, airborne radioactivity / atmospheric emissions, environmental monitoring

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INTRODUCTION

The National Atmospheric Release Advisory Center (NARAC) at Lawrence Livermore National Laboratory (LLNL) was activated by the Department of Energy / National Nuclear Security Agency (DOE/NNSA) Office on Emergency Response on March 11, 2011, to respond to events at the Fukushima Dai-ichi nuclear power plant. Although the reactors at the plant shut down automatically following the devastating Tohuku earthquake, the subsequent tsunami caused the loss of electrical power to the plant and damaged the backup generators. This in turn resulted in loss of cooling and heat build-up in the reactor cores and spent fuel pools leading to the release of radioactive materials into the atmosphere.

NARAC was asked to provide a wide range of simulations and analyses during the crisis including weather forecasts, dose calculations from hypothetical scenarios for emergency planning, predictions of arrival times and dose levels reaching U.S. territories, and source estimates based on the incorporation of field measurement data. By the time NARAC ended its active operations in late May, 32 members of its staff, supplemented by other LLNL scientists, had invested more than 5000 person-hours of time and produced more than 300 analyses and predictions.

Atmospheric plume modeling for Fukushima Dai-ichi posed an extremely complex problem due to the rapidly changing meteorological conditions (e.g., on and off-shore wind directions, precipitation events), Japan's complex topography, and the variety and number of reactor units experiencing problems over an extended time period. NARAC efforts were complicated by the difficulties in obtaining accurate information from Japan, particularly in the early stages of the response. During the first few days following the tsunami, only limited meteorological and radiological measurements were available. Much larger volumes of data were later received from Japanese weather and radiological monitoring stations, the DOE Aerial Measuring System (AMS), deployed U.S. and Japanese ground monitoring teams, and public Web sites and e-mail streams. However throughout the response, very little information was available regarding reactor and spent fuel pool conditions.

The remainder of this paper provides background on NARAC capabilities, presents some examples of the center's atmospheric dispersion analyses, and discusses some of the operational and scientific challenges encountered during the response.

NATIONAL ATMOSPHERIC RELEASE ADVISORY CENTER CAPABILITIES

The National Atmospheric Release Advisory Center (NARAC) provides tools and services to map the spread of hazardous materials accidently or intentionally released into the atmosphere (Nasstrom et al. 2007; Sugiyama et al. 2010). NARAC was created in 1979 during the Three Mile Island nuclear power plant accident. Since that time, the center has responded to dozens of nuclear emergencies, including the 1986 Chernobyl nuclear reactor disaster and the 1999 nuclear fuel accident in Tokaimura, Japan. The center also provides capabilities to model the impacts of radiological dispersal devices, nuclear detonations, nuclear weapons accidents, and other radiological, chemical, biological, and natural releases.

The center's products provide information on affected areas and populations, potential casualties, health effects and protective action guides, contaminated areas, and damage zones to assist decision makers and responders in taking actions to protect the public and the environment. NARAC is the atmospheric dispersion modeling center for DOE/NNSA emergency operations and one of the components of the Consequence Management Home Team (CMHT). The center supports other sponsors and missions and serves as the operations hub for the Department of Homeland Security (DHS)-led Interagency Modeling and Atmospheric Assessment Center (IMAAC), whose role is to coordinate plume modeling during events requiring federal coordination.

NARAC utilizes a distributed modeling system to predict the potential impacts of hazardous atmospheric releases. The system incorporates a suite of source term, meteorological, dispersion and dose-response models, databases of hazardous material properties, and graphical and statistical analysis tools. It contains extensive global geographical databases and obtains real-time global meteorological data from the National Oceanic and Atmospheric Administration (NOAA), the Department of Defense (DoD), regional networks, and other sources. Both a meteorological data assimilation model (ADAPT) and the Weather Research and Forecasting (WRF) model are used to develop analysis and forecast atmospheric fields. The center's dispersion model, LODI, solves the advection-diffusion equation using a Lagrangian stochastic Monte Carlo approach. Other specialized modeling capabilities are available to estimate nuclear prompt effects, blast damage, fallout, resuspension, urban impacts, and corrections to indoor exposures based on sheltering/shielding. During responses, the center acquires chemical, biological, and/or radiological monitoring data for use in refining model predictions.

Model outputs of air and ground contamination are post-processed to calculate radiological dose from inhalation, air immersion and ground-shine, chemical exposures, and/or lethal dose (chemical/biological) concentration levels, which are related to available federal protective action guide levels for evacuation/sheltering, worker protection, relocation, and agricultural impacts as appropriate. A Web portal provides access to the NARAC system and allows authorized users to run their own simulations, obtain expert analyses from the center, and/or share model predictions with other users. Response capabilities range from fully-automated three-dimensional plume model initial predictions available in 5 to 15 minutes to detailed analyses by the center's subject matter experts.

NARAC personnel provide 24/7 technical and scientific expertise until all airborne releases end, the hazardous areas are defined and mapped, and the long-term impacts are assessed. Staff quality assure model input data, meteorological observations, weather forecasts, and dispersion predictions, estimate unknown source amounts, refine simulations based on field measurement data, and provide information on model product interpretation. In addition, the center provides training and supports exercises and drills. Center personnel also conduct research, develop new modeling tools, and perform risk assessments and other studies.

ATMOSPHERIC AND DISPERSION MODELING ANALYSES FOR FUKUSHIMA

During the Fukushima response, NARAC was simultaneously tasked with providing a wide range of modeling analyses including:

- Daily Japanese weather forecasts and atmospheric transport predictions to inform planning for field monitoring operations and to provide U.S. government agencies with on-going situational awareness of meteorological conditions

- Estimates of possible dose in Japan based on hypothetical U.S. Nuclear Regulatory Commission (NRC) scenarios of potential radionuclide releases to support protective action planning for U.S. citizens
- Predictions of possible plume arrival times and possible dose levels at U.S. locations
- Source estimation and plume model refinement based on atmospheric dispersion modeling and available monitoring data

Each of these efforts is described in more detail in the following sections.

Meteorological forecasting

NARAC provided up to thrice-daily meteorological forecasts to inform field operations and mission planning throughout the three-month period that DOE monitoring teams were deployed in Japan. These forecasts were also distributed to the NRC, elements of the DOD, and other agencies. Animations of generic gas releases were constructed for each forecast period to graphically communicate hourly changes in predicted wind and plume directions, accompanied by tables of wind speed, wind direction, atmospheric stability and precipitation at specified locations.

Weather forecasts were generated using the community Weather Research and Forecast (WRF) model (Skamarock et al. 2008) driven by NOAA Global Forecast System (GFS) model output (Environmental Modeling Center 2003). Periodic consistency checks were made against independent NOAA forecasts and available Japanese meteorological data. Wind fields from 5-km WRF forecasts were primarily used for routine operational support.

Higher-resolution WRF wind fields were developed to support reconstruction of the Fukushima releases (see below) using analysis nudging (Stauffer and Seaman 1994) for the outer model domains (27, 9, and 3 km grid spacing) and observational nudging (Liu et al. 2005) for the innermost domain (1 km grid spacing). These WRF four dimensional data assimilation (FDDA) simulations were repeatedly updated in order to assimilate Japanese meteorological observations as additional data became available. WRF FDDA wind fields at 1 km grid resolution were found to provide limited additional benefit in source reconstruction analyses relative to the 3-km forecasts.

Release scenario modeling

NARAC worked closely with the Department of Energy, the Nuclear Regulatory Commission (NRC), and the White House Office of Science and Technology Policy (OSTP) to construct and predict the impacts from a wide range of hypothetical scenarios. Scenario modeling results provided policy-makers with scientifically-based guidance on possible impacts in Japan and U.S. territories and informed decisions on potential actions that might be needed to protect U.S. citizens in Japan.

The scenario simulations were developed from a range of hypothetical reactor and spent fuel pool source terms provided by the NRC, based on limited available information on conditions in the Fukushima reactor units. Fig. 1 shows the results from one such NARAC ADAPT/LODI model calculation. Although there was a high degree of uncertainty in the NRC-provided source terms, the results provided insight into potential plume arrival times at critical locations and the types of protective actions (sheltering / evacuation, iodine administration, worker protection, relocation) that might need to be considered as reactor conditions evolved. Both separate and combined impacts for the reactor units and spent fuel pools were considered.

A variety of meteorological conditions were used in this “what-if” scenario modeling, including real-world meteorology and artificial conditions with wind directions targeted towards areas with large populations. Although initially NARAC used CMHT-provided Derived Response Levels (DRLs) to convert marker radionuclide concentrations to dose exposures, most of the scenario impacts were simulated by direct calculation of approximately twenty primary dose contributing nuclides, determined in consultation with the NRC and the CMHT.

Estimated U.S. plume arrival times and radiation dose

NARAC simulated trans-Pacific plumes in order to predict potential plume arrival times and possible dose levels in U.S. locations. NARAC modeled the atmospheric transport and dispersion of unit releases of ^{137}Cs and ^{131}I (and in some cases ^{133}Xe) using multiple successive 12 or 24-hour release periods. Dose estimates were derived by scaling the resulting predicted air and ground concentrations by the quantities in selected NRC release scenarios (see above).

Fig. 2 shows four panels from an animation of one trans-Pacific NARAC LODI-model calculation based on NOAA GFS 0.5 degree resolution global meteorological forecasts and/or analyses. The panels portray two-dimensional projections of the modeled marker particles from the LODI model at different times, with particles from each separate 24-hour release period colored differently. The complex nature of the trans-Pacific transport and dispersion process are evident in the patterns shown in the figure.

