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ATMOSPHERIC DISPERSION MODELING:

CHALLENGES OF THE FUKUSHIMA DAI-ICHI RESPONSE

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ABSTRACT

The Department of Energy's (DOE) National Atmospheric Release Advisory Center (NARAC) provided a wide range of predictions and analyses as part of the response to the Fukushima Dai-ichi nuclear power plant accident 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 dose levels at U.S. locations
- Source estimation and plume model refinement based on atmospheric dispersion modeling and available monitoring data

This paper provides an overview of NARAC response activities, along with a more in-depth discussion of some of NARAC's preliminary source reconstruction analyses. NARAC optimized the overall agreement of model predictions to dose-rate measurements, using statistical comparisons of data and model values paired in space and time. Estimated emission rates varied depending on the choice of release assumptions (e.g., time-varying vs. constant release rates), the radionuclide mix, meteorology, and/or the radiological data used in the analysis. Results were found to be consistent with other studies within expected

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uncertainties, despite the application of different source estimation methodologies and the use of significantly different radiological measurement data. The paper concludes with a discussion of some of the operational and scientific challenges encountered during the response, along with 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 Administration (DOE/NNSA) Office of Emergency Response on 11 March 2011, to respond to events at the Fukushima Dai-ichi nuclear power plant. Although the reactors 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 throughout the crisis including weather forecasts, dose calculations for hypothetical scenarios to inform 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, particularly in the early stages of the response. During the first few days following the tsunami, only limited meteorological and radiological measurements were available. Subsequently larger volumes of data were received from Japanese weather and radiological monitoring stations, the DOE/NNSA 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 during the Fukushima response, and discusses some of the operational and scientific challenges encountered.

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). The center's products provide information on affected areas and populations, potential casualties, health effects and protective action guides, contamination levels, and damage zones to assist decision makers and responders in taking actions to protect the public, workers and the environment.

NARAC was created in 1979 during the Three Mile Island nuclear power plant accident. Since that time, the center has responded to other nuclear emergencies, including the 1986 Chernobyl nuclear reactor disaster and the 1999 nuclear fuel accident in Tokaimura, Japan. NARAC also provides capabilities to model the impacts of radiological dispersal devices, nuclear detonations, nuclear weapons accidents, and other radiological, chemical, biological, and natural releases.

NARAC is the atmospheric dispersion modeling center for DOE/NNSA emergency operations and one of the components of its 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 world-wide 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. NARAC'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 concentrations 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, NARAC 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 dose levels at U.S. locations
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Each of these efforts is described in more detail in the following sections.

Meteorological forecasting

NARAC provided regular meteorological forecasts to inform field operations and mission planning throughout the three-month period that DOE/NNSA monitoring teams were deployed in Japan. Forecast output was also distributed to the NRC, elements of the DOD, and other agencies. Initially, hourly forecasts for the next 24-to-48 hour period were produced three times each day due to rapidly changing meteorological conditions and mission planning needs. Later, the forecasting interval was reduced to once per day. 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). Wind fields from 5-km resolution WRF forecasts were used for routine operational support. Periodic consistency checks were made against independent NOAA forecasts and available Japanese meteorological data.

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.

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. 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 (DRL) factors to convert marker radionuclide concentrations to dose, 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.

Fig. 1 shows an example of one NARAC ADAPT/LODI model calculation of a NRC-provided hypothetical release scenario. The changing wind directions over the assumed 14 day release period resulted in the multi-lobe plume pattern seen in the figure. Although there was a high degree of uncertainty in the source terms, model predictions provided insight into areas that potential could be affected, 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 unit conditions evolved.

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) over multiple successive 12 or 24-hour release periods. Dose estimates were derived by scaling the modeled air and ground concentrations by the time-varying release rates in selected NRC release scenarios.

Fig. 2 shows four panels from an animation of one trans-Pacific NARAC 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 11-12 March would arrive on the West Coast on15-16 March. 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 surface detections or a substantial amount of ground 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) and ^{131}I concentrations to child thyroid dose exposures to determine if they exceeded the U.S. Environmental Protection Agency (EPA) / Food and Drug Administration (FDA) Protective Action Guide (PAG) levels (Sandia National Laboratories, 2010). These NARAC/CMHT dose estimates were relatively low-confidence predictions due to the uncertainties in both long-range global weather forecasting and emission scenarios. However, in all cases examined, the 96-hour TED dose projections were well below the EPA/FDA 0.01 Sv (1 rem) TED early phase evacuation/sheltering 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 0.05 Sv (5 rem) child thyroid PAG level even for the most 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. 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 limited due to the resources invested in, and priorities given to, some of the other activities described above.

During a response, NARAC typically provides an initial plume prediction to deploying field teams to assist in prioritizing areas for monitoring and sample collection. As aerial measurement survey and ground monitoring data are collected, they are electronically transferred to NARAC and/or downloaded from the DOE/NNSA CMHT quality-assured 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 typically performed using data and model results paired in both space and time (Foster et al. 2000). Below threshold measured and/or predicted values are not used in the comparisons, and outlier values may be removed as appropriate. The primary statistics used in the model-data comparisons are the percentage of predicted values that fall

within a factor, R, of the measured values (where R = 0.5, 2, 3, 5,7, 10, 20, 50, 100, and 1000) supplemented by a bias analysis (e.g., consideration of the relative magnitude and number of values over or under predicted). Statistics that use the ratio of measured and computed values are useful in comparing values that vary over many orders of magnitude, such as air concentration and ground deposition measurements. Additional statistical measures used in the analysis include the (absolute and signed) bias, the normalized mean square error, and the average and standard deviations of the ratios of measurement to calculated value. The predicted spatial and temporal concentration patterns also are compared to spatial plots of the data and time series plots at each measurement location (example of these comparisons are shown in some of the figures in this paper). Input assumptions (e.g., release rates, release heights, activity and particle size distributions, meteorological data) are then varied to find the best fit to these data and the average measured-to-predicted value ratio is used to scale the release amounts to best match the measurements.

Source estimation is almost always an under-constrained non-linear optimization problem, which requires taking into account meteorology, geography, source characteristics (e.g., emission rates, radionuclide mix, release height, particle size distribution), and dry and wet deposition processes. Reconstruction of the Fukushima Dai-ichi releases posed a uniquely complicated challenge due to rapidly changing winds and precipitation conditions, complex terrain, land-sea interfaces, long-running time-varying sources, and multiple potential reactor and spent fuel pool releases. Items that needed to be addressed 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
- 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 radionuclide 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 options, NARAC used a range of meteorological simulations generated by 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 11 March, shifting to on-shore northward flow on 12 March , back to off-shore flow on 13 March, followed by a clockwise rotation pushing plumes first to the south (14 – 15March), then west, northwest, and north (15 March), and off-shore again on 16 March. Winds remained primarily off-shore until 21 March when the wind direction again sent radioactive material southward in the general direction of Tokyo. Initial NARAC forecasts captured the overall pattern of wind directions and the occurrence of precipitation, with subsequent higher resolution forecasting providing increased accuracy in modeling the timing of the wind shifts and precipitation patterns. NARAC primarily used WRF 3-km FDDA simulated wind fields in its source reconstruction analyses as WRF fields at 1 km grid resolution were not found to result in significant differences in the dispersion patterns of interest. It should be noted that this does not necessarily imply

that 1 km resolution forecasts do not provide more accurate *meteorological* fields, only that use of such data did not affect NARAC's initial source reconstruction analyses. However, additional studies are needed before the benefits of higher-resolution weather information can be completely assessed.

