DETECTION OF RADIONUCLIDES EMITTED DURING THE FUKUSHIMA NUCLEAR ACCIDENT WITH THE CTBT RADIONUCLIDE NETWORK

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ABSTRACT

Radioactivity is monitored on a global scale by the International Monitoring system (IMS) radionuclide network, which is being built for the Verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). This network consists of 80 particulate stations. Forty of them will also be equipped with complementary noble gas monitoring systems. Sensitivity, number, and location of the particulate and noble gas stations are defined to ensure a 90 % detection probability of a release from a 1 kt nuclear explosion in the atmosphere after 10 days. As of March 2011, 60 particulate stations have been certified and 27 noble gas stations have been installed.

Following the accident of the Fukushima Dai-ichi nuclear power plant radioactive emission to the atmosphere started on 12 March. Radioactive particles and gases were transported across the Pacific towards North America. The first detection in the CTBT network was on 13 March at Takasaki, Gunma, Japan and after 2 weeks the whole northern atmosphere was affected. In the following months the CTBT radionuclide network continuously monitored enhanced levels of radioactivity. But by mid June 2011, activity concentrations at most stations where down to background level.

Atmospheric transport modeling (ATM) played an important role during the first days since they provided means to forecast which stations were going to be affected by the release.

Although the CTBT radionuclide system performed well during this event, the operation of the network and data processing and analysis of radioactive samples highlighted a number of challenges. This paper will focus on a summary of activity concentrations of radioactive particulates (mainly I-131, Cs-137 and Cs-134) and noble gases (mainly Xe-133) detected during this event. It will include a discussion on isotopic ratios and will identify lessons learned about procedures relevant for this kind of situation, taking into account the dynamic range of the existing measurement equipment and possible future improvements.

OBJECTIVES

From March to June 2011, the global radionuclide network of the CTBTO , which has been designed for the verification of the CTBT, was faced with a new situation. All stations on the northern hemisphere detected radionuclides emitted from the Fukushima nuclear power plant for a period of more than 6 weeks. Very high levels of activity concentrations were observed which in some cases even exceeded the dynamic range of the high sensitivity monitoring systems. Despite these challenges the network performed well and provided high-quality information on the dispersion of the radioactive plume on a daily basis, which was of high value for estimating the global impact of the release from Fukushima Dai-ichi nuclear power plant.

RESEARCH ACCOMPLISHED

The IMS Radionuclide Network

The status of the IMS network is shown in Figure 1. In March 2011, 63 stations for particulate monitoring and 27 noble gas systems are installed, 60 of the particulate and 3 noble gas systems have been certified.

Figure 1. IMS Stations installed in March 2011.

Network for the monitoring of particulates

Each station in the radionuclide particulate monitoring network consists of a high-volume aerosol sampler with a flow rate of typically around 900 – 1000 m^3/h. Aerosol-bound particulates are sampled for a period of 24 hours on filter material. The particulate fraction is filtered through conventional filters, mainly glass fibers or polypropylene fibers with a collection efficiency of > 80% for 0.2 µm particles (aerodynamic equivalent diameter).

After sampling, the filter decays 24 hours to minimize the contribution of radon progenies. In the final step the samples are measured using HPGe detector systems with a typically relative efficiency of 70 – 100% (Schulze et. al., 2000). The entire measurement cycle for one sample consists of three steps, sampling, decay, and measurement, and each of these steps lasts 24 hours; therefore the overall duration of a measurement is 72 hours. During the measurement cycle preliminary results are transmitted every 2 hours to the International Data Center (IDC) of the

Provisional Technical Secretariat (PTS) in Vienna, so the first information about sample analysis is available 50 hours after the start of the sampling cycle. The final gamma spectrum is sent to the IDC for detailed analysis and review after the full 24 hour measurement cycle is completed. The equipment installed and the measurement schedule provide minimum detectable concentrations (MDC) (Currie, L., 1968) of a few $\mu Bq/m^3$ for many nuclides. Table 1 shows the detection limits of some key nuclides.

Network for noble gas monitoring

The noble gas network consists of monitors for the surveillance of air-borne xenon isotopes Xe-131m, Xe-133. Xe-133m and Xe-135 (half-lives between 9.14 hours and 11.84 days). Due to the high yields in uranium and plutonium fission and half lives long enough to be detected within days and weeks after release from the source, these noble gases are of particular interest for the CTBTO monitoring system. Atmospheric background concentration of radioxenon is low due to the short half lives of these isotopes avoiding significant accumulation in the atmosphere. Typical background levels observed in IMS are below 1 mBq/m^3 which are mainly due to releases from civil sources (such as medical isotope production facilities or reactors). Ratios of xenon isotopes can discriminate nuclear explosions from other sources.