NARAC calculations conducted during the first week of the response showed that releases on March 11-12 would arrive on the West Coast on March 15-16. This prediction was later found to be consistent with detected plume arrival times (Bowyer et al 2011). However, calculated U.S. arrival times, affected areas, and impacts varied considerably depending on the meteorological conditions during the March to May time period. It also should be noted that upper-level winds transported some the release material faster than near surface winds, but this upper-level plume did not necessarily result in a substantial amount of surface contamination.

Dose conversion factors and derived response levels provided by the DOE/NNSA CMHT were used to convert model-predicted ^{137}Cs deposition levels to early-phase 4-day Total Effective Dose (TED) dose and ^{131}I child thyroid dose to determine if they exceeded the Environmental Protection Agency (EPA) / Food and Drug Administration (FDA) Protective Action Guide (PAG) levels (Sandia National Laboratories, 2010). NARAC/CMHT doses estimates were relatively low-confidence predictions due to the uncertainties in both long-range global weather forecasting and NRC emission scenarios. However in all cases examined, the 96-hour TED dose projections were well below the EPA/FDA 1 rem TED early phase PAG. The grass-cow-milk pathway for child thyroid dose was found to be the dose pathway of greatest concern, but in nearly all locations considered, doses were predicted to be well below the EPA/FDA 5 rem child thyroid PAG level even for conservative NRC emissions scenarios. Measurement data collected by the EPA (EPA 2011) and other agencies later confirmed that levels of concern were not reached in U.S. land areas.

It should be noted that precipitation was not included in the trans-Pacific calculations in order to provide a more conservative estimate of the amount of material that might reach the U.S. Precipitation is a very effective means of removing particulate material from the plume. However, given the known limitations of predicted rain rates and locations in global-scale meteorological predictions, inclusion of precipitation scavenging results could result in unwarranted depletion of the plume.

NARAC source reconstruction and model refinement based on measurement data

In standard DOE/NNSA radiological monitoring support, source estimation and model-refinement are a key component of NARAC's mission. Model predictions are used to guide monitoring and sampling plans and collected data in turn are used to refine model predictions in an iterative process that continues until the contaminated areas are characterized. During the Fukushima emergency, NARAC conducted an initial series of source term estimation and model refinement calculations, although the effort dedicated to this was relatively limited due to the resources invested in, and priorities given to, some of the other activities described above.

During typical responses, NARAC provides an initial plume prediction to deploying field teams to assist in prioritizing areas for initial monitoring and sample collection. Aerial measurement survey and ground monitoring data are electronically transferred to NARAC and/or downloaded from the DOE/NNSA CMHT quality-assured electronic database of monitoring and sampling data. Specialized NARAC software is used to select, filter, and statistically compare these data to a range of model predictions based on different input assumptions. Statistical analyses are performed using data and model results paired in both space and time, supplemented by graphical comparisons. Input assumptions and/or meteorology are then adjusted to improve the model fit to data.

Reconstruction of the Fukushima Dai-ichi atmospheric releases poses a uniquely complicated challenge. Source estimation is an under-constrained non-linear optimization problem, which requires taking into account rapidly changing meteorology (e.g., winds, precipitation), complex terrain, land-sea interfaces, time-varying source terms (e.g., emission rates, radionuclide mix, release height, particle size distribution), multiple potential reactors and spent fuel sources, radioactive decay, and dry and wet deposition processes. Special considerations which needed to be addressed during the Fukushima response included:

- Determination of the key time periods when releases were likely to have occurred, based on a preliminary review of meteorological conditions and environmental radiological measurements, including monitoring data from the nuclear power plant (when available)
- Identification, acquisition, and quality assurance of available Japanese meteorological observations from routinely-available data feeds as well as special Japan networks (provided courtesy of the Japan Atomic Energy Agency)
- Selection, processing, and quality assurance of radiological aerial survey and/or ground monitoring data for model-data analyses
- Determination of the key isotopic dose contributors to be modeled or otherwise accounted for (e.g., ^{131}I, ^{137}Cs, ^{134}Cs, ^{133}Xe) and *a priori* estimation of the approximate activity ratios of the selected radionuclides based on measurement data, reactor analyses, or other information
- Statistical and graphical comparisons of multiple model simulations (using different source terms and meteorological analyses), including use of below-threshold data (null measurements) to constrain possible release periods
- Updated source estimation as identified inconsistencies and/or data gaps were resolved

Meteorology. Continuously-changing complex wind conditions occurred throughout the Fukushima Dai-ichi accident, with multiple periods of on-shore and off-shore flow. As modeled plume and deposition patterns are sensitive to the quantity and quality of meteorological data, grid resolution, and model physics used, NARAC conducted a range of meteorological simulations using both the diagnostic ADAPT model and the predictive WRF model to investigate the accuracy of the resulting predictions of wind fields, precipitation, and other quantities of interest.

Initial NARAC meteorological analyses showed off-shore winds on March 11, shifting to on-shore northward flow on March 12, back to off-shore flow on March 13, followed by a clockwise rotation pushing plumes first to the south (March 14 – 15), then west, northwest, and north (March 15), and off-shore again on March 16. Winds remained primarily off-shore until March 21 when the wind direction again sent radioactive material southward in the general direction of Tokyo.

NARAC's meteorological simulations were later found to be consistent with independent weather analyses (Stohl et al. 2011; Takemura et al. 2011) which showed a well-organized region of surface low-pressure that moved south of Tokyo on March 14 resulting in southward winds at the Fukushima nuclear power plant. A weak low-pressure system moved across central Japan on March 15 bringing light precipitation and southward to northwestward winds at Fukushima Dai-ichi. The two low-pressure systems merged off the east coast of Japan late on March 15 and rapidly intensified. This well-developed storm resulted in strong vertical motion that lifted radioactive material from the boundary layer into the upper atmosphere where it could be transported by the westerly jet stream towards the west coast of the United States (Takemura et al. 2011).

As the first step in its source reconstruction process, NARAC examined meteorological conditions to determine key periods of interest when available radiological data were correlated with prevailing wind directions. Based on the this preliminary analysis, NARAC focused its model refinement efforts on March 14 – 16, a critical time frame in which the largest releases appeared to have occurred during periods of on-shore flow.

NARAC also found evidence of a second period of interest on March 21 – 23 when the wind directions rotated back toward the south and were correlated with elevated radiological monitoring data readings in the direction of Tokyo. Although NARAC did not examine this second period in detail, an analysis of ^{131}I and ^{137}Cs deposition measurements from monitoring stations in 15 Japanese prefectures by Morino et al. 2011 confirmed an increase in deposition rates around Fukushima during 21 to 23 March 2011 due to on-shore winds and precipitation scavenging. A meteorological analysis by Kinoshitaa et al. 2011 similarly concluded that deposition observed in Ibaraki, Tochigi, Saitama, and Chiba prefectures and in Tokyo likely occurred on 21 March.

Precipitation. Precipitation occurred sporadically throughout the Fukushima releases and was found by NARAC to be a significant factor affecting radionuclide transport and deposition during both of the

March periods mentioned in the previous paragraphs. Fig. 3 shows measured precipitation near Fukushima and Tokyo (Japan Weather Agency 2011). A recent paper by Kinoshitaa et al. 2011 confirmed that rainfall occurred over central-eastern Japan during the periods of interest, with precipitation observed between 15 March 0800 UTC to 15 March 1900 UTC in northern Fukushima prefecture and 20 March 2300 UTC to 22 March 21 UTC in Ibaraki, Chiba, Tochigi, and Saitama prefectures and Tokyo.

NARAC used both grid-wide precipitation based on Japanese meteorological observations and precipitation fields derived from NARAC's 3-km-resolution WRF model forecasts in its source reconstruction analyses. As can be seen in Fig. 4, the spatially and temporally varying WRF-generated precipitation fields were found to be generally consistent with measured precipitation data.

Spatially and temporally varying precipitation and associated scavenging due to both in-cloud and below-cloud processes significantly impact deposition patterns. Precipitation may reduce downwind transport, but create local areas of enhanced deposition. Fig. 5 shows an illustrative comparison of relative deposition with and without precipitation for the same uniform release rate and meteorology. These deposition patterns were generated using the FLEXPART model (Stohl et al. 2005; Fast and Easter 2005) from WRF wind and precipitation fields. As can be seen in the figure, a prominent deposition pattern extending northwest of the Fukushima Dai-ichi plant and continuing along a northeast-to-southwest valley further downwind can be produced from precipitation scavenging of airborne radioactivity by rain and possibly snow in the higher elevation areas to the west of the plant. A qualitatively similar high deposition footprint was seen in the AMS measurement data (Lyons et al. 2012; US DOE/NNSA 2011).

Radiological Data. NARAC used a variety of radiological data in its source estimation and model refinement process, although it should be noted that the selection of radiological (and meteorological) data was often determined by data availability at the time the analysis was performed. During the response, NARAC primarily focused on the following sources of radiological data:

- Limited on-site Tokyo Electric Power Company (TEPCO) measurements from mobile instrumentation obtained from the DOE/CMHT electronic radiological database

- Time series of dose-rates provided by the Government of Japan (GOJ) Ministry of Education, Culture, Sports, Science and Technology (MEXT) environmental monitoring stations (GOJ 2011d†)
- Dose-rate data provided by the DOE Aerial Measuring System (AMS)
- DOE and DoD monitoring data provided via the DOE/CMHT electronic radiological database

Key time gaps existed in the TEPCO data due to the failure of plant monitoring stations during the earthquake or tsunami and a site evacuation that occurred on March 15 (GOJ 2011c). Most MEXT regional prefectural monitoring station data were available only for the period following 0900 UTC March 15, although some Fukushima prefecture data are available earlier. The MEXT dose rate measurements were assumed to include contributions from both air immersion and ground exposure.

During the response, NARAC worked closely with the other national laboratory components of DOE/NNSA's Consequence Management Home Team (CMHT) to acquire, process, and quality assure the radiological data. However, further review of these data for modeling purposes is necessary to identify inconsistencies and gaps, exclude unrepresentative data, take background into account, and make sure the data is properly interpreted for comparison against NARAC model results. Analysis of additional data also should be undertaken in order to confirm or further refine existing source term estimates.