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 14 March resulting in southward winds at the Fukushima nuclear power plant. A weak low-pressure system then moved across central Japan on 15 March 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 15 March 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 environmental radiological data were correlated with prevailing wind directions. Based on this preliminary analysis, NARAC focused its model refinement efforts on 14 – 16 March, 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 21 – 23 March 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 – 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 around 21 March.

Precipitation. Precipitation occurred sporadically throughout the Fukushima releases and was found 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 from 15 March 0800 UTC to 15 March 1900 UTC in northern Fukushima prefecture and from 20 March 2300 UTC to 22 March 2100 UTC in Ibaraki, Chiba, Tochigi, and Saitama prefectures and Tokyo.

NARAC used both uniform grid-wide precipitation based on Japanese meteorological observations, and spatially varying precipitation fields derived from NARAC's 3-km-resolution WRF model simulations, in its source reconstruction analyses. As illustrated in Fig. 4, WRF-generated simulations captured the approximate timing and location of precipitation, although not all of the details of the rainfall patterns. In looking at Fig 4, it should be noted that the measurements of precipitation rate were only reported to the nearest 1 mm h^{-1} (e.g., 0, 1, 2, or 3 mm h^{-1}), which limits the accuracy of comparisons especially for rates less than 1 mm h^{-1}. Comparisons of time series of measured and WRF-modeled precipitation rates (not shown) show good agreement for stations located near Tokyo and Fukushima City.

Spatially and temporally varying precipitation and associated scavenging due to both in-cloud and below-cloud processes can 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 (apart from precipitation). 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 and Colton 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/NNSA Aerial Measuring System (AMS)
- DOE and DoD monitoring data provided via the DOE/CMHT electronic radiological database

Unfortunately, several key time gaps existed in the TEPCO data due to the failure of plant monitoring stations during the earthquake and/or tsunami and a site evacuation that occurred on 15 March (GOJ 2011c). MEXT regional prefectural monitoring station data were available only for the period following 15 March 0900 UTC, although a few Fukushima prefecture locations reported data from earlier time periods.

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 for modeling purposes. The data were also reviewed to identify inconsistencies and gaps, exclude unrepresentative data, take background into account, and make sure data were properly

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

interpreted for comparisons against NARAC model results. Analysis of additional data still needs to be undertaken in order to further refine existing source term estimates.

Preliminary analysis of the MEXT data showed progression of the plume over the 14-16 March 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 20 March, a hypothesis 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 primarily focused its calculations on the radionuclides ^{133}Xe, ^{131}I, ^{137}Cs, ^{134}Cs, using assumed relative activity ratios of 100:10:1:1 for ^{133}Xe:^{131}I:^{137}Cs:^{134}Cs , although some variants on these values were explored. Later, ^{132}I and ^{132}Te were added to the mix because of their potentially significant contributions to dose within the first few days following their release. In the examples discussed in this paper, ^{133}Xe:^{131}I: ^{132}I: ^{132}Te:^{137}Cs:^{134}Cs relative activity ratios of 100:20:20:20:1:1 and 100:10:10:10:1:1 were used, as described below.

The relative activity ratios were derived from DOE/NNSA analyses of spectra from *in situ* measurements[‡] and NRC reactor scenario radionuclide activity ratios. The average ^{134}Cs:^{137}Cs activity ratio of approximately 1:1 is consistent with a wide range of DOE/NNSA *in situ* spectra analyses, although there is considerable variation between individual values (Musolino et al. 2012). The ^{131}I:^{137}Cs activity ratios used are 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 ^{137}Cs: ^{131}I ratios vary over 2 orders of magnitude (Table 2 in Chino et al. 2011).

Source term estimates. NARAC conducted a number of source reconstruction analyses using a range of possible release assumptions and meteorological conditions. As time constraints during the response prevented a comprehensive and systematic study, this paper only discusses several illustrative analysis examples that provide insight into the range of possible source terms consistent with the available data. In

[‡] Wimer, N., Lawrence Livermore National Laboratory, Livermore, CA; 2011 (private communication).

all of the 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, radionuclide mixes, 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 during the entire modeled period. The NARAC analyses used Japanese MEXT dose rate and/or AMS ground-shine dose rate measurements (Lyons and Colton 2012). The MEXT dose rate measurements were assumed to include contributions from both "cloud-shine" (air immersion) and "ground-shine" (ground exposure). 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 application of a ground roughness shielding factor of 0.82 to the predicted ground exposure dose (Likhtarev et al. 20 02).

A number of NARAC source reconstruction analyses were conducted using WRF FDDA 3-km resolution simulations, which were believed to provide the most accurate meteorology. One such analysis was performed using ADAPT/LODI simulations of ^{137}Cs, ^{134}Cs, ^{131}I, ^{132}I, ^{132}Te, and ^{133}Xe in the relative activity ratios of 1:1:20:20:20:100 over the critical period from 14 March 0600 UTC to 16 March 0600 UTC. For the purposes of this paper, this analysis will be referred to as the "NARAC baseline" case. NARAC estimated the source term for this case using 451 hourly dose rate measurements at 22 MEXT stations from Fukushima and the surrounding prefectures, although as discussed above the majority of these data were from the period after 15 March 0900 UTC. Assuming a constant release rate over the entire 48-hour period, the model fit to data resulted in total release quantities of 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 (Table 1).

Fig. 6 shows a comparison of NARAC baseline case results to MEXT dose rates for a representative time period on 15 March 1800 UTC. Overall, 20% of the baseline case predicted values were within a factor of 2 of the MEXT measurements in Fukushima prefecture (i.e., the ratios of measured and predicted values for the same time and location were between 0.5 and 2), 49% were within a factor of five, and 71% within a factor of 10. For MEXT stations in other prefectures, more than 51% of model predicted values were within a factor of 2 of measured dose rates, 75% within a factor of five, and 91% within a factor of 10. NARAC model-predicted values were found to provide similar agreement to AMS data (34% within a factor of 2, 78% with a factor of 5, and 88% within a factor of 10), even though these data were not used in the NARAC source estimation process.

Predictions using time-varying release rates can better capture some of the time variations in the MEXT dose rate data time series, although this does not necessarily improve the overall statistical agreement with data. One such analysis was performed for an assumed release period from 12 March 0100 UTC to 18 March 1400 UTC (although 90% of the activity is released from 14 March 1500 UTC to 15 March 0700 UTC). This analysis was based on meteorology developed from Japanese weather observations and used a uniform grid-wide precipitation rate of 2 mm h^{-1} on 15 March. The total release over the simulation period was 3.7×10^{15} Bq (9.9×10^{4} Ci) each of ^{137}Cs and ^{134}Cs, 3.7×10^{16} Bq (9.9×10^{5} Ci) each of ^{131}I. ^{132}I, and ^{132}Te, and 3.7×10^{17} Bq (9.9×10^{6} Ci) of ^{133}Xe.