Due to the requirements on sensitivity and 95% data availability, noble gas monitoring systems were specifically developed for the IMS and in 2002 the first commercial systems became operational. Currently, three different types of noble gas systems are deployed in the IMS network. Sample processing of xenon gas in all three systems is based on similar techniques and the minimum detectable concentration for all systems is below 1 $\mu Bq/m^3$ for the xenon isotopes. Xenon gas is extracted from atmospheric air, accumulated and separated based on absorption of xenon on activated charcoal. Xenon activity concentrations are measured using either high resolution gamma spectrometry (SPALAX) or beta-gamma coincidence techniques (SAUNA and ARIX). The systems have different sampling times varying between 12 and 24 hours. Results are available between 31 and 48 hours after acquisition start.

The IMS particulate network of 80 stations will be complemented by 40 noble gas systems, with an option for extension to 80 after entry into force of the CTBT. By early 2011, 27 stations had been installed, of which 24 stations were transmitting data to the IDC during the relevant time period between March and June 2011.

Table 1. System Specifications for IMS Stations

Nuclide	Minimum Detectable Concentrations ($\mu Bq/m^3$)		Sampling Duration	Reporting Time
I-131	1-7	Particulate	24 hours	72 hours
Cs-137	1-5			
La-140	2-20			
Xe-133	200	Noble Gas	12 or 24 hours	31 or 48 hours

Data analysis

At the IDC each spectrum is automatically processed, analyzed and manually reviewed for the presence of fission and activation products considered relevant for CTBT verification.

Characteristic gamma ray energies (I-131: 364.5 keV; Cs-134: 604.7 and 795.9 keV; Cs-137: 661.7 keVs) are used to identify and quantify the radionuclides. Summation effect corrections are applied to the determination of Cs-134. This correction is especially required for the determination of Cs-134/Cs-137 ratios. The level of naturally occurring Be-7 was used for quality control since typical specific activities of the cosmogenic Be-7 are known for each station.

From the automatic processing of samples so-called Automatic Radionuclide Reports (ARR) are generated. Final products after interactive review of samples by analysts at IDC are the so-called Reviewed Radionuclide Reports (RRR). Each sample is interactively reviewed. Due to IDC policies the review is performed on a peak by peak basis. Manual changes of results provided by the automatic processing have to be verified and commented by analysts.

In the first days after the accident at the Fukushima nuclear power plant (NPP) analysts had to deal with complex spectra including multi-line isotopes like I-132. It turned out that the software had to be re-adjusted to deal with more than 100 comments provided by analysts to comply with IDC rules. The quality of some of the hottest samples reported from Takasaki station had bad energy resolution due to the loss of power at the station and subsequent loss of resolution in the spectrometer. Therefore it is important to consider comments in the RRR. Furthermore, identification and concentration of some radionuclides (especially Pm-151 and I-135) in these samples may remain uncertain and should not be used for further assessment.

In some cases where radionuclides are daughter products (which are produced during the sampling, decay, and acquisition of the sample), the half life in the reports is listed as 9.9999 years. This half life is needed in the processing to allow association of radionuclides in question since parent-daughter correction has not yet been implemented in the analysis software. In these cases the actual half life of the radionuclide is shorter. Nuclides which are in this category include for example I-132, Pr-144, and Te-129.

The IMS station JPP38 in Takasaki, Japan

The IMS is operating one station in Takasaki, 200 km south-west of Fukushima. The station is equipped with one fully automatic high volume air sampler and measurement system of the RASA type and one beta-gamma Noble Gas station of the SAUNA type. The activity concentrations observed at Takasaki are shown in Figure 2.