Preliminary analysis of the MEXT data showed progression of the plume over the March 14-16 period to the south, west, northwest, and then north of the Fukushima Dai-ichi plant, consistent with the meteorological analysis described above. It was assumed that most of the on-shore radiological deposition to the west and north of the plant occurred before March 20, a hypothesis that was supported by later AMS measurements that showed no significant additional deposition in those areas after that date (US DOE/NNSA 2011).

Radionuclide mix. During the response, NARAC focused its source estimation calculations on a few key radionuclides: ^{137}Cs, ^{134}Cs, ^{131}I, and ^{133}Xe. The relative activity ratios of ^{133}Xe:^{131}I:^{137}Cs:^{134}Cs were

† During the response, the DOE/NNSA and NARAC received Japanese radiological monitoring data by email from the GOJ. Most of this data are posted on the web site cited in GOJ 2011d.

usually taken to be 100:10:1:1. These ratios were derived from limited DOE/NNSA radionuclide sample analysis data and NRC reactor scenario radionuclide mixes, although a few excursions from these values were explored. A ^{131}I:^{137}Cs: average activity ratio of 10:1 is approximately consistent with independent estimates made by Chino et al. 2011 and the Government of Japan (GOJ 2011c), although the former paper also shows an analysis in which ^{131}I: ^{137}Cs ratios vary over 2 orders of magnitude (Table 2 in Chino et al., 2011).

Although not included in all modeling cases, ^{134}Cs activity may be inferred from ^{137}Cs activity assuming an average ^{134}Cs:^{137}Cs activity ratio of approximately 1:1. This ratio is consistent with a wide range of DOE laboratory sample analyses, although there was found to be wide variation between individual samples (Musolino et al. 2012). If other potentially released radionuclides (e.g., ^{132}I, ^{132}Te) are included, the NARAC-estimated release activities of ^{137}Cs, ^{131}I, and ^{133}Xe are expected to decrease.

Source term estimates. NARAC conducted a number of source reconstruction analyses using a range of possible release assumptions and meteorological conditions. In these simulations, reactor and spent fuel pool emissions were treated as a single time-varying source. 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. Varying the latter factors generally resulted in changes that were small compared to that produced by different emission rates and meteorology.

For the source estimates presented in this paper, NARAC optimized the fit to dose-rate data using comparisons of model predicted values paired in space and time to the available measurement data. The emission rates were taken to be the values that improved the *overall* agreement with the data at all locations covering the entire modeled period. The NARAC analyses used Japanese MEXT measurements for total dose rate, assumed be the sum of "cloud-shine" (air immersion) plus "ground-shine" (ground exposure) dose rate, and/or AMS ground-shine dose rate measurements (Lyons et al. 2012). NARAC model-predicted air and ground activity concentrations were converted to dose rate using air immersion and ground exposure dose conversion factors (Eckerman and Leggett, 2008) and applying a ground roughness factor of 0.82 to the predicted ground exposure dose (Likhtarev et al. 2002).

For example, NARAC performed a source reconstruction using ADAPT/LODI simulations of ^{137}Cs, ^{131}I, and ^{133}Xe released during the critical period from March 14 0600 UTC to March 16 0600 UTC. The meteorology was derived from WRF-generated 3-km wind and precipitation fields. The ^{137}Cs: ^{131}I:^{133}Xe activity ratios were assumed to be 1:10:100. NARAC estimated the source term for this case using approximately 400 hourly dose rate measurements at 19 regional and Fukushima prefecture MEXT stations, although as discussed above the majority of these data were from the period after March 15 0900 UTC. Assuming a constant release rate over the entire 48-hour period, the model fit to data resulted in total release quantities of 1.85×10^{16} Bq (5×10^{5} Ci) of ^{137}Cs, 1.85×10^{17} Bq (5×10^{6} Ci) of ^{131}I, and 1.85×10^{18} Bq (5×10^{7} Ci) of ^{133}Xe (Table 1). For the purposes of this paper, this analysis will be referred to as the “NARAC baseline” case.

Fig. 6 shows an example comparison of NARAC baseline case results to MEXT dose rates for one March 15 1200 UTC. Using all available observations, NARAC found that approximately 30% of the baseline case predicted values were within a factor of 2 of the MEXT measurements in Fukushima prefecture (e.g., the ratios of measured and predicted values for the same time and location were between 0.5 and 2), and approximately 60% were within a factor of five. For regional MEXT stations located farther away from the plant, more than 40% of model predicted values were within a factor of 2 of measured dose rates and 80% within a factor of five. NARAC model-predicted values also were found to be within a factor five of the AMS data more than 60% of the time, even though these data were not used in the NARAC source estimation process,

It should be noted that other release rates from different NARAC analyses provided comparable statistical agreement to the available dose-rate data, with the corresponding NARAC estimates of release quantities varying by as much as a factor of three from the baseline case. The range of possible source estimates resulted from the use of different input assumptions (e.g., time-varying vs. constant emission rates), the choice of meteorology, assumptions on the modeled radionuclide mix and activity ratios, and the selection of the source of radiological data to preferentially match in the model refinement process. In general, NARAC found that high-resolution meteorological fields were needed to capture many of the detailed features of plume transport and deposition.

Not surprisingly, time-varying release rates provided better fits to the time variations in MEXT station dose rate data and captured some of the peaks in the measurement data time series. Fig. 7 shows the results of one such analysis for the period March 12 0100 UTC to March 18 1400 UTC based on meteorology developed from Japanese weather observations and uniform grid-wide precipitation rates. The time-varying emission rate model fit to data totaled to $4.7x10^{15}$ Bq of ^{137}Cs, $4.7x10^{16}$ Bq of ^{131}I, and $4.7x10^{17}$ Bq of ^{133}Xe over the simulation period. The figure compares NARAC ADAPT/LODI predicted values against gamma dose-rate data for two locations– Aizu-wakamatsu and Iwaki (GOJ 2011d), which are located approximately 30 km west and 100km south of the Fukushima Dai-ichi respectively, with somewhat less consistent agreement in terms of magnitudes and timing of peaks found for other stations used in the analysis. More than 50% of the model predicted values were within a factor of two of MEXT measurements in Fukushima prefecture and 70% within a factor of five. Similar comparison statistics for more distant regional MEXT measurements show 40% within a factor of two and 80% within a factor of five. Approximately 80% of predicted values were within a factor of five of AMS measurement values.

Fig. 8 shows another illustrative source term analysis based on the Aerial Measuring System (AMS) data alone. This ADAPT/LODI simulation from March 15 0300 UTC to March 16 0200 UTC used time-dependent releases of ^{137}Cs, ^{134}Cs, and ^{131}I (with an assumed a relative activity ratio of 1:1:10), with meteorological observations and a uniform grid-wide precipitation. The model fit to data resulted in total release quantities for ^{137}Cs, ^{134}Cs, and ^{131}I over the simulation period of 1.1 x 10^{16} Bq (3×10^5 Ci), 1.1 x 10^{16} Bq (3×10^5 Ci) and 1.1 x 10^{17} Bq (3×10^6 Ci), respectively.

The left panel of Fig. 8 shows a comparison of LODI model predicted dose rate (color-filled contours) to the March 18 AMS data (small circles with values color coded in the same manner as the contours). The number of AMS data points has been significantly thinned in order to improve visualization of the comparison. Approximately 40% of predicted values were within a factor of two of AMS measurement values from this date and more than 80% within a factor of five.

The March 26 comparison shown in the right hand panel of Figure 8 provides a confirmation of the original source reconstruction, as these data were not used in developing the source estimate but model the results of radioactive decay over the intervening eight days. In this case, more than 80% of the model values agreed with the AMS data within a factor of two and 98% within a factor of five.

The AMS data (Lyons et al. 2012) used in most NARAC analyses were primarily derived from the aerial surveys that measured the highest deposition area extending to the northwest of the Fukushima Dai-ichi site. Dose rate predictions for this time period have significant uncertainty because of the uncertainty in predictions of precipitation scavenging, which caused much of the deposition in this region. AMS data for this period also needed substantial corrections to account for terrain elevation and for the contributions of any airborne plume to the measurements.

The complete set of NARAC source term analyses showed that multiple emission rates can be consistent with the available data within model and measurement uncertainties. Source term estimates based on measurement-model comparisons are sensitive to details and timing of wind shifts, precipitation, and release rates, the selection of the type of data which is preferentially optimized (e.g., AMS, MEXT), or different assumptions regarding release characteristics, radionuclide inventories, and/or reactor conditions. Predicted ground-shine deposition patterns are heavily influenced by precipitation scavenging, especially the northwest deposition "footprint" measured by the Aerial Measuring Survey (AMS).

COMPARISON TO OTHER SOURCE ESTIMATES

In this section, the NARAC baseline case is compared to previous studies. Table 1 summarizes activity release estimates from several recent studies, which used different computer models, measurement data, and source estimation techniques. An analysis conducted by Chino et al. 2011 estimated a total discharge of 1.3×10^{16} Bq of ^{137}Cs and 1.5×10^{17} Bq of ^{131}I from March 12 0100 UTC to April 51500 UTC (March 12 1000 JST to April 6 0000 JST) based primarily on "air dust sampling measurements" of those radionuclides, as well as some dose rate measurements. In this analysis, if multiple measurements were available from different locations for a time, only the maximum value was used. The authors estimated the error in their release quantity estimates as "at least a factor of 5".