Fig. 7 compares time series of NARAC ADAPT/LODI predicted values from this second example analysis against hourly gamma dose-rate data for two locations – Aizu-wakamatsu and Iwaki (GOJ 2011d), which are located approximately 30 km west and 100 km south of Fukushima Dai-ichi, respectively. For all 860 MEXT measurements in Fukushima prefecture, more than 35% of the model predicted values were within a factor of two of the data, 84% within a factor of five, and 90% within a factor of 10, which is an improvement over the baseline case statistics. For the 770 measurements from the MEXT stations in other prefectures, 32% were within a factor of two of predicted values, 72% within a factor of five, and 92% within a factor of 10.

Fig. 8 shows results from a third representative analysis for the period from March 15 0300 UTC to March 16 0200 UTC that was based on the Aerial Measuring System (AMS) data alone. The NARAC ADAPT/LODI simulation shown in the figure used time-dependent releases of ^{137}Cs, ^{134}Cs, ^{131}I, ^{132}I, and ^{132}Te with an assumed relative activity ratio of 1:1:20:20:20, meteorological fields derived from observational data and a uniform grid-wide precipitation of 2 mm h^{-1} over the entire simulation period. The model fit to data resulted in estimates of total release quantities of 5.6×10^{15} Bq (1.5×10^{5} Ci) each of ^{137}Cs and ^{134}Cs and 1.1×10^{17} Bq (3×10^{6} Ci) each of ^{131}I, ^{132}I, and ^{132}Te. ^{133}Xe was not included in this analysis as it does not contribute to the ground shine dose rate measured by the AMS.

The left panel of Fig. 8 shows a comparison of LODI model predicted dose rates (color-filled contours) to the 18 March 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. For the 1959 points in this dataset, 43% of predicted values were within a factor of two of AMS measurement values, 84% within a factor of five and 94% within a factor of 10.

The 26 March comparison shown in the right side panel of Figure 8 provides a confirmation of the original source reconstruction, as these data were not used in developing the source estimate. The measurements reflect the effects of radioactive decay over the intervening eight days and show no signs of significant additional deposition in the period after 18 March. In this case, for the 1717 data points, more than 64% of the model predicted values agreed with the AMS data within a factor of two, 97% within a factor of five, and 98% within a factor of 10. There was very little overall bias in the model predictions, with the model over-predicting 1906 of all the AMS measurement values and under-predicting 1770 values.

In general, AMS-based NARAC source reconstruction analyses led to somewhat higher release rate estimates than those based on MEXT data. This may be due to one or more of the following factors. NARAC analyses primarily focused on AMS data derived from the aerial surveys that measured the highest deposition area extending to the northwest of the Fukushima Dai-ichi site (Lyons and Colton 2012). Plume predictions for this period have a higher degree of uncertainty due to the complexity and

importance of precipitation scavenging, which caused much of the deposition in this region. The AMS data also needed to be corrected to account for terrain elevation and for the contributions of any airborne plume to the measurements. However, the model agreement with the AMS data during both periods provides confidence that the analyses are capturing key features of the meteorology, release, dispersion, and deposition.

NARAC source reconstruction analyses showed that a range of emission rates were consistent with the available dose-rate data within model and measurement uncertainties. NARAC estimates of release quantities varied within a factor of three from the baseline case for the same radionuclide mix. Source term estimates based on measurement-model comparisons were sensitive to source term input assumptions (e.g., time-varying vs. constant emission rates, the radionuclide mix and activity ratios, other release characteristics, reactor conditions), the choice of meteorology, and the selection of the radiological data (e.g., AMS, MEXT) to preferentially match in the model refinement process. 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

Table 1 summarizes activity release estimates from the NARAC baseline case along with several recent studies, which utilized different computer models, measurement data, and source estimation techniques. 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 12 March 0100 UTC to 5 April 1500 UTC (12 March 1000 JST to 6 April 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. Table 1 includes the most recently published values (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 (12 March – 5 April) 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 derived from 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 includes the Stohl et al. 2011 release estimate of 3.6×10^{16} Bq of ^{137}Cs for 11 March to 20 April, which had an estimated uncertainty range of 2.33×10^{16} to 5.01×10^{16} Bq. 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 16 March.

The DOE/NNSA CMHT used AMS ground-shine dose rate data collected out to 80 km from the plant during April 6–29 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 (e.g., non-deposited) material. Assuming that 19% of the released ^{137}Cs was deposited on land (as estimated by Stohl et al 2011), the AMS-based analysis leads to an estimated release of approximately1.4×10^{16} Bq of ^{137}Cs.

Table 2 compares the NARAC baseline case activity release estimate for the period from 0600 UTC on 14 March 2011 to 0600 UTC on 16 March 2011 to estimates of the release rate for the same period derived from several of the previous studies discussed above. NARAC calculated the latter values using time-varying release rates provided in the cited papers. The two-day period compared is estimated to cover approximately 25-50% of the total release based on the references in Table 2.

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

It is both interesting and encouraging to note that all of the source term estimates in Table 2 apart from ^{133}Xe are within a factor of approximately six (and most are within a factor of three) despite the different source reconstruction methodologies, meteorological models, types of radiological data, and reactor conditions assumed. Without further analysis, as discussed below, it is difficult to determine which 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. Furthermore, although the time frame in Table 2 covers the primary period of on-shore transport of radioactivity, 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, requiring the use of sparser longer-range measurements and model calculations.

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 in great demand as it was critical to developing and quality assuring new non-standard and/or complex analyses required to answer key unanticipated questions.
- More efficient means were needed to analyze complex (multiple reactor unit and spent fuel pool) nuclear power plant scenarios, 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
- Construct 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 items above, 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 enhanced interagency communications. Such efforts are leading to improvements in DOE/NNSA and NARAC's ability to respond to a future Fukushima-scale 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 currently 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 (Stohl et al. 2011 make a similar recommendation). 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
- Investigation of the use of ensemble forecasts to develop probabilistic arrival times and impact estimates for both regional (e.g. Japan) and long-range (e.g., trans-Pacific) cases
- 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), and U.S. nuclear power plant data, and comparison of source estimation based on these data to values derived using Japan-based measurement data

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 capabilities for responding to future incidents.

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REFERENCES

Bowyer RW, Biegalski SR, Cooper M, Eslinger PW, Haas D, Hayes JC, Miley HS, Strom DJ, Woods V. Elevated radioxenon detected remotely following the Fukushima nuclear accident. J. Environ. Radioactivity 102:681-687; 2011.

Chino M, Nakayama H, Nagai H, Terada H, Katata G, Yamazawa H. Preliminary Estimation of Release Amounts of I-131 and Cs-137 Accidentally Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere. J. Nuc. Sci and Tech. 48: 1129-1134; 2011.

Eckerman KF, Leggett RW. User Guide to DCFPAK 2.2. Oak Ridge National Laboratory, Oak Ridge, TN; 2008.

Environmental Modeling Center. The GFS Atmospheric Model. NCEP Office Note 442, Global Climate and Weather Modeling Branch, EMC, Camp Springs, Maryland, 2003. Available at: http://nws.noaa.gov/ost/climate/STIP/AGFS_DOC_1103.pdf.