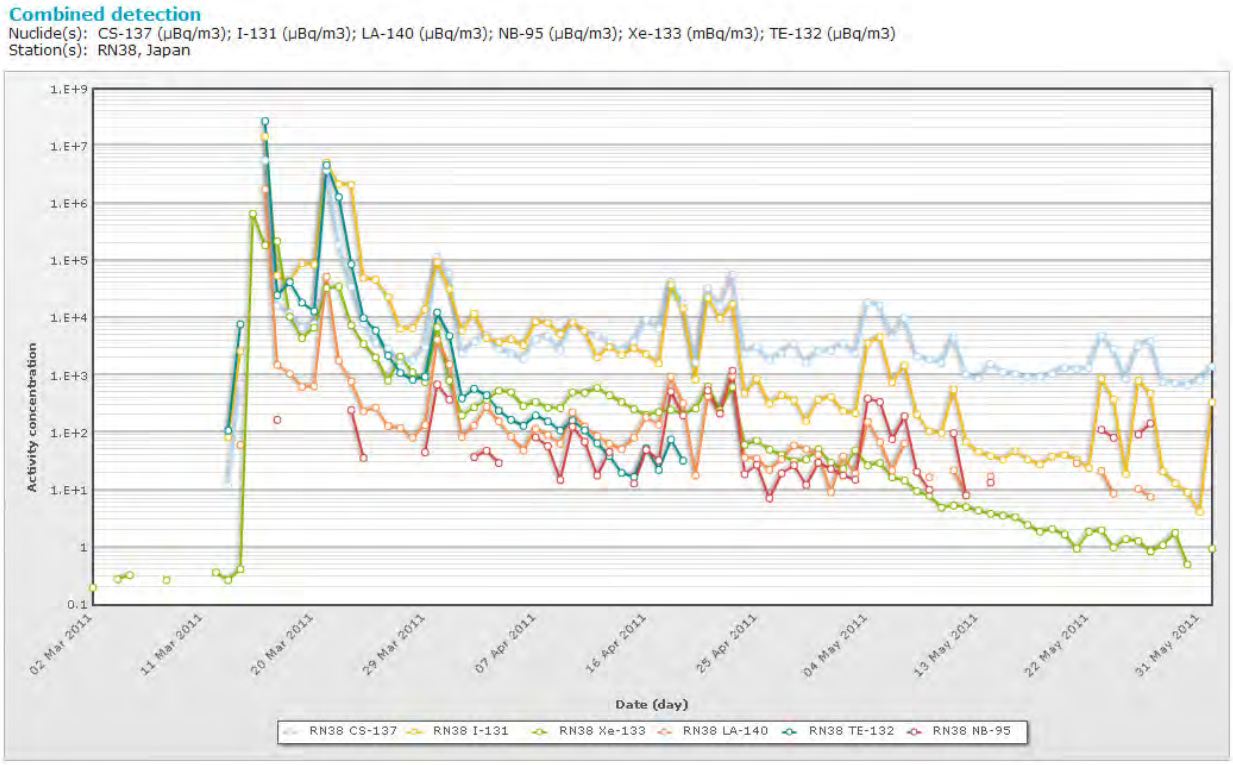

Figure 2. Activity concentrations at Takasaki particulate station (JPP38) 200 km south-west of Fukushima.

The first detection was reported from JPP38 for the sample with collection period 12.3.2011, 06:55 to 13.3.2011, 06:55 Coordinated Universal Time (UTC). Several gaseous fission products like Cs-134, Cs-136, Cs-137, I-131, I-131, I-133, Te-132, Ba-136m and Xe-133 were detected. But it has to be pointed out, that the sample in the measurement position was clean and the detector did not measure radionuclides originating from the sample, but rather from the contaminated air entering the station and the shielding of the detector. Therefore, calculated activity concentrations are not reliable, but could be used as a clear indicator that the Takasaki station was affected by releases from Fukushima NPP.

In the 3 days following the initial release, there were several power outages at the Takasaki station, which reduced the daily sampling time and stopped the cooling of the HPGe detector. Consequently, the detector was switched off and the sample with collection period 14.3.2011, 06:55 to 15.3.2011, 06:55 (UTC) was not measured. On the following day the station operator was able to cool the detector down to operational temperature and measurements could be performed again at the station. The next sample that could be measured for about one hour was from collection period 15.3.2011, 06:55 - 16.3.2011, 06:55 (UTC), although the loss of cooling decreased the energy resolution of the detector. Since there were also a large number of peaks in the spectrum, analysis of the sample at IDC was challenging, especially with respect to multi-line nuclides like Cs-136 and I-132.

Table 2. Activity concentrations for I-131 and Cs-137 on the first days after 11 March 2011

Collection Stop	I-131 / µBq/m3	Cs-137/ µBq/m3	Comment
2011/03/14 06:55	2.7E+03	7.1E+02	Nuclides are not originating from the sample, but rather from the contaminated air entering the station and the shielding of the detector.
2011/03/15 06:55	-	-	Sample not measured due to power outage at the station – hottest sample
2011/03/16 06:55	1.5E+07	5.6E+06	
2011/03/17 08:11	5.6E+04	1.6E+04	
2011/03/18 06:57	4.4E+04	1.2E+04	

In the days following 11 March 2011, the PTS was in close contact with the Takasaki station operator both to assure the health and safety of the station operator and to keep the system operational. The station operator was instructed to carry gamma dose rate meters when entering the station and to not touch the sample. To reduce contamination of the RASA system, the air flow of the particulate air sampler was reduced from 900 to 300 m^3/h. Furthermore, the station operator was asked to measure the gamma dose rate of individual samples to assess the activity concentrations in the sampling period – especially for those samples which could not be analyzed by the HPGe station detector. From these measurements it became obvious that the sample with collection stop on 15 March has the highest activity. The sample is still at the station since it cannot be sent due to public transport regulations.