The Government of Japan (GOJ 2011a; GOJ 2011b, GOJ 2011c) has provided several estimates of total release rates based on different methodologies. Table 1 also includes the latest published GOJ release estimates (GOJ 2011c) from two sources – the JAEA and NISA. The JAEA estimated that a total quantity of 1.1×10^{16} Bq of ^{137}Cs was released over approximately 24 days (March 12 – April 5) in what appears to be a revision to the Chino et al. 2011 analysis. An alternative Nuclear and Industrial Safety Agency (NISA) estimate based on a plant behavior analysis estimated a release total of 1.5×10^{16} Bq of ^{137}Cs over approximately 4 days shortly after accident initiation (GOJ, 2011c). This estimate is approximately 4.5 greater than the JAEA estimate for the same 4-day period.

Stohl et al. 2011 used an estimation procedure that combined *a priori* assumed release rates based on information on plant conditions with atmospheric dispersion computer model predictions and comparisons to measurement data. Their analysis was based on long-range Comprehensive Test Ban Treaty Organization (CTBTO) ^{133}Xe and ^{137}Cs air concentration measurements and a limited set of regional Japanese ^{137}Cs air concentration measurements. Table 1 shows the Stohl et al. 2011 release estimates of 3.6×10^{16} Bq of ^{137}Cs, with an estimated uncertainty range of 2.33×10^{16} to 5.01×10^{16} Bq, for March 11 to April 20. This estimate is a factor of 2-3 times higher than the Government of Japan (GOJ 2011c) estimates. Stohl et al. 2011 also provided an estimate for ^{133}Xe, based on the assumption that all xenon was released prior to March 16.

The DOE/NNSA CMHT used AMS ground-shine dose rate data collected out to 50 miles from the plant during April 6–29, 2011, to estimate a total deposited activity of 2.7×10^{15} Bq for both ^{134}Cs and ^{137}Cs (with an estimated range of 0.7×10^{15} to 3.7×10^{15} Bq)[‡]. This estimate does not account for airborne (non-deposited) material. Using the Stohl et al. 2011 estimate that 19% of the ^{137}Cs emission was deposited on land, this AMS-based estimate would correspond to an estimated ^{137}Cs release quantity of approximately1.4×10^{16} Bq.

Table 2 compares the NARAC baseline case activity release estimate for the period from 06:00 UTC on March 14, 2011 to 0600 UTC on March 16, 2011 to estimates of the release rate derived from

[‡] Okada C, Remote Sensing Laboratory, Las Vegas NV; 2011 (private communication).

several of the previous studies discussed above. NARAC estimated the latter values from time-varying release rates provided in the cited papers. The two-day period compared is estimated to cover between 25-50% of the total release based on the references in Table 2. It should be noted that since NARAC estimated total dose-rate using only a few key isotopes, corrections for the presence of other radionuclides (e.g., ^{132}I, ^{132}Te) are likely to reduce estimates of total activity of the three radionuclides quoted in the table.

It is both interesting and encouraging to note that all of the source term estimates in Table 2 are within a factor of approximately six (and more than half are within a factor of approximately three) despite the different source reconstruction methodologies, meteorological models, types of radiological data , and reactor conditions assumed. At this point in time, it is difficult to determine if any of these analyses is to be preferred, especially given the uncertainty and large variability in the spatial and temporal patterns of air concentration and deposition and the limitations of the available data. It is important to note that Table 2 covers the primary period of on-shore transport of radioactivity. However, wind directions were off-shore for considerable periods of time. Source estimates for off-shore-wind times are significantly more speculative as Japanese radiological measurement data are generally unavailable for these periods.

CHALLENGES OF THE FUKUSHIMA RESPONSE

The Fukushima response involved the greatest sustained level of NARAC effort in the more than three decade long existence of the center. Although NARAC successfully provided a wide range of highly-valued products and analyses during the response, the experience also identified a number of scientific and operational challenges that are being documented as part of the DOE/NNSA After Action Review process. Some of the key operational challenges encountered by NARAC are summarized below.

- Both personnel and computations resources were strained to support the many different types of analyses requested and to meet the desired response times.
- High-level expertise was at a premium as it was critical to developing and quality assuring new non-standard and/or complex analyses required to answer key questions.

- More efficient means to address complex (multiple reactor unit) nuclear power plant scenario analyses were needed, including improved source term estimation tools and closer NARAC-NRC ties, documentation, and procedures.
- Communications and sharing of key information with other DOE assets and other federal government agencies was limited by the available time and resources.
- Management and archiving of the overwhelming information flow was challenging and time consuming.

The After Action process has also identified scientific needs to:

- Develop a set of well-understood nuclear power plant scenarios for different reactor conditions that can be used in future accidents
- Build a complete quality-assured data set of all available Fukushima-related meteorological and radiological data, especially for the period covering the first week following the tsunami when the data are relatively sparse
- Improve modeling of complex meteorology and precipitation on both the local and global scales and further investigate the impacts of precipitation in order to reduce the uncertainty in model predictions
- Complete a comprehensive analysis that combines knowledge of nuclear reactor conditions, data from field measurement and lab sample analyses, and modeling to improve source estimates and radionuclide inventories and develop a more accurate reconstruction of the accident

Actions are underway to address some of the above needs and challenges, including the development of upgraded NARAC computational hardware and software to increase throughput capacity and reduce turn-around time, procedures to improve connectivity among teams involved in the response, approaches for handling large volume information and data flow, and improved interagency communications. Such efforts are leading to improvements in DOE and NARAC's ability to respond to a Fukushima-scale domestic event.

CONCLUSION

NARAC provided a wide range of predictions and analyses during the Fukushima Dai-ichi crisis, including weather forecasts, simulations of dose levels in Japan resulting from hypothetical release scenarios, predictions of arrival times and dose levels reaching U.S. territories, and source estimates based on the incorporation of field measurement data. A number of scientific and operational challenges were encountered during the response, some of which are already being addressed.

The releases from the Fukushima Dai'ichi nuclear power plant are still incompletely characterized due to the long-term duration of the event, the rapidly changing and still unknown reactor and spent fuel conditions at multiple units, the complicated geography of the region, the highly-variable meteorological conditions, and the relatively limited data available during the early stages of the event when the most significant releases are likely to have occurred. NARAC found that a range of emission rates and quantities are consistent with the available data. To reduce the range in uncertainty in source estimates, additional high-resolution studies using all available data are needed. Future activities to be considered include:

- Collection, quality assurance, and verification of all available meteorological and radiological data from Japan, including consideration of background, instrument thresholds, measurement uncertainties, and data interpretation (e.g., separation of air immersion from ground-shine dose)
- Development of a better understanding of the complex interplay between time-varying release characteristics and meteorological conditions
- Improvements in meteorological modeling to more accurately simulate rapidly shifting wind conditions, spatially and temporally-varying precipitation, and long-range trans-oceanic/continental transport in order to improve predictions of plume arrival times and spatial patterns
- Determination of the degree to which different reactor unit releases can be distinguished via time-varying radionuclide signatures and/or reactor analyses and whether actinide signatures indicative of core material releases were detected

- Investigation of the sensitivity of modeling results to details of release characteristics (e.g., time-varying rates, release heights, radionuclide mix, particle size distribution)
- Determination of the degree to which on-shore radiological data from Japan can be used to constrain release rate estimates during off-shore periods for which local and regional data are unavailable
- Analysis of the complete set of long-range radiological data sets, including Comprehensive Test Ban Treaty Organization (CTBTO), EPA RadNET (EPA 2011), data from U.S. nuclear power plants, and comparison of source estimation based on these data to values based on measurements in Japan

The Fukushima event provides a unique and voluminous data set, only a small portion of which has been analyzed and incorporated into this and previous studies. Additional data should be analyzed, and used to advance and evaluate methodologies for meteorological forecasting, dispersion modeling, data assimilation, dose assessment, and source reconstruction. Such improvements will lead to a better understanding of the Fukushima accident and will enhance our capabilities for responding to future incidents.

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FIGURE CAPTIONS

Fig. 1. This figure shows the results from a NARAC simulation of ground level dose used to predict areas exceeding protective action guides in Japan. These results are for an example hypothetical release scenario, one of many provided by the Nuclear Regulatory Commission. Scenario calculations used both actual and artificial meteorology and were used to inform U.S. government emergency planning.

Fig 2. The panels show four frames from an animation of the trans-Pacific transport and dispersion of LODI modeled marker particles from potential releases at the Fukushima Dai-ichi plant (times are marked on each frame). Particles of the same color were released during the same 24-hour interval.

Fig 3. This figure plots observed daily precipitation at Tokyo (red) and Fukushima City (blue) from March 12 to May 31, 2011 obtained from JWA 2011.

Fig 4. The panels show comparisons of 3-km resolution WRF-modeled precipitation rates (square color pixels) to Japan Meteorological Agency (JMA) station observations (circles color coded in the same manner as the pixels) at four different times. The precipitation data was obtained courtesy of the Japan Atomic Energy Agency (JAEA).

Fig 5. The two panels compare the NARAC-predicted relative deposition resulting from a constant release rate of a normalized amount of material over the period for March 14 1000 UTC to March 15 1800Z without precipitation scavenging (left panel) and with precipitation scavenging (right panel). The simulation was performed using FLEXPART based on 1-3 km resolution WRF modeled winds and precipitation. Colors correspond to the following normalized deposition values: blue $>2\times10^{-13}$, green $>1\times10^{-10}$, yellow $>7\times10^{-10}$, orange $>9\times10^{-10}$, red $>1.1\times10^{-9}$ m^{-2}. The yellow cross shows the location of the Fukushima Dai-ichi plant.

Fig 6. 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 1200 UTC. The contour levels were selected to best show the comparison to data. The red contour is the area where the model predicts that 60 µGy/h (6.0 mR h^{-1}) is exceeded; pink shows 2-60 µGy h^{-1} (0.2-6.0 mR h^{-1}), orange 0.6-2 µGy h^{-1} (0.06 – 0.2 mR h^{-1}), light orange 0.004-0.6 µGy h^{-1} (0.004-0.06 mR h^{-1}), and yellow 0.006-0.04 µGy h^{-1} (0.0006-0.004 mR h^{-1}). The blue circle indicates the location of the Fukushima Dai-ichi plant.