Environmental Protection Agency (EPA). Japanese Nuclear Emergency: Radiation Monitoring; 2011. Available at http://www.epa.gov/japan011/rert/radnet-data.html

Fast J, Easter R. A Lagrangian particle dispersion model compatible with WRF. 7th Annual WRF User's Workshop, 19-22 June 2006, Boulder, CO; 2006. Available at: http://www.mmm.ucar.edu/wrf/users/workshops/WS2006/abstracts/PSession06/P6_02_Fast.pdf

Foster KT, Sugiyama G, Nasstrom JS, Leone, Jr. JM, Chan ST, Bowen BM. The Use of an Operational Model Evaluation System for Model Intercomparison. Int. J. Environment and Pollution 14: 77-88; 2000.

Government of Japan. Ministry of Economy Trade and Industry News Release: INES (the International Nuclear and Radiological Event Scale) Rating on the Events in the Fukushima Dai-ichi Nuclear Power Station by the Tohoku District – off the Pacific Ocean Earthquake, 12 April, 2011; 2011a. Available at: http://www.nisa.meti.go.jp/english/files/en20110412-4.pdf

Government of Japan. Report of Japanese Government to IAEA Ministerial Conference on Nuclear Safety – The Accident at TEPCO's Fukushima Nuclear Power Stations. Transmitted by Permanent Mission of Japan to IAEA, 7 June 2011; 2011b. Available at: http://www.iaea.org/newscenter/focus/fukushima/japan-report/

Government of Japan. Additional Report of the Japanese Government to the IAEA – Accident at TEPCO's Fukushima Nuclear Power Stations. Transmitted by Nuclear Emergency Response Headquarters, Govermnment of Japan, 15 September 2011; 2011c. Available at: http://www.iaea.org/newscenter/focus/fukushima/japan-report2/

Government of Japan. Ministry of Education, Culture, Sports, Science & Technology in Japan (MEXT). Reading of environmental radioactivity level (English version); 2011d. Available at: http://www.mext.go.jp/english/incident/1303962.htm

Kinoshitaa N, Suekia K, Sasaa K, Kitagawaa J, Ikarashia S, Nishimuraa T, Wonga Y-S, Satoua Y, Handaa K, Takahashia T, Satob M, Yamagatab T. Assessment of individual radionuclide distributions from the Fukushima nuclear accident covering central-east Japan. Proc Natl Acad Sci USA 108:19526–19529; 2011.

Japan Weather Association (JWA): Meteorology Data Service; 2011. Available at: http://tenki.jp.

Likhtarev IA, Kovgan LN, Jacob P, Anspaugh LR. Chernobyl Accident: Retrospective and Prospective Estimates of External Dose of the Population of Ukraine. Health Physics 82(3): 290 -303; 2002.

Liu Y, Bourgeois A, Warner T, Swerdlin S, Hacker J. An implementation of obs-nudging-based FDDA into WRF for supporting ATEC test operations. 2005 WRF user workshop paper 10.7; 2005. Available at http://www.rap.ucar.edu/projects/armyrange/references/publications/Liu_WRF-WK.2005.pdf

Lyons C, Colton D. Aerial Measuring System in Japan, Health Physics; 2012 (in this issue)

Morino Y, Ohara T, Nishizawa M. Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant. Geophys Res Lett 38, L00G11, doi:10.1029/2011GL048689, 2011

Musolino S, Clark H, McCullough T, Pemberton W, Wimer N. Environmental measurements in an emergency: this is not a drill. Health Physics; 2012 (in this issue)

Nasstrom JS, Sugiyama G, Baskett RL, Larsen SC, Bradley MM. The NARAC modeling and decision support system for radiological and nuclear emergency preparedness and response. Int. J. Emergency Management 4:524-550; 2007.

Sandia National Laboratories, *FRMAC Assessment Manual Volume 1, Overview of Methods*. SAND2010-1405P, Sandia National Laboratories, Albuquerque, NM; 2010.

Skamarock WC, Klemp JB , Dudhia J, Gill DO, Barker DM, Duda MG, Huang X-Y., Wang W, Powers JG. A Description of the Advanced Research WRF Version 3. NCAR Technical Note, NCAR/TN–

475+STR, National Center for Atmospheric Research, Boulder CO; 2008. Available at: http://www.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf

Stauffer DR, Seaman NL. On Multi-Scale Four-Dimensional Data Assimilation. J. Appl. Meteor. 33:416-434; 1994.

Stohl A, Forster C, Frank A, Seibert P, Wotawa G. The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys. 5:2461–2474, doi:10.5194/acp-5-2461-2005; 2005.

Stohl A., Seibert P, Wotawa G, Arnold D, Burkhardt JF, Eckhardt S, Tapia C, Vargas A, Yasunari T. Xenon-133 and caesium-137 releases into the atmospheric from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition. Atmos. Chem. Phys. Discuss. 11:28319–28394; 2011. Available at www.atmos-chem-phys-discuss.net/11/28319/2011/doi:10.5194/acpd-11-28319-2011

Sugiyama G, Nasstrom JS, Baskett R, Simpson M. National Atmospheric Release Advisory Center (NARAC) Capabilities for Homeland Security. Proceedings of the 5th International Symposium on Computational Wind Engineering, Chapel Hill, NC May 23-27, 2010; 2010. Available at narac.llnl.gov/documents.html

US DOE/NNSA. Response to 2011 Fukushima Incident- Raw Aerial Data and Extracted Ground Exposure Rates and Cesium Deposition; 2011. Available at http://explore.data.gov

Takemura T, Nakamura H, Takigawa M, Kondo H, Satomura T, Miyasaka T, Nakajima T. A numerical simulation of global transport of atmospheric particles emitted from the Fukushima Daiichi Nuclear Power Plant. SOLA 7:101–104, doi:10.2151/ sola. 2011-026; 2011.

Yasunari J, Andreas S, Hayano RS, Burkhart JF, Eckhardt S, Yasunari T. Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. *PNAS* 108:19530-19534; 2011. Available at www.pnas.org/cgi/doi/10.1073/pnas.1112058108

Table 1. Released radioactivity estimates (and release dates, in UTC) from previously published studies, and NARAC baseline release estimate from this study.

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	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	3.7×10^{15} Bq
^{134}Cs	1.8×10^{16} Bq	–	–	–	–	1.4×10^{16} Bq	3.7×10^{15} Bq
^{131}I	1.6×10^{17} Bq	1.5×10^{17} Bq	1.3×10^{17} Bq	–	–	–	7.4×10^{16} Bq
^{133}Xe	–	–	–	1.2×10^{19} Bq	1.7×10^{19} Bq (March 11-15)	–	3.7×10^{17} Bq

Table 2. This table compares the NARAC baseline case released activity estimate, for the release period from 14 March 2011 0600 UTC to 16 March 2011 0600 UTC, to estimated values from previously published studies. The release quantities were estimated from tables and figures covering longer periods in the cited references (except for the GOJ 2011c estimate for which the published data ends on 15:00 UTC March 15). When available, the estimated percentage of the total released activity is given.