Detections in the particulate and noble gas network of IMS

The emission of radioactive material into the atmosphere after the accident at the Fukushima NPP started on 12 March. The dominant radionuclides were xenon isotopes and especially Xe-133 together with I-131, Cs-134, and Cs-137, and further short-lived radionuclides like Te-132 and I-132 were also detected.

Figure 2 shows the time development of particulate detections for each day after the accident in the 3 month period from 13 March to 15 June 2011.

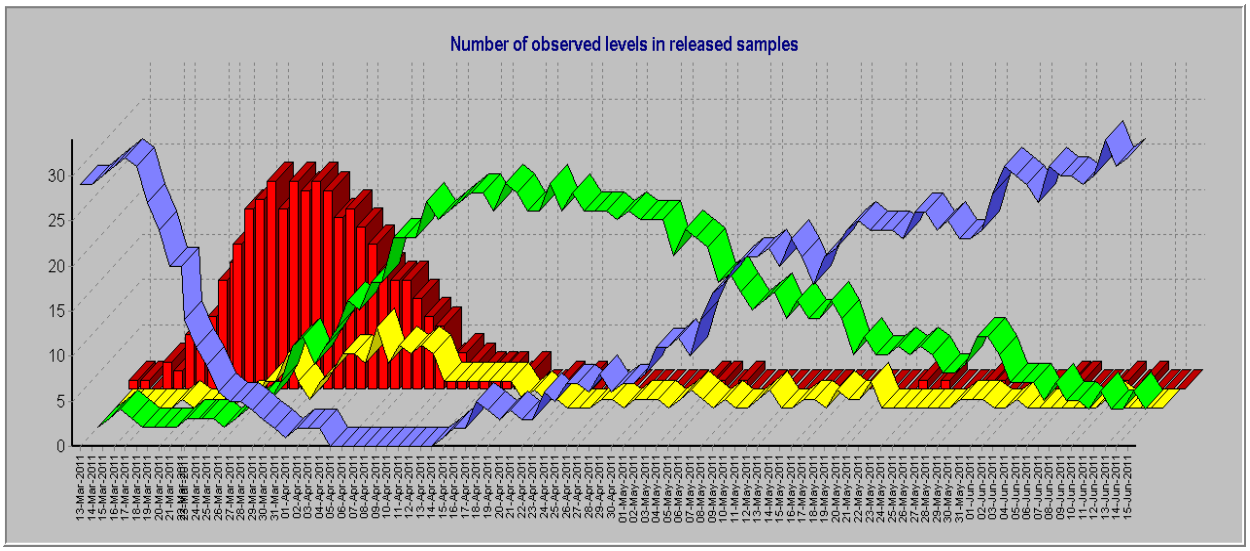

Figure 2. Time development of particulate detections for each day after the accident from 13 March to 15 June 2011. The number of observed levels of detections is shown. Level 5 = multiple fission products detected, Level 4 = one fission product detected, Level 3 = fission products typical for the station detected; Level 1 and 2 = only natural radioactivity detected.

Figures 2 and 3 show that the main global impact from Fukushima NPP was in the first 4 weeks when multiple radionuclides were detected in the samples, but it took about 2 months until the radiological situation came back close to normal background conditions.