Fig 7. Comparisons of NARAC-predicted dose rates (red) and MEXT measured dose rates (blue) are shown for two locations – Aizu-wakamatsu and Iwaki. The NARAC simulation used a time-varying source estimate as discussed in the text.

Fig 8. The results of a NARAC analysis based on a time-varying release of ^{137}Cs, ^{134}Cs, and ^{131}I developed from AMS data from March 18, 2011. The panel compares LODI model predicted dose rates (color-filled contours) with AMS data (circles colored coded in the same manner as the contours) at two different times – March 18 and March 26. The dark red, red, dark orange, orange, and yellow contour levels shown correspond to levels greater than 100, 10, 1, 0.1, 0.01 µGy h^{-1} (10, 1, 0.1, 0.01, 0.001 mrad

h^{-1}), respectively. For visual clarity the number of AMS data points plotted has been significantly reduced. The blue circle indicates the location of the Fukushima Dai-ichi plant.

Table

Table 1. Released radioactivity estimates from previously published studies and the NARAC baseline case. The analyses cover different time periods as summarized in the table. Note that the NARAC baseline release estimates for the listed radionuclides in this table may decrease with the inclusion of additional radionuclides in the analysis.

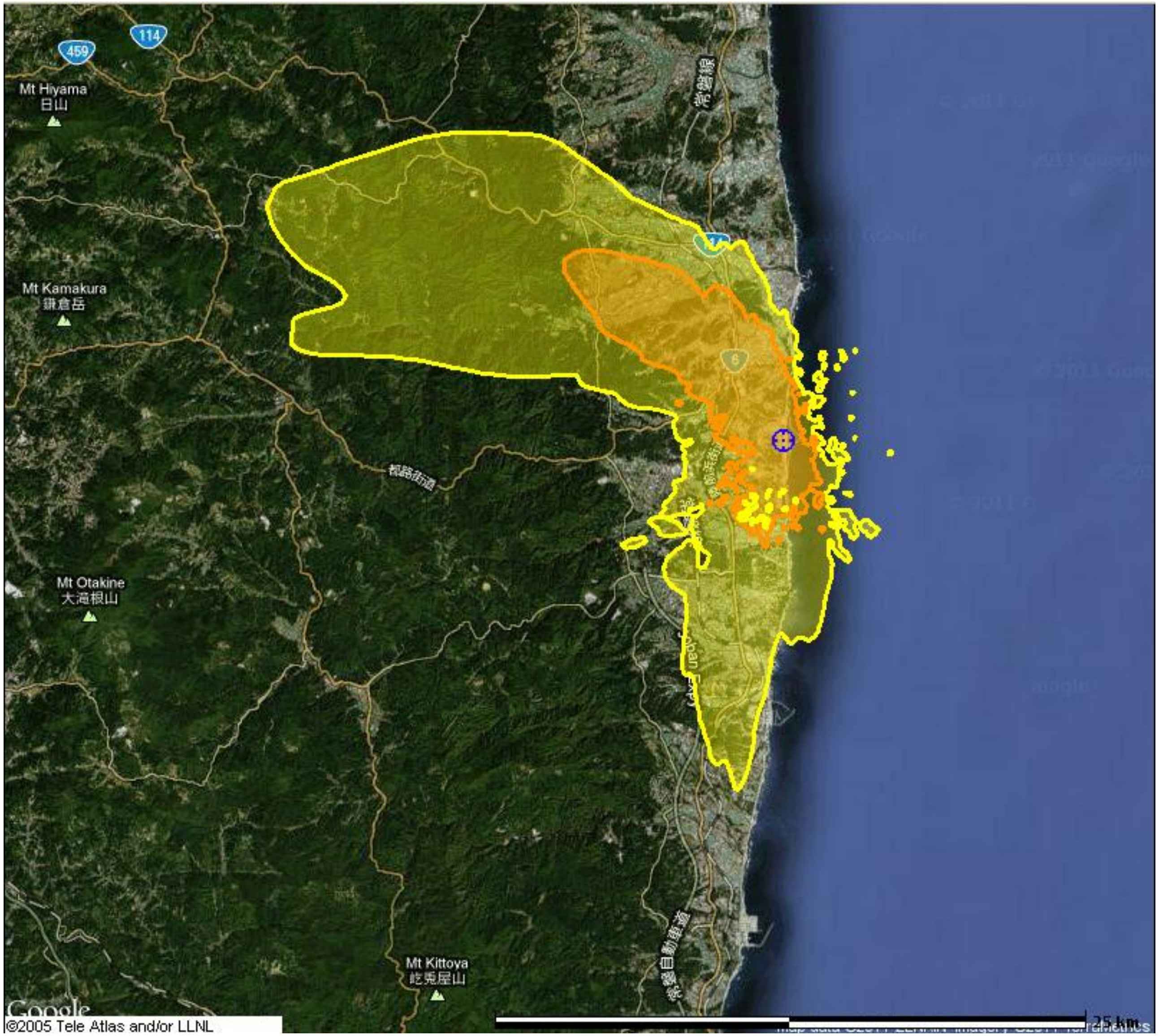
Radionuclide	NISA (GOJ 2011c)	JAEA (Chino et al. 2011)	JAEA (GOJ 2011c)	Bowyer et al. 2011	Stohl et al. 2011	CMHT analysis based on AMS data from April 6-29	NARAC Baseline
Release Time Period (UTC)	March 12-16	March 12-April 5	March 12-April 5	March 11-14	March 11-April 20	Before April 29	March 14-16
^{137}Cs	1.5×10^{16} Bq	1.3×10^{16} Bq	1.1×10^{16} Bq	□	3.6×10^{16} Bq	1.4×10^{16} Bq	1.85×10^{16} Bq
^{134}Cs	1.8×10^{16} Bq	□	□	□	□	1.4×10^{16} Bq	□
^{131}I	1.6×10^{17} Bq	1.5×10^{17} Bq	1.3×10^{17} Bq	□	□	□	1.85×10^{17} Bq
^{133}Xe	□	□	□	1.2×10^{19} Bq	1.7×10^{19} Bq (March 11-15)	□	1.85×10^{18} Bq

Table

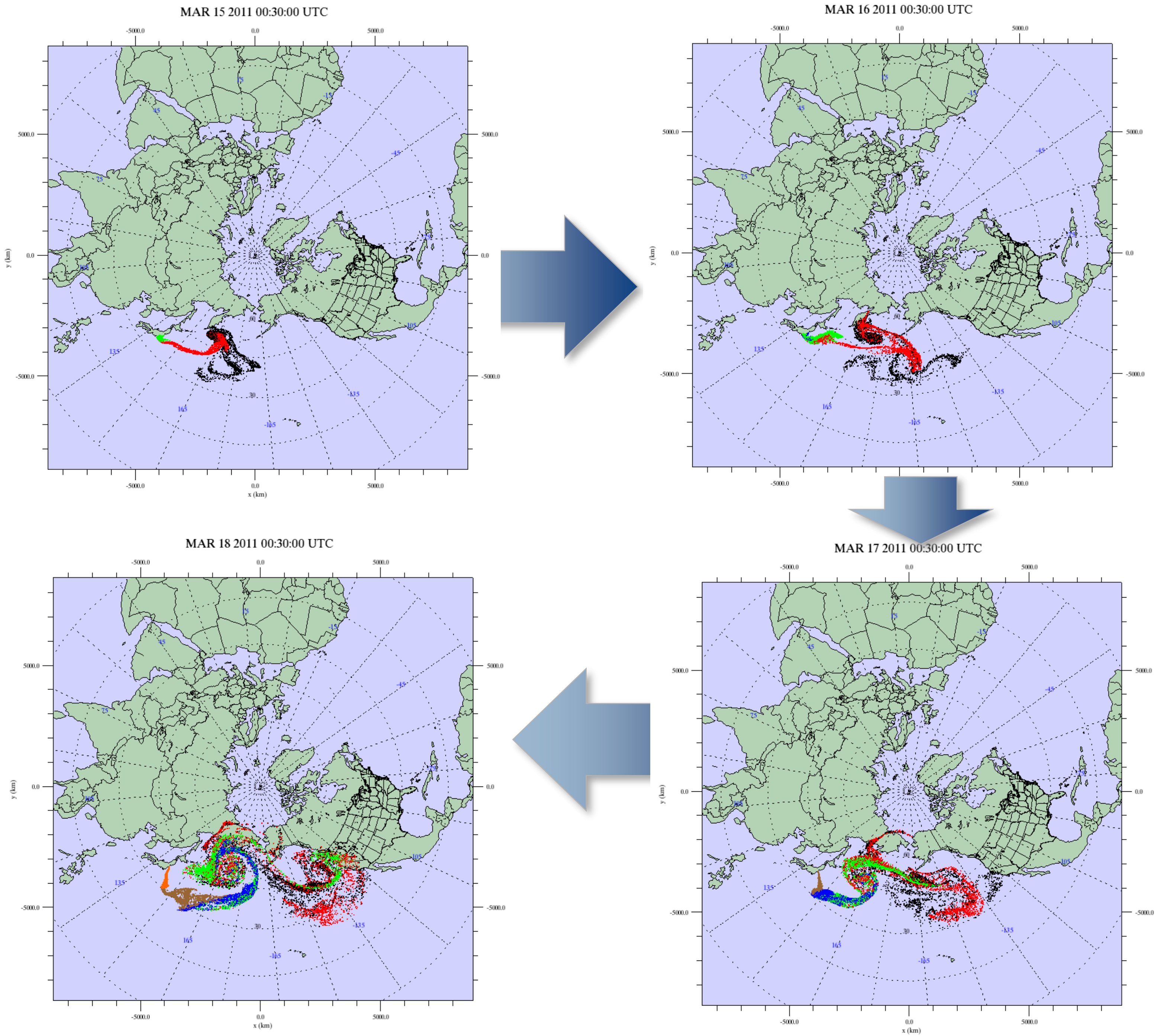
Table 2. This table compares the NARAC baseline case released activity estimate for the period from 06:00 UTC March 14, 2011 to 06:00 UTC March 16, 2011, to estimated values from previously published studies. NARAC estimated the release quantities and the percentage of the total release quantity over the two day period from tables and figures in the cited references that covered longer release periods (except for the GOJ 2011c estimate for which the published data ends on 15:00 UTC March 15). NARAC baseline release estimates for the radionuclides in this table may decrease with the inclusion of additional radionuclides in the analysis.