Radionuclide	JAEA (Chino et al. 2011)	JAEA (GOJ 2011c)	Stohl et al. 2011	NARAC Baseline
^{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%	3.7×10^{15} Bq (1×10^{5} Ci)
^{134}Cs	–	–	–	3.7×10^{15} Bq (1×10^{5} Ci)
^{131}I	6.87×10^{16} Bq (1.86×10^{6} Ci) 45%	2.94×10^{16} Bq (7.96×10^{5} Ci) 23%	–	7.4×10^{16} Bq (2×10^{6} Ci)
^{133}Xe	–	–	5.68×10^{18} Bq (1.54×10^{8} Ci) 34%	3.7×10^{17} Bq (1×10^{7} Ci)

FIGURES

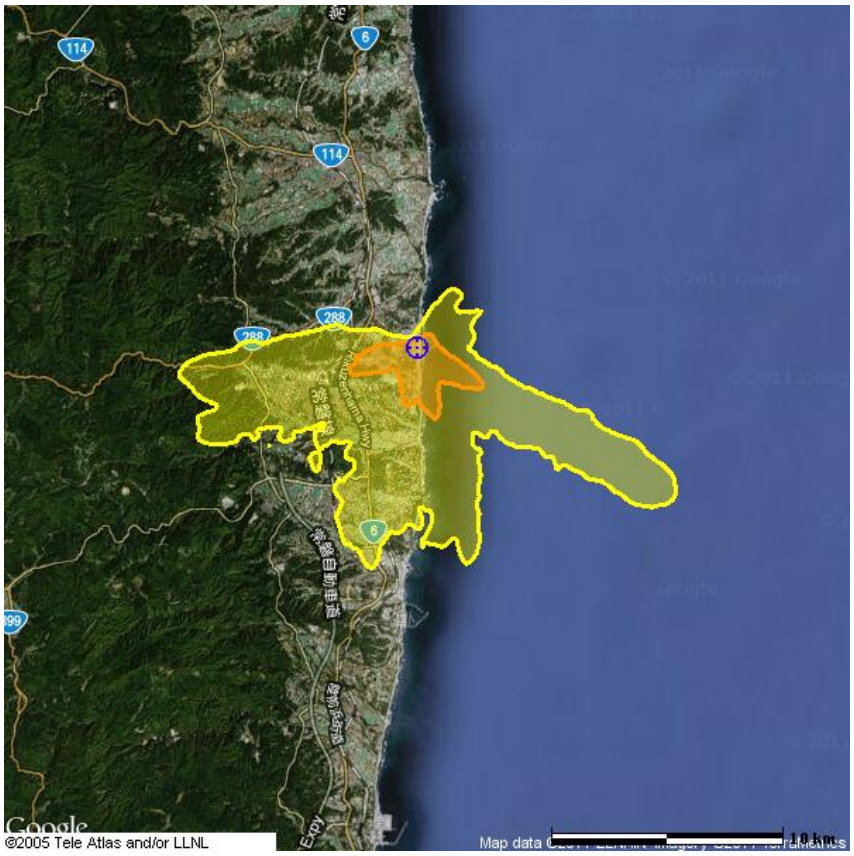

Fig. 1. This figure shows the results of a NARAC ADAPT/LODI simulation of Total Effective Dose (TED) over a 14-day period from 12 March to 26 March 2011 for a hypothetical reactor release scenario provided by the Nuclear Regulatory Commission. The orange and yellow color-filled contours show the areas where the dose is predicted to exceed 0.05 Sv (5 rem) and 0.01 Sv (1 rem), respectively. The top 20 radionuclide contributors to dose and WRF-generated meteorological fields were used in this simulation.

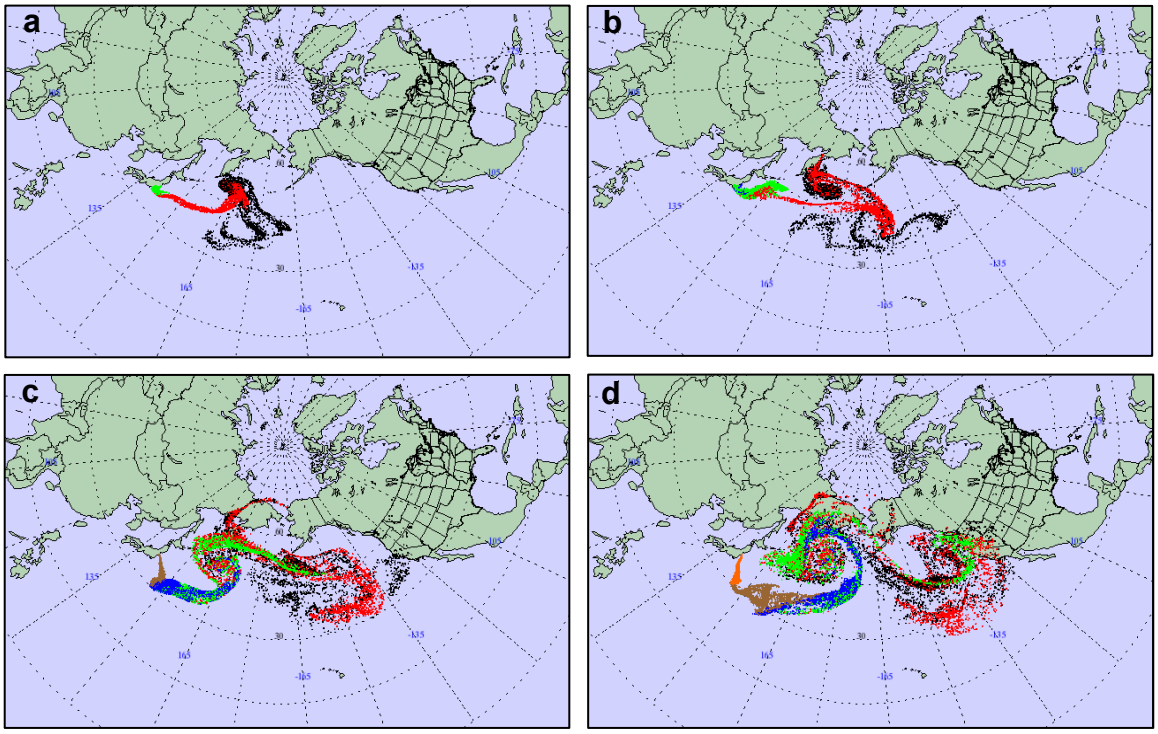

Fig 2. The panels show four frames from an animation of the trans-Pacific transport and dispersion of marker particles from a LODI model simulation of hypothetical releases from the Fukushima Dai-ichi plant. Particle locations are shown for four times: (a) 00:30 UTC 15 March 2011, (b) 00:30 UTC 16 March 2011, (c) 00:30 UTC 17 March 2011 and (d) 00:30 UTC 18 March 2011. Particles of the same color were released during the same 24-hour interval.