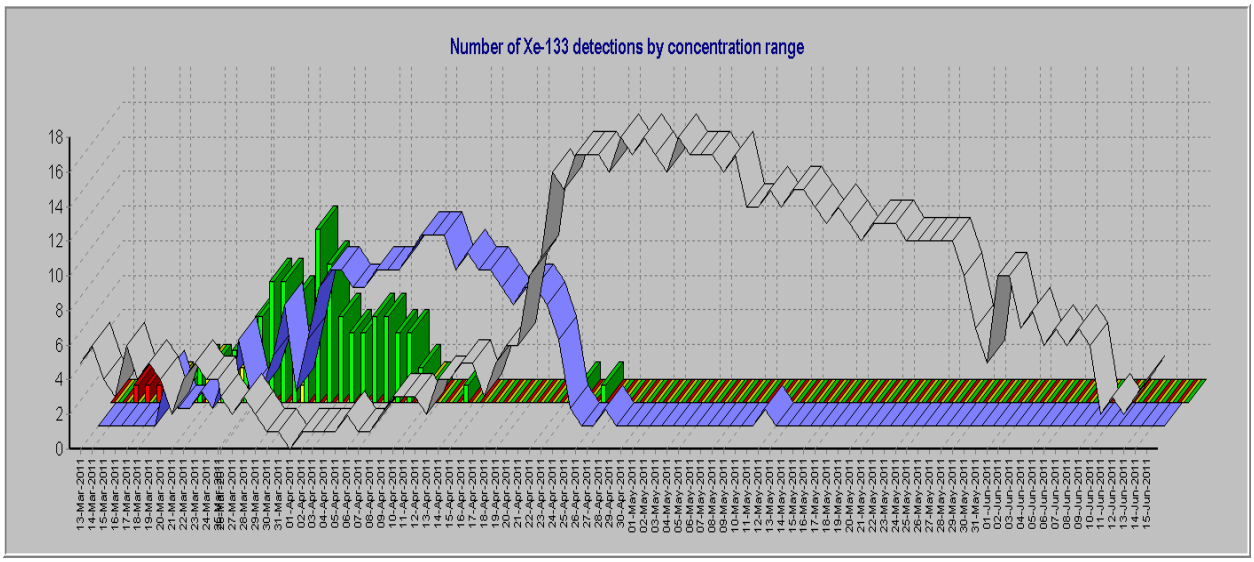

Figure 3. The daily time development of Xe-133 detections from 13 March to 15 June 2011. The number of observed Xe-133 of detections is shown with values >100 Bq/m3, 10-100 Bq/m3, 1-10 Bq/m3 , 1-0.1Bq/m3 and <0.1 Bq/m3.

In the period from April to July 2011, more than 40 stations detected radionuclides released from Fukushima NPP and more than 1600 samples contained radiation originating from this event. Today, enhanced levels are only reported from the closest station in Takasaki, Japan. This is mainly caused by the contamination of the sampler and detector of the IMS stations. Furthermore re-suspension causes spurious increases of mainly Cs-137 levels.

Activity concentrations observed in the particulate network of IMS

Two weeks after 12 March, all operational particulate stations in the northern hemisphere had reported at least one I-131 detection and all noble gas stations detected Xe-133. Three stations in the southern hemisphere detected I-131 (RN51, RN26 and RN39). The highest level measured was in the range of 100 Bq/m^3 for I-131. Further detections reported from almost all particulate stations were due to gaseous releases of I-130, I-133, Te-129m, Te-129, Te-131m, Te-132, Cs-132, Cs-134, Cs-136, and Cs-137. Particulate releases of Zr-95, Nb-95, Ba-140, La-140, Mo-99, and Tc-99m were detected, but the release was mostly gaseous; solid materials were well contained. Due to the early observation of Te-132 it can be concluded that the fuel in the reactor core was damaged shortly after the earthquake and tsunami since the detection of tellurium indicates temperatures above 1000˚C. With a long half-life of 30.2 years and by re-suspension of soil particles containing traces from former deposition (fallout from nuclear weapon tests or from the Chernobyl accident), Cs-137 is observed sporadically in the IMS network with concentrations close to the detection limit of about 1 $\mu Bq\ m^{-3}$. Cs-134 with a short half-life of 2.06 years is rarely detected.

The gaseous fraction of I-131

Cesium is rapidly bound to aerosols, but iodine is mainly found in gaseous form. To determine the total level of airborne I-131 activity concentrations, both the gaseous and particulate fractions of I-131 must be accounted for. This is especially important for dose assessment. However, sampling of gaseous radioiodine requires activated charcoal traps, which are not required for CTBT monitoring and not implemented in the IMS radionuclide network.

During the Fukushima event, data for gaseous iodine in Europe were made available from a network called “Ring of Five (Ro5)”, which is an informal network of national authorities in Europe. The network was started in 1983 and comprises more than 150 sampling systems equipped with high volume samplers for particulates and some with activated charcoal traps. Based on the results provided by Ro5, the average gaseous/total ratio for I-131 is 77.2% +/- 13.6% (Masson et al., (2011)). This value is close to the average reported after the Chernobyl accident (Cambray et al., 1987) and to the 71% +/- 11% average reported from Fukushima NPP site during the period from 22 March to 4 April. Therefore a factor of 4 for the gaseous in comparison to the particulate fraction is considered as appropriate estimate.

Isotopic ratios

Figure 5 shows selected isotopic ratios measured at Takasaki from the Fukushima release. As can be seen from this figure, the Cs-134/Cs-137 ratio was close to 1 and constant over time. This differs from the ratios in the range 0.5 - 0.6 (De Cort et al., 1998) reported during Chernobyl. The smooth and constant Cs-134 /Cs-137 ratio suggests that radioactive material was predominantly released from the same type of source material. The variation in the Cs-136/Cs-137 ratio may indicate that source material included a mixture of different irradiated batches of fuel. The Te-132/Cs-137 provides information on the behaviour of metallic elements. Since this indicator did not change significantly over time, it can be interpreted that gaseous fission products are constantly dominating.