Radionuclide	JAEA (Chino et al. 2011)	JAEA (GOJ 2011c)	Stohl et al. 2011	NARAC Baseline
^{131}I	6.87×10^{16} Bq (1.86×10^{6} Ci) 45%	2.94×10^{16} Bq (7.96×10^{5} Ci) 23%	□	1.85×10^{17} Bq (5×10^{6} Ci)
^{137}Cs	6.49×10^{15} Bq (1.75×10^{5} Ci) 51%	2.94×10^{15} Bq (7.96×10^{4} Ci) 27%	1.72×10^{16} Bq (4.65×10^{5} Ci) 48%	1.85×10^{16} Bq (5×10^{5} Ci)
^{134}Cs	□	□	□	□
^{133}Xe	□	□	5.68×10^{18} Bq (1.54×10^{8} Ci) 34%	1.85×10^{18} Bq (5×10^{7} Ci)

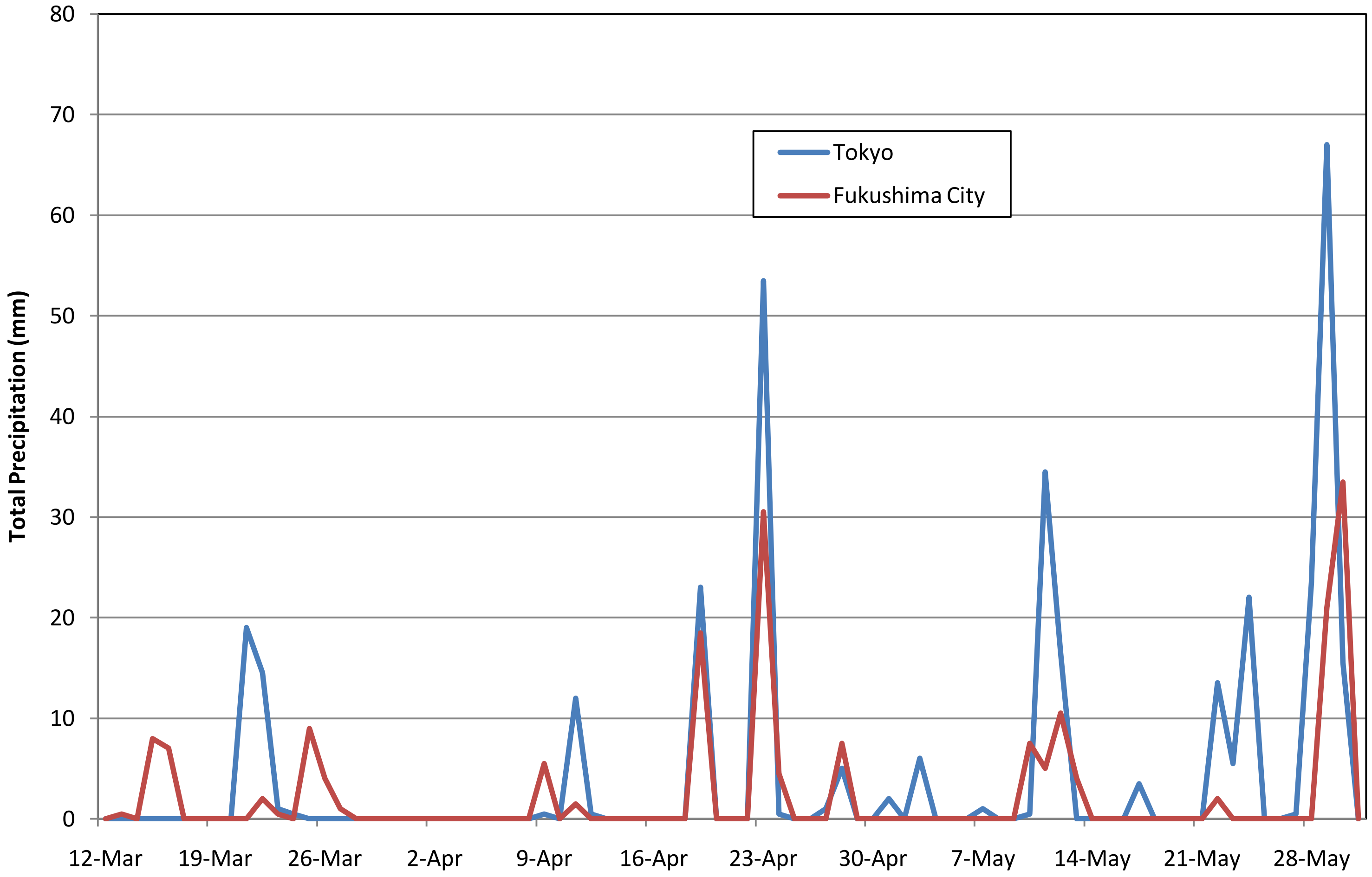
Figure


459
114
Mt Hiyama
日山
Mt Kamakura
鎌倉岳
Mt Otakine
大滝根山
Mt Kittoya
屹兎屋山
常磐線
常磐自動車道
Google
©2005 Tele Atlas and/or LLNL
25 km

Figure

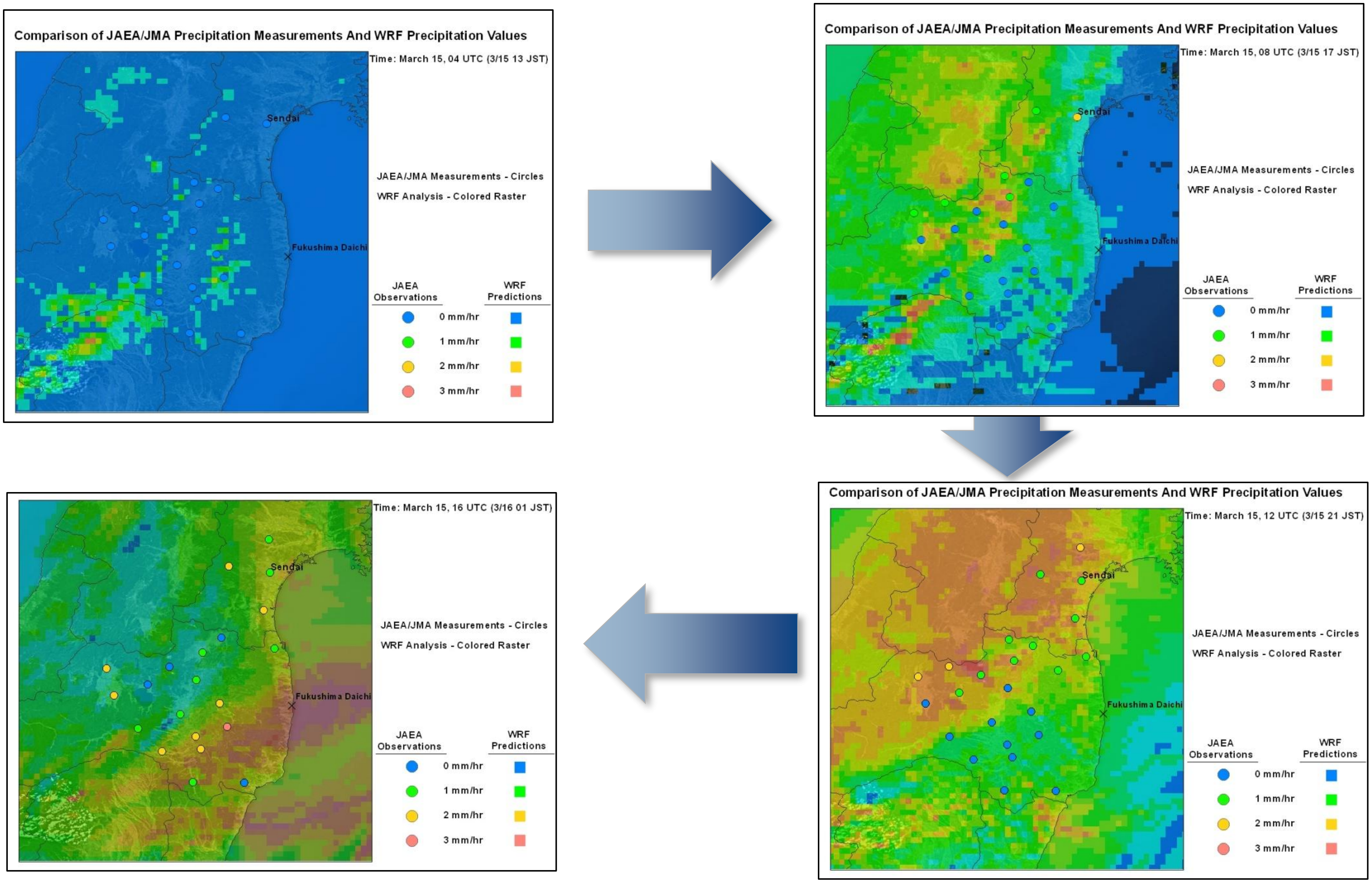

MAR 15 2011 00:30:00 UTC
MAR 16 2011 00:30:00 UTC
MAR 17 2011 00:30:00 UTC
MAR 18 2011 00:30:00 UTC
x (km)
y (km)
-5000.0
0.0
5000.0