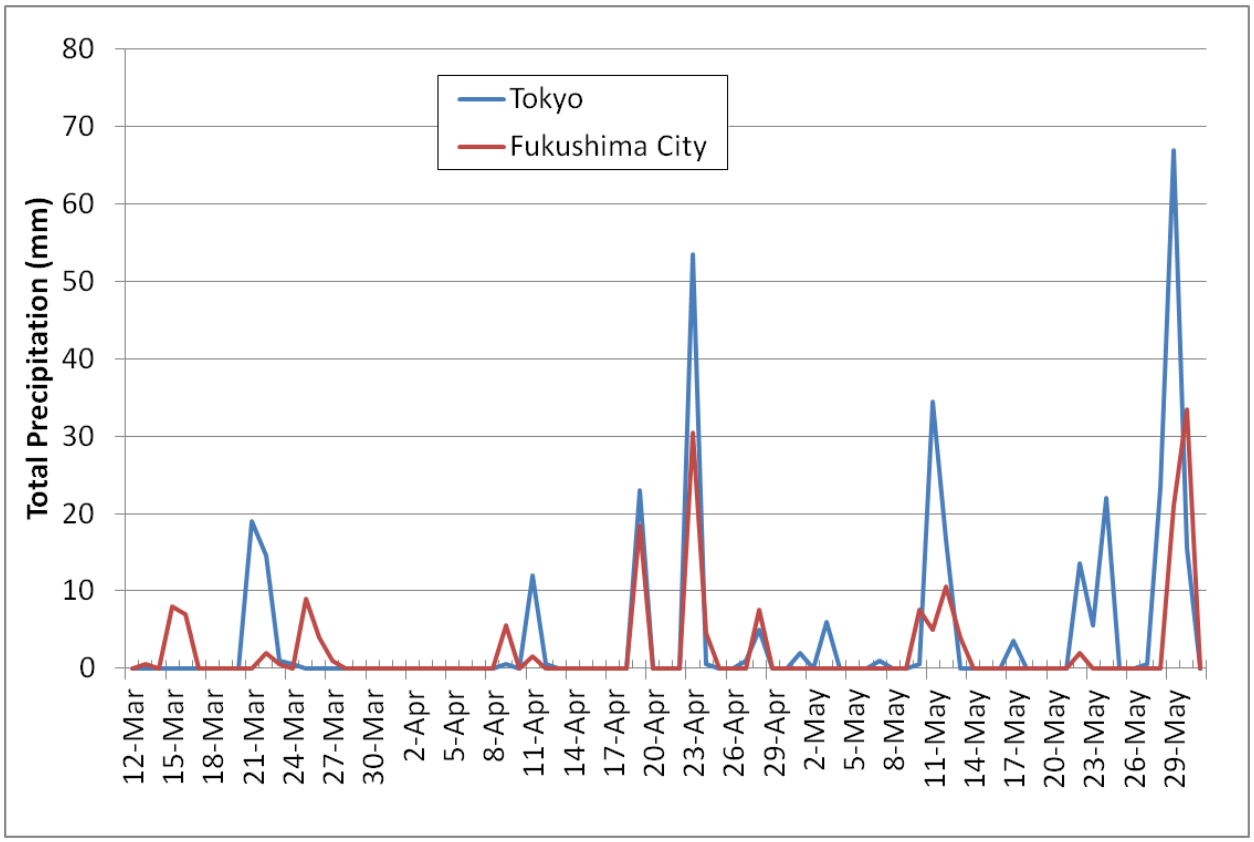

Fig 3. This figure plots observed daily precipitation at Tokyo (blue) and Fukushima City (red) from 12 March to 31 May 2011 JST (data obtained from JWA 2011).

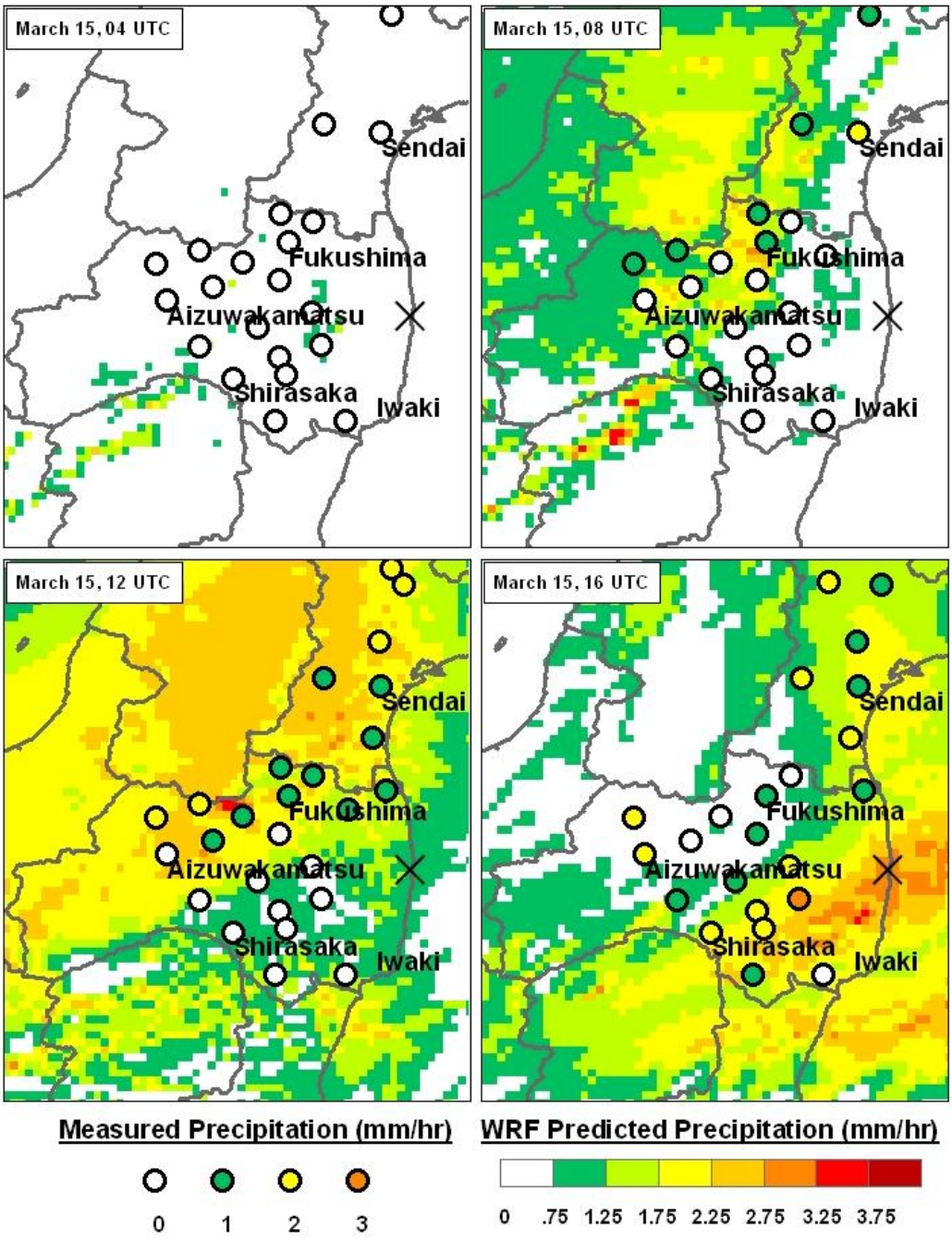

Fig 4. The panels show comparisons of 3-km resolution WRF-modeled precipitation rates (square color pixels) to Japan Meteorological Agency (JMA) station observations (color coded circles) at four different times. The observed precipitation rates were provided courtesy of the Japan Atomic Energy Agency (JAEA) and were reported to the nearest 1 mm h^{-1}. The cross shows the location of the Fukushima Dai-ichi plant. City names are listed just below and to the right of their locations.