Detections in the noble gas network of IMS

The sample from the SAUNA system installed in Takasaki with collection stop at 15.3.2011, 08:55 - 20:55 (UTC) showed clear detection of radioxenon, but far above the dynamic range of the measurement system. Normal spectrum analysis was not possible and the high level xenon concentration (in the kBq/m3 range) caused the detector to suffer a prolonged memory effect. Therefore the reported activity concentrations from this station are only estimates. In addition dead time corrections of nearly 10% are necessary for samples around 1 Bq/m3. These are typically below 1 % for normal background samples.

As seen in Figure 4, detected activity concentrations of Xe-133 with values above the dynamic range of the measurement system (>100 Bq/m3) were observed only on the first days at Takasaki station. For all other stations, the maximum levels were between 1 and 0.1 Bq/m3. Since mid of June 2011 the global xenon activity concentrations were back to normal.

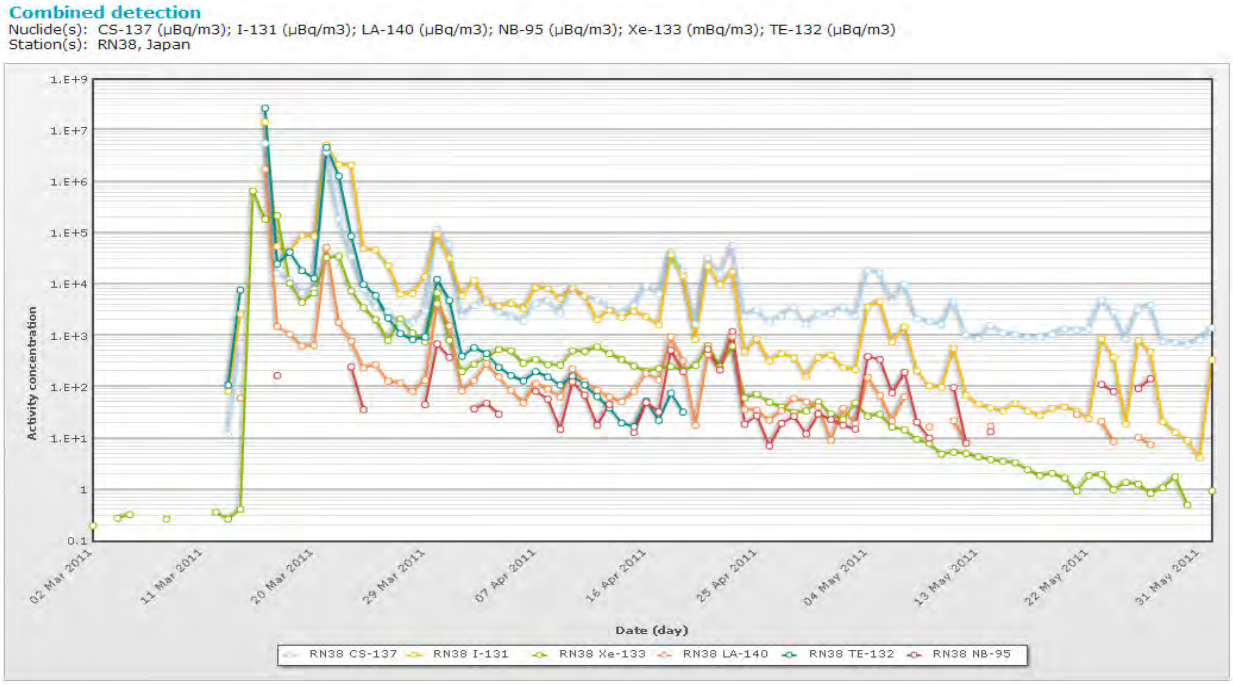

Figure 4. Activity concentrations at Takasaki particulate station (JPP38) 200 km south-west of Fukushima.