Figure

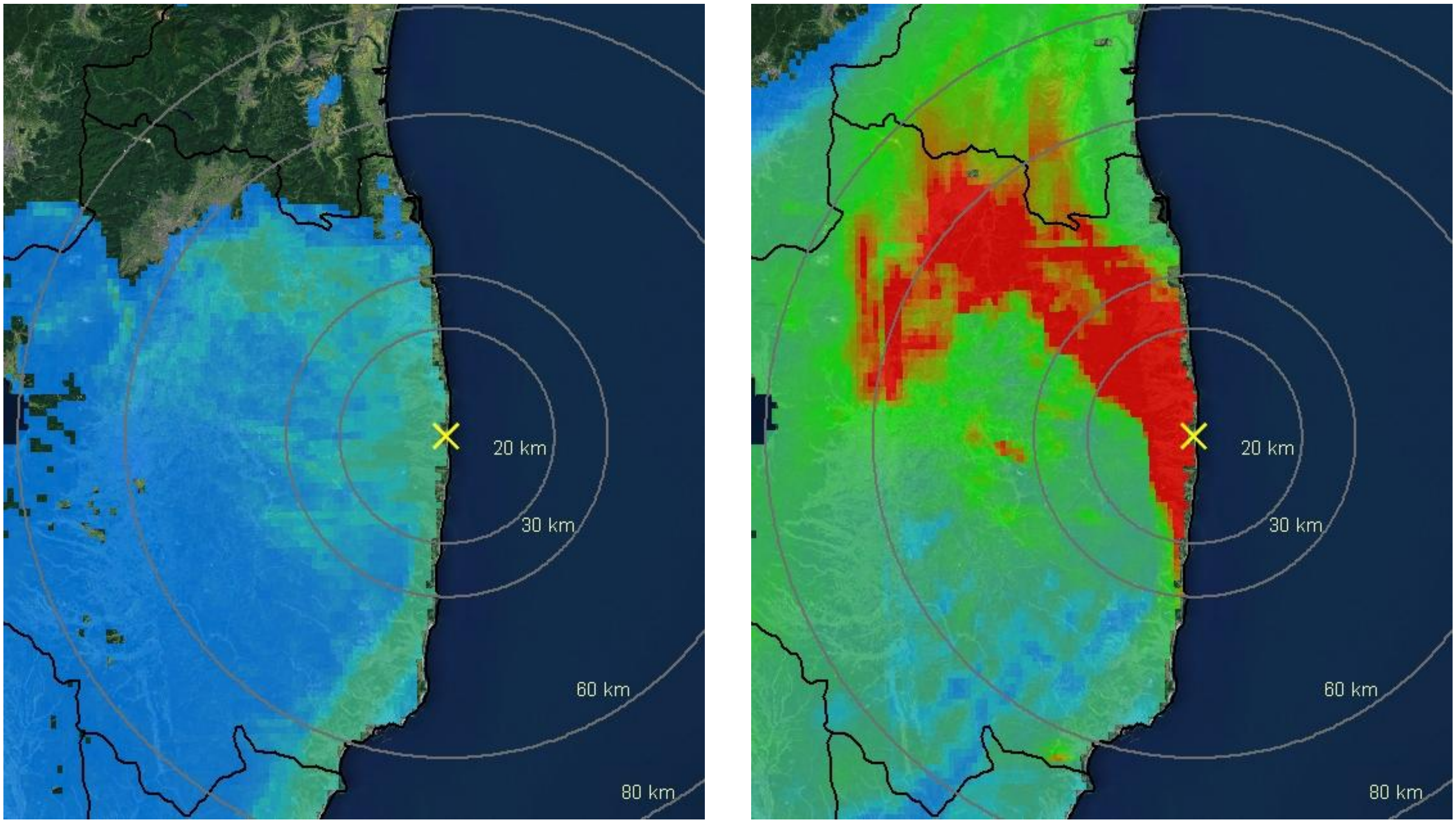


Tokyo
Fukushima City
Total Precipitation (mm)
80
70
60
50
40
30
20
10
0
12-Mar
19-Mar
26-Mar
2-Apr
9-Apr
16-Apr
23-Apr
30-Apr
7-May
14-May
21-May
28-May

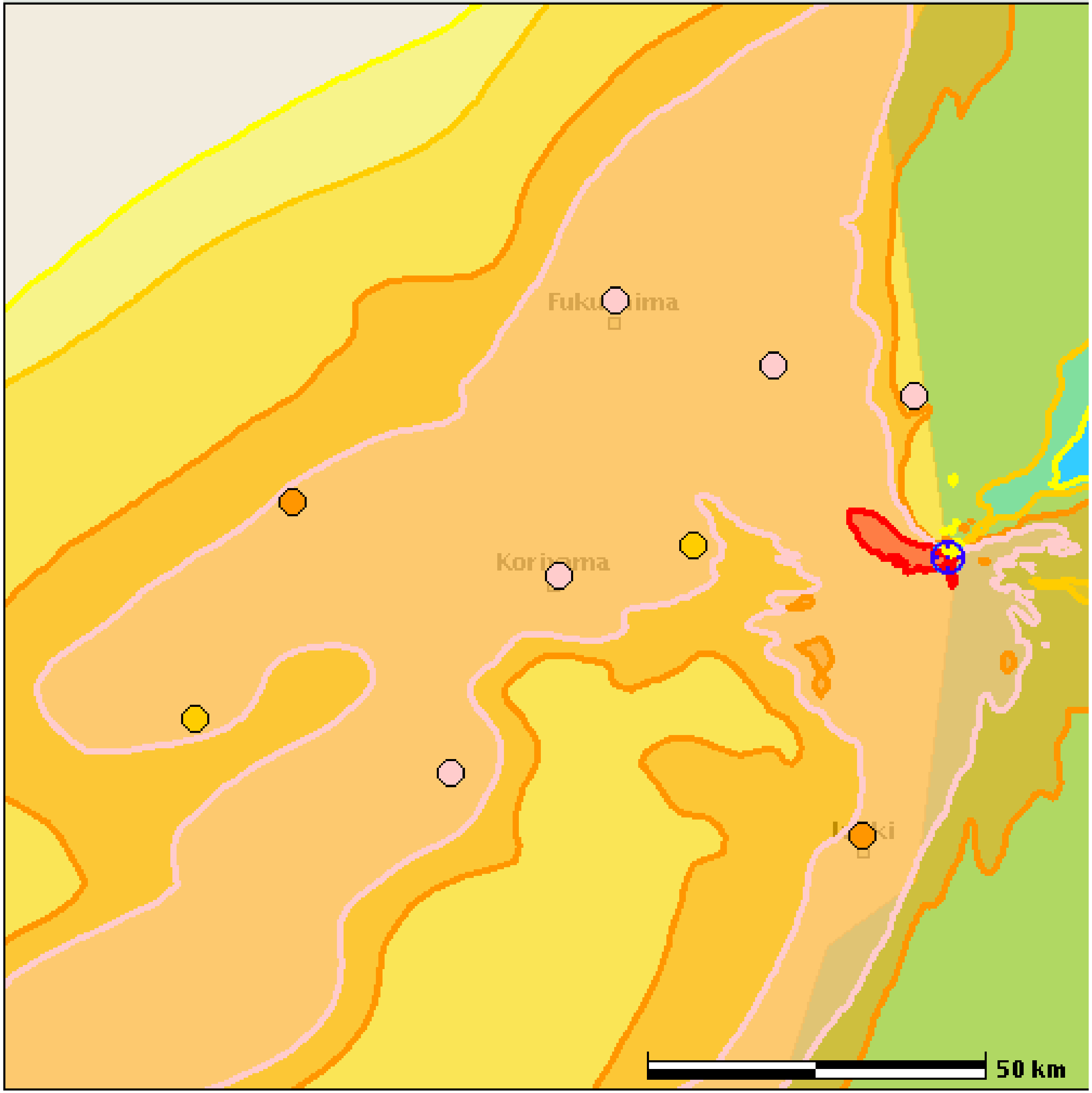
Figure


Comparison of JAEA/JMA Precipitation Measurements And WRF Precipitation Values
Time: March 15, 04 UTC (3/15 13 JST)
Sendai
Fukushima Daichi
JAEA/JMA Measurements - Circles
WRF Analysis - Colored Raster
JAEA Observations
WRF Predictions
0 mm/hr
1 mm/hr
2 mm/hr
3 mm/hr
Comparison of JAEA/JMA Precipitation Measurements And WRF Precipitation Values
Time: March 15, 08 UTC (3/15 17 JST)
Sendai
Fukushima Daichi
JAEA/JMA Measurements - Circles
WRF Analysis - Colored Raster
JAEA Observations
WRF Predictions
0 mm/hr
1 mm/hr
2 mm/hr
3 mm/hr
Comparison of JAEA/JMA Precipitation Measurements And WRF Precipitation Values
Time: March 15, 12 UTC (3/15 21 JST)
Sendai
Fukushima Daichi
JAEA/JMA Measurements - Circles
WRF Analysis - Colored Raster
JAEA Observations
WRF Predictions
0 mm/hr
1 mm/hr
2 mm/hr
3 mm/hr
Time: March 15, 16 UTC (3/16 01 JST)
Sendai
Fukushima Daichi
JAEA/JMA Measurements - Circles
WRF Analysis - Colored Raster
JAEA Observations
WRF Predictions
0 mm/hr
1 mm/hr
2 mm/hr
3 mm/hr