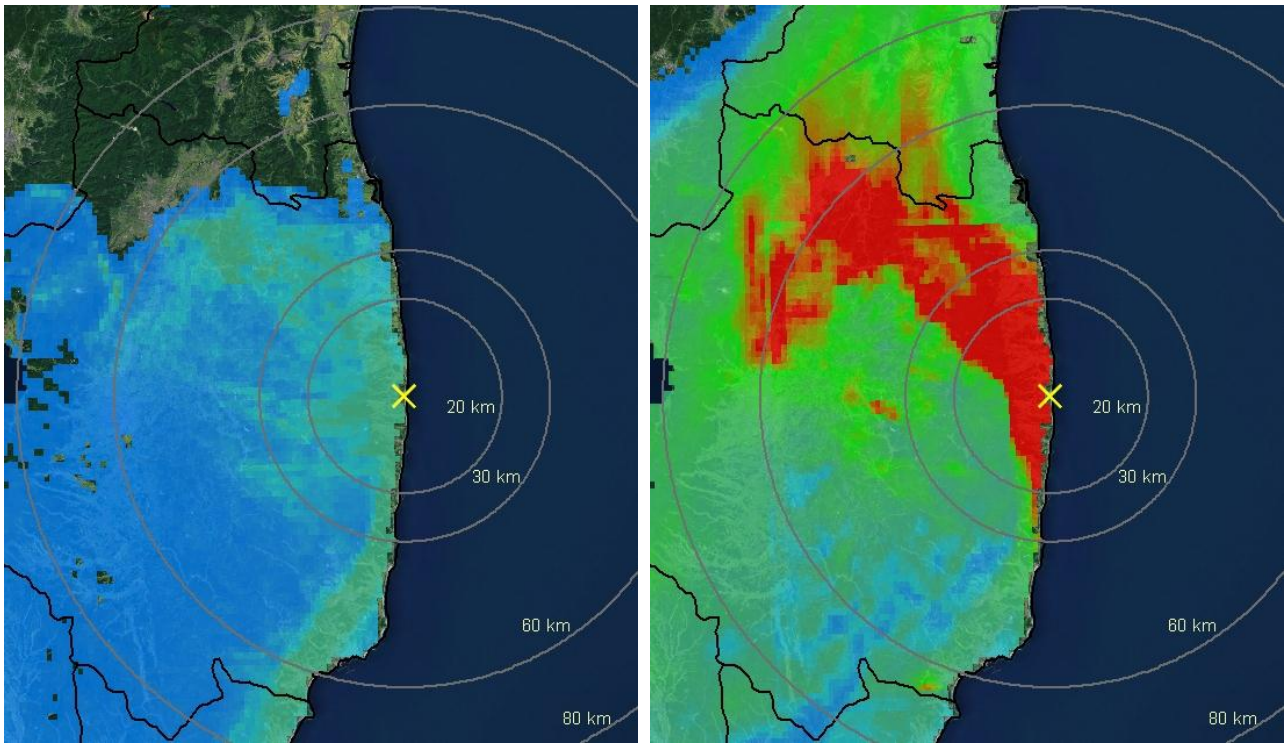

Fig 5. The two panels compare the relative deposition resulting from a FLEXPART simulation of a constant release rate of a normalized amount of material over the period for 14 March 1000 UTC to 15 March 1800Z without precipitation scavenging (left panel) and with precipitation scavenging (right panel). The simulation used 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.

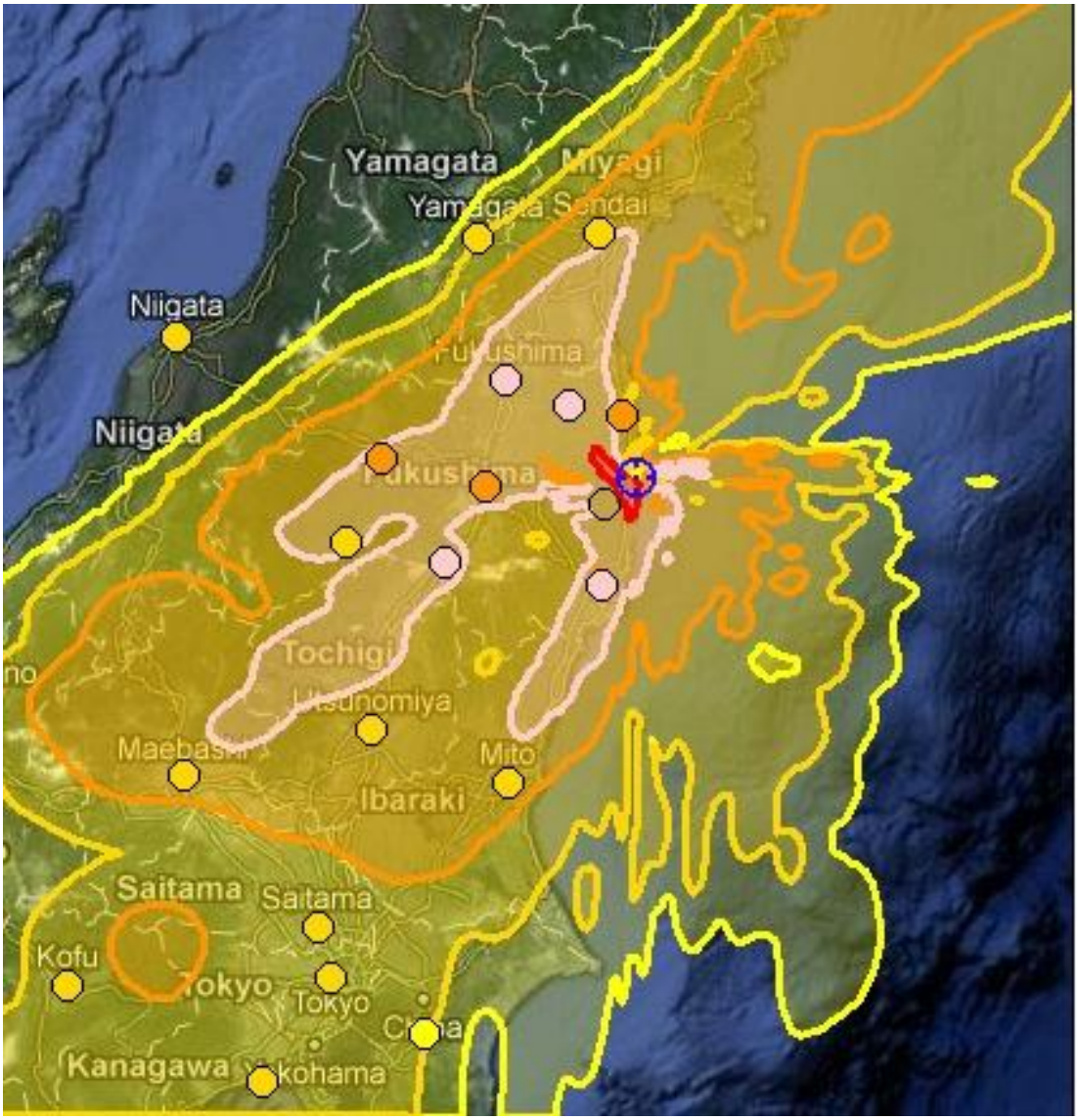

Fig 6. Dose rate results from the NARAC-modeled baseline case (color-filled contours) are compared with MEXT data (circles color coded to the same levels as the contours) for 15 March 1800 UTC. The contour levels were selected to best show the comparison to data. The innermost red contour is the area where the model predicts that 120 µGy/h (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 Dai-ichi plant. (Background map courtesy of Google)