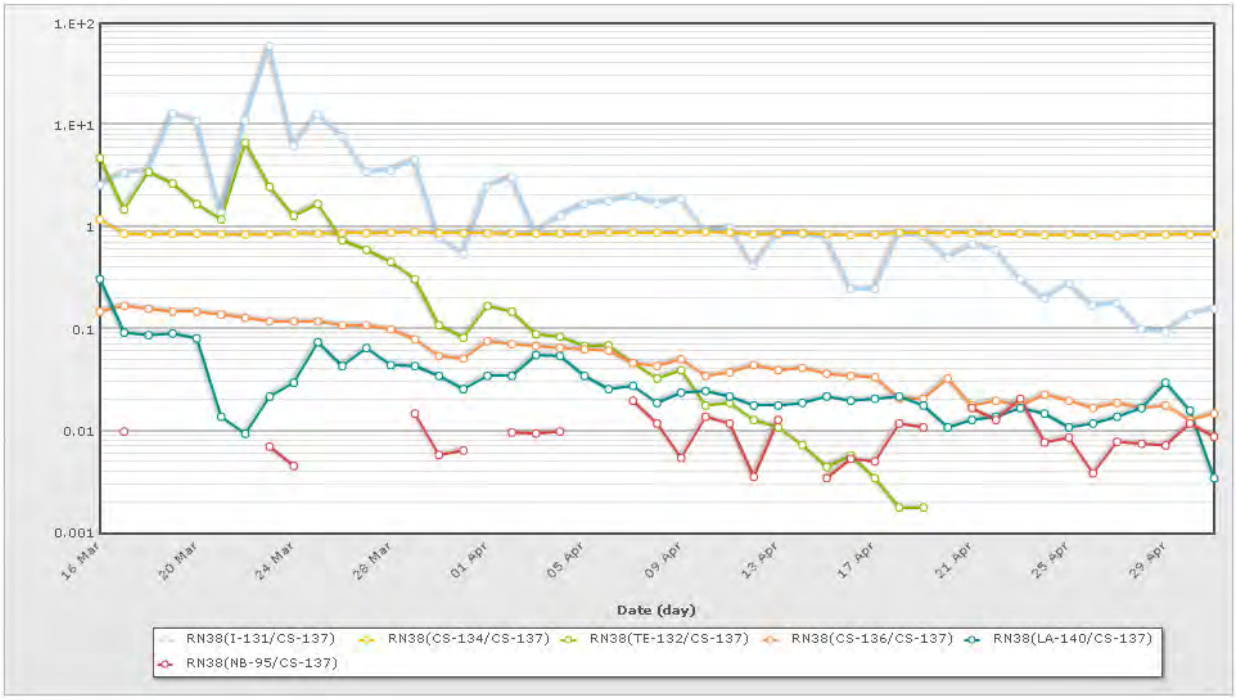

Figure 5. Isotopic ratios for selected radionuclides at Takasaki particulate station (JPP38) 200 km south-west of Fukushima.

Atmospheric Transport Modeling

After the Fukushima incident, atmospheric transport forward modeling was used to qualitatively predict the IMS radionuclide stations likely to be affected by the radioactive release and to estimate the time of the first detections. ATM capabilities have been developed at IDC to support treaty verification.

Once per day forward ATM calculations were performed and the results were posted on the IDC secure website. Snapshots of the plume were taken to illustrate its propagation in time every 24 hours and were assembled in pdf documents. In addition animations were posted to illustrate the dynamics of the plume. Each day's calculations covered the period from 12 March 2011 at 00:00 UTC to the recent day plus 3 days in the future at 00:00 UTC. The calculations were driven by the analyzed meteorological wind fields for the past dates and by the forecasts for the future dates. The atmospheric transport model came from FLEXPART version 5 using the meteorological fields from National Centers for Environmental Prediction (NCEP) with 1x1 degree resolution. The source term assumed a constant release over the time interval covered by the calculations with a total release equivalent to a 1 kton atmospheric nuclear weapons test (approximately 10^{16} Bq) beginning 12 March 2011 at 05:30. A passive tracer was assumed, so the model did not take into account the special characteristics of aerosol bound radionuclides like washout (removing particles by precipitation) or radioactive decay. Further uncertainties are linked to the use of forecast meteorological fields. Therefore model results were only qualitative not intended for isotopic concentration estimates.

The spread of the release was such that all the IMS radionuclide stations in the northern hemisphere and three stations in the south reported one or more detections. In early April 2011, the radiological situation became stable and no new detections from IMS stations were reported. Consequently, forward ATM modeling could no longer provide significant additional information. Backtracking atmospheric transport modeling continues as part of routine IDC operations and those results are attached in each RRR.

CONCLUSIONS AND RECOMMENDATIONS

The CTBTO radionuclide network established to detect nuclear test explosions is capable of detecting anthropogenic radioactivity in the $\mu Bq/m^3$ range on a global scale. Together with atmospheric transport calculations these detections can help emergency response authorities assess situations before plumes arrive. The high sensitivity of the measurements also allows in-depth analysis of the nature and severity of a release. The IMS network continuously monitored the radiological situation. According to the measurement results, it became obvious that the radiological situation was of no general public health concern.