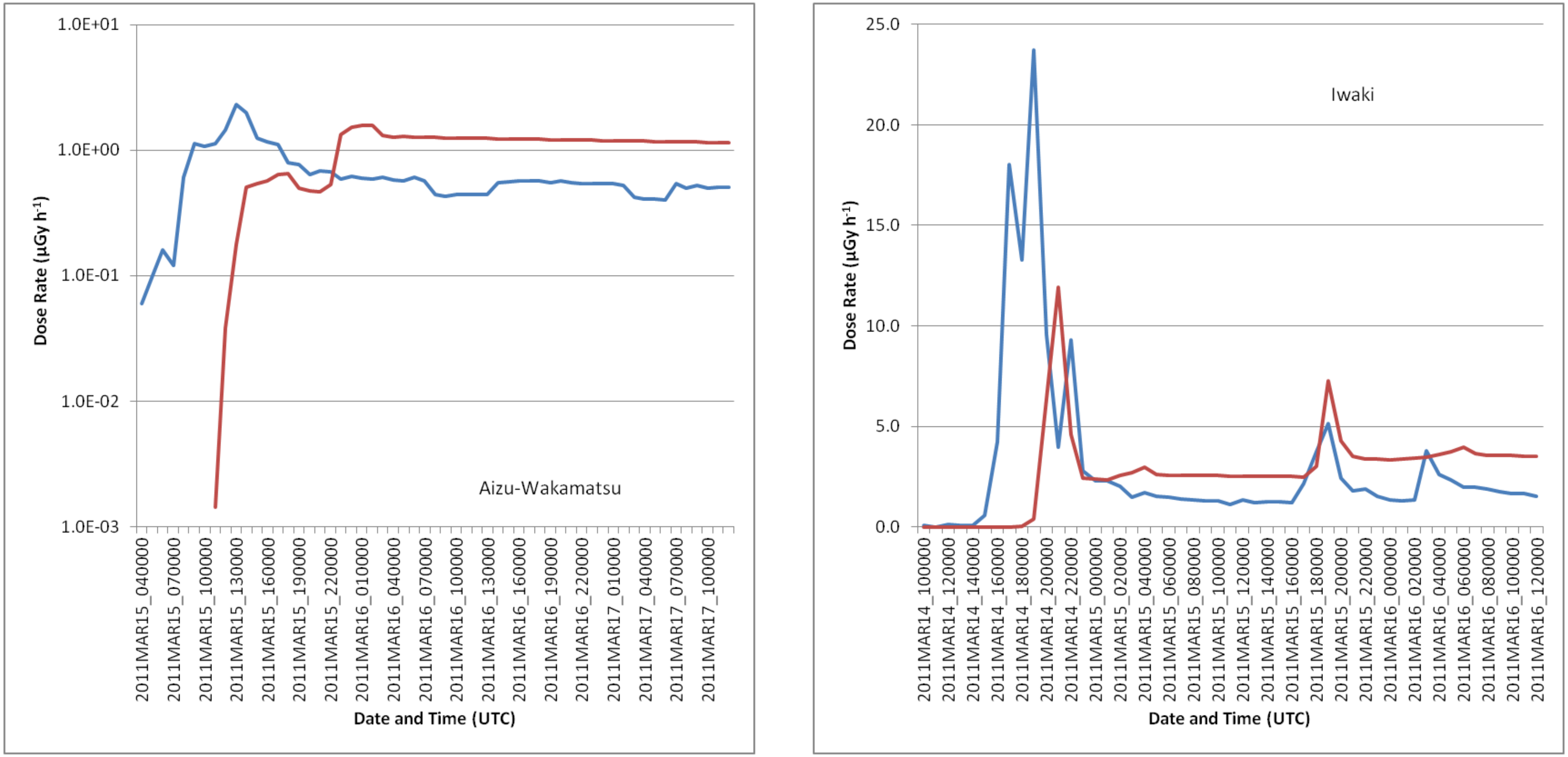
Figure



Figure

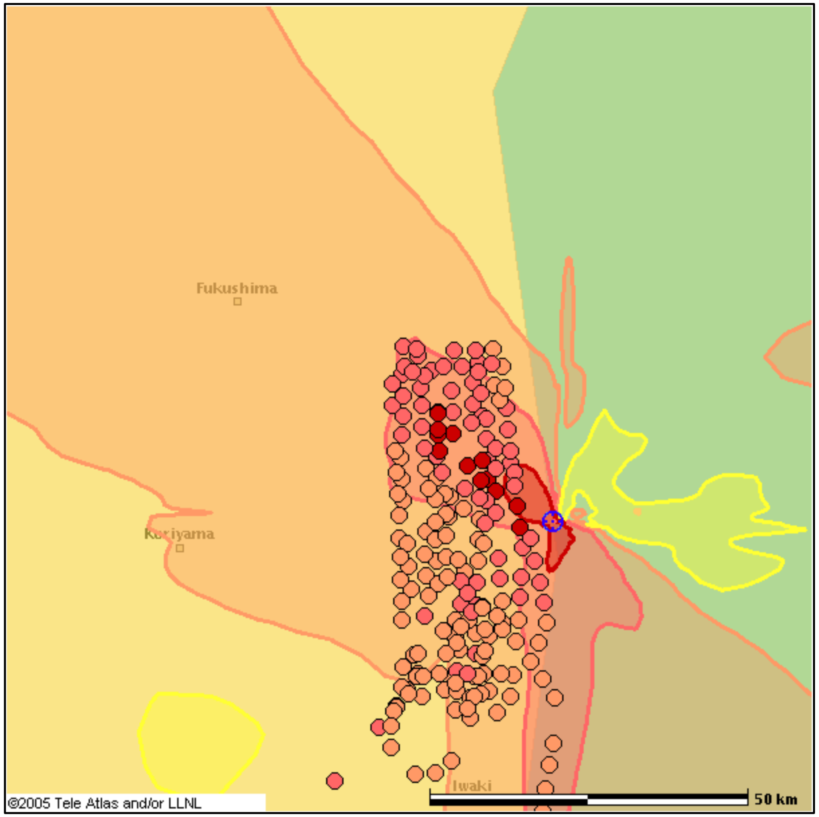


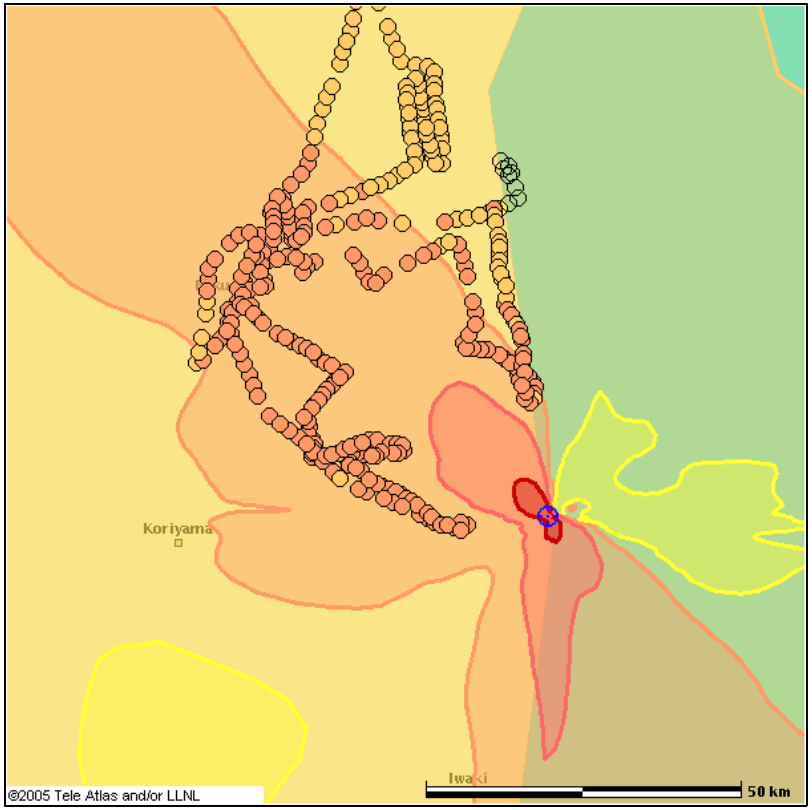
Figure



1.0E+01
1.0E+00
1.0E-01
1.0E-02
1.0E-03
Dose Rate (µGy h-1)
Aizu-Wakamatsu
2011MAR15_040000
2011MAR15_070000
2011MAR15_100000
2011MAR15_130000
2011MAR15_160000
2011MAR15_190000
2011MAR15_220000
2011MAR16_010000
2011MAR16_040000
2011MAR16_070000
2011MAR16_100000
2011MAR16_130000
2011MAR16_160000
2011MAR16_190000
2011MAR16_220000
2011MAR17_010000
2011MAR17_040000
2011MAR17_070000
2011MAR17_100000
Date and Time (UTC)
25.0
20.0
15.0
10.0
5.0
0.0
Dose Rate (µGy h-1)
Iwaki
2011MAR14_100000
2011MAR14_120000
2011MAR14_140000
2011MAR14_160000
2011MAR14_180000
2011MAR14_200000
2011MAR14_220000
2011MAR15_000000
2011MAR15_020000
2011MAR15_040000
2011MAR15_060000
2011MAR15_080000
2011MAR15_100000
2011MAR15_120000
2011MAR15_140000
2011MAR15_160000
2011MAR15_180000
2011MAR15_200000
2011MAR15_220000
2011MAR16_000000
2011MAR16_020000
2011MAR16_040000
2011MAR16_060000
2011MAR16_080000
2011MAR16_100000
2011MAR16_120000
Date and Time (UTC)

Figure


Fukushima
Koriyama
Iwaki
©2005 Tele Atlas and/or LLNL
50 km


Koriyama
Iwaki
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50 km