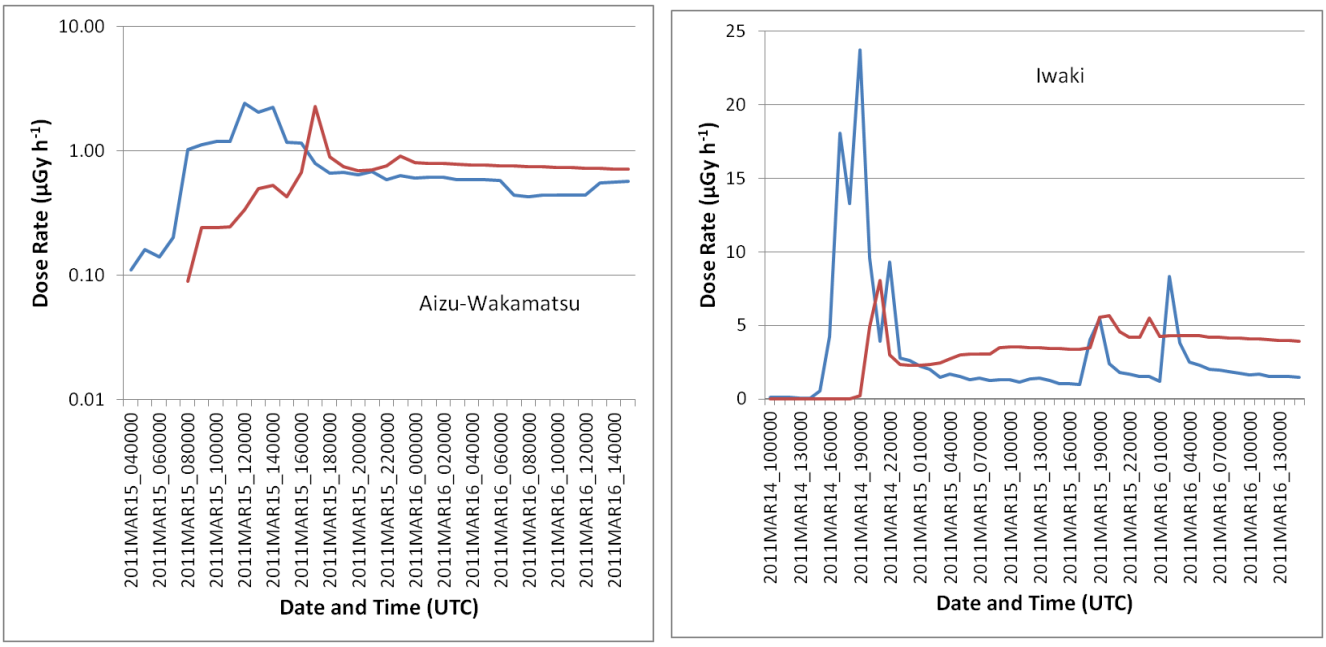

Fig 7. Comparisons of NARAC-predicted dose rates based on a time-varying source estimate (red) and MEXT measured dose rates (blue) are shown for two locations – Aizu-wakamatsu (left panel) and Iwaki (right panel). Different time periods are shown for the two locations which do not cover the entire time frame of the simulation. The measurement data were below threshold for the periods prior to the starting times shown in the panel.

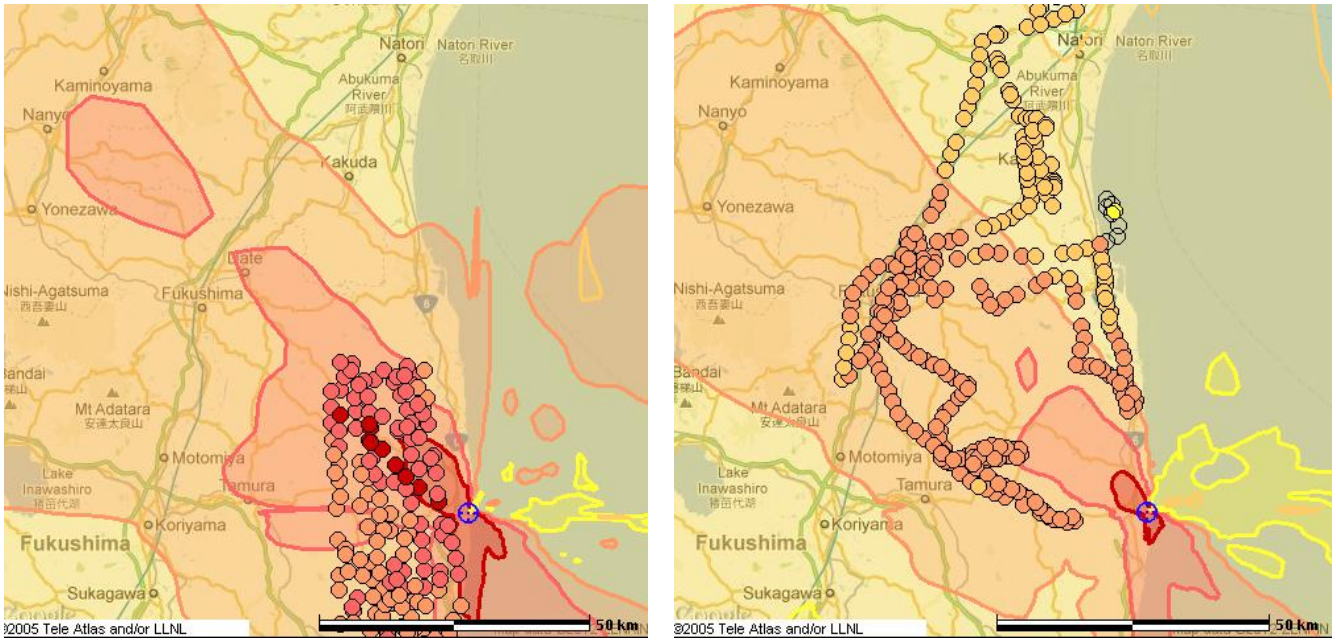

Fig 8. The two panels show the results of a NARAC analysis based on a time-varying release developed from AMS data from 18 March 2011. LODI model predicted dose rates (color-filled contours) are compared with AMS data (circles colored coded in the same manner as the contours) at two different times – 18 March (left panel) and 26 March (right panel). The innermost dark red, red, dark orange, orange, and outermost yellow contours 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.