Maximum activity concentrations of radioactive particulates (dominated by I-131, Cs-137 and Cs-134) and noble gases (mainly Xe-133) were detected at the IMS station in Takasaki, Japan on the 16 March (collection stop) with values for I-131, Cs-137 and Cs-134 and noble gases below 1kBq/m3 (external dose rate factor 10,000 below typical natural radiation (2mSv per year)).

One day later, the IMS station at Petropavlovsk-Kamchatskiy, Russian Federation was affected with activity concentrations for I-131 in the mBq/m3 range.

The data obtained from all stations of the IMS network in the northern hemisphere showed an almost constant ratio of Cs-137 and Cs-134. This ratio is an indicator of the burn-up of the radioactive fuel in the reactor core. it implies that the release of radioactivity to the atmosphere was very probably from the same source during the whole event ATM calculations were performed on a daily basis through April at IDC. The prediction of the plume agreed with the real detections of enhanced activity concentrations reported from IMS highlighting the predictive capabilities of this modeling technique applied at IDC.

The CTBT radionuclide system, including the operation of the network, data processing, and analysis of radioactive samples performed well during this event. From mid March to mid June 2011, more than 400 samples indicated abnormal concentrations of more than one CTBT relevant anthropogenic radionuclides (Level 5 samples). In this same period about 1600 samples contained traces of radionuclides released from Fukushima NPP.

During the Fukushima event CTBTO collaborated with other international organizations, especially the International Atomic Energy Agency, the World Meteorological Organization, and the World Health Organization. The IMS includes a unique radioactivity network covering the entire globe and providing high-quality data. The information provided by CTBTO was of high value to estimate the global impact of the release from Fukushima Dai-ichi nuclear power plant.

REFERENCES

Cambray, R. S. P.A. Cawse, J.A. Garland, J.A.B. Gibson, P. Johnson, G.N.J. Lewis, D. Newton, L. Salmon, and B.O. Wade (1987). Observations on radioactivity from the Chernobyl accident. *Nuclear Energy*, 26, issue 2: 77–101.

Currie, L. (1968). Limits for qualitative detection and quantitative determination. *Anal. Chem.*, 40: 586–593.

De Cort M., G. Dubois, Sh. D. Fridman, M.G. Germenchuk, Yu. A. Izrael, A. Janssens, A. R. Jones, G. N. Kelly, E. V. Kvasnikova, I. I. Matveenko, I. M. Nazarov, Yu. M. Pokumeiko, V. A. Sitak, E. D. Stukin, L. Ya. Tabachny, and Yu. S. Tsaturov (1998). Atlas of Caesium Deposition on Europe after the Chernobyl Accident, EUR report nr. 16733, *Office for Official Publications of the European Communities, Luxembourg.*

Masson, O., A. Baeza, J. Bieringer, K. Brudecki, S. Bucci, M. Cappai, F.P. Carvalho, O. Connan, C. Cosma, A. Dalheimer, G. Depuydt, L.E. De Geer, A. De Vismes, L. Gini, F. Groppi, K. Guðnason, R. Gurriaran, D. Hainz, Ó. Halldórsson, D. Hammond, K. Holý, Z. Homoki, A. Ioannidou, K. Isajenko, C. Katzlberger, M. Kettunen, R. Kierepko, R. Kontro, P.J.M. Kwakman, M. Lecomte, A.-P. Leppänen, B. Lind, G. Lujaniene, P. Mc Ginnity, C. Mc Mahon, H. Malá, S. Manenti, M. Manolopoulou, A. Mattila, A. Mauring, J.W. Mietelski, B. Møller, S.P. Nielsen, R.M.W. Overwater, S. E. Pálsson, C. Papastefanou, I. Penev, M. Pham, P.P. Povinec, H. Ramebäck, M.C. Reis, W. Ringer, A. Rodriguez, P. Rulík, P.R.J. Saey, V. Samsonov, C. Schlosser, G. Sgorbati, B. V. Silobritiene, C. Söderström, R. Sogni, L. Solier, M. Sonck, G. Steinhauser, T. Steinkopff, P. Steinmann, S. Stoulos, I. Sýkora, N. Tooloutalaie, L.Tositti, J. Tschiersch, A. Ugron,E. Vagena, A. Vargas, H. Wershofen, and O. Zhukova (2011). Tracking of Airborne Radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. *To be published in Science.*

Schulze, J. M. Auer and R Werzi (2000). Low level radioactivity measurement in support of the CTBTO. *Appl. Radiat. Isot*. 53: 23–30.

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