ACDIS Research Report

Preparation of a Global Radioxenon Emission Inventory: Understanding Sources of Radioactive Xenon Routinely Found in the Atmosphere by the International Monitoring System for the Comprehensive Nuclear-Test-Ban Treaty

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Martin B. Kalinowski holds a PhD in nuclear physics dealing with international tritium control. He was a scientific assistant for a decade in the Interdisciplinary Research Group on Science, Technology, and Security (IANUS) at Darmstadt University of Technology, Darmstadt, Germany. He co-founded the International Network of Scientists and Engineers Against Proliferation (INESAP) and acted as the Scientific Coordinator of this non-governmental organization from 1994 to 1997. In October 1998, Kalinowski joined the International Data Center of the Provisional Technical Secretariat of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), Vienna, Austria. In spring 2005, he was an Assistant Professor in the Department of Nuclear, Plasma, and Radiological Engineering at the University of Illinois. Beginning in spring 2006, Kalinowski will be a professor for science and peace research at the University of Hamburg, Germany. He will be the founding director of the Carl-Friedrich von Weizsäcker Center for Science and Peace Research (http://www.uni-hamburg.de/fachbereiche-einrichtungen/znf/index e.html). His current research deals with atmospheric radioactivity monitoring and analysis as a means to detect clandestine nuclear testing. More generally, his research interests are scientific-technical based innovations to nuclear disarmament and non-proliferation towards a nuclear weapons-free world; verification of arms control treaties; concepts for international control of nuclear weapons-usable materials (plutonium, highly enriched uranium, tritium); and anthropogenic sources of atmospheric radioactivity and their transport through the atmosphere.

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- BWR Boiling water reactor
- CTBT Comprehensive Nuclear-Test-Ban Treaty
- HWR Heavy water reactor
- IAEA International Atomic Energy Agency
- INGE International Noble Gas Experiment
- LWR Light water reactor
- PRIS Power Reactor Information System
- PWR Pressurized water reactor

INTRODUCTION

Atmospheric Radioxenon Background Concentration

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) was opened for signature in 1996. In 1997 the Provisional Technical Secretariat began building up the verification system as part of the International Monitoring System (IMS). When completed, forty to eighty stations will monitor the atmosphere for radioactive xenon.

Four radioactive xenon isotopes are the best indicators of nuclear weapon tests: Xe-135, Xe-133m, Xe-133 and Xe-131m. The challenge for verification of compliance with the CTBT based on atmospheric radioxenon is created by the fact that all nuclear reactors are constantly emitting these isotopes as normal operational releases. The background concentration of Xe-133 in the northern hemisphere is in the order of the detection limit at about 1 mBq/m³. The other three isotopes have typical concentrations below the detection limit and are seen only during the passage of a plume with elevated concentrations over the detector site.

In order to improve the interpretation of measurements as possible indications of a nuclear test in the presence of high background caused by civilian facilities, it is crucial to establish an understanding of what levels of concentrations could be expected as normal at the radionuclide station sites, which may vary depending upon seasonal and weather related conditions.

The goal of the project described in this research paper is to prepare a database on global radioxenon emissions from nuclear reactors. There are more than 400 power reactors and about fifty large research reactors that have to be taken into consideration. The most accurate data are based on effluent data reported by the respective facility operator. The emissions of those reactors for which no reported data are available are estimated from suitable reactor parameters such as fuel inventory or power output. Various reactor parameters are investigated and the ones providing the most precise relationships with fission gas activity releases are used for scaling.

The resulting emission inventory shall be used as input to global atmospheric transport models to determine the global distribution of the four relevant radioactive xenon isotopes. Their atmospheric background concentration as well as the deviations from their averages shall be determined climatologically, i.e. depending on seasonal weather patterns. The background concentration at all IMS measurement sites is particularly of high interest. These stations have a time resolution of eight, twelve, or twenty-four hours. This is much better than the current standard of taking weekly samples, and it will be possible to resolve passages of fresh plumes with sharp concentration gradients. This time resolution will allow the application of methods for atmospheric transport modeling in order to determine the source location.

PART ONE Reported Nuclear Power Reactor Effluents

Data Sources for Power Reactor Effluents

Most available data on xenon releases are extracted from quarterly effluent reports for the years 1999 to 2003 as made available by the North American Technical Center (2000, 2001, 2002, 2003, and 2004). The North American Technical Center (http://hps.ne.uiuc.edu) is located at the Nuclear, Plasma, and Radiological Engineering Department of the University of Illinois at Urbana-Champaign. During the time covered by these reports, the United States had 104 nuclear reactors licensed to operate in thirty-one states. Thirty-five of these are Boiling Water Reactors (BWRs) and sixty-nine are Pressurized Water Reactors (PWRs). In many cases, the discharge values for certain noble gases are reported to be below the detection limit. The quarterly reports distinguish between gas and liquid phases as well as between continuous and batch releases. In some reports the gas releases are reported for different heights (ground level, semi-elevated or roof vent, and elevated level).

Further data are taken from the report on releases to the environment of radioactive materials in airborne and liquid effluents from the period 1995–2003 (Van der Stricht and Janssens 2001; EC 2004). It covers discharges from operational nuclear power stations of capacity greater than 50 MWe in the European Union (EU). During the time covered by the report, seventy-three nuclear power stations totalling 148 reactors were operational—these are spread over sixty-four different sites within the territory of the European Union. Xenon release data are available for forty-three nuclear reactors at thirty-two sites in Finland, Germany, Spain, and Sweden. In some cases, the discharge values for noble gases are reported to be below the detection limit.

In addition, release data from the four Swiss nuclear power plants are included in this work (BAG 2000, 2001, and 2002). Data on four CANDU type Heavy Water Reactors (HWRs) are available (ISR 2001).



Fig. 1: Distribution of quarterly effluents of four xenon isotopes reported for U.S. and European light water reactors (LWRs). The dotted line represents the median emission and the solid line represents the logarithmic mean.

Characterization of Xenon Effluents from Nuclear Power Plants

Fig. 1 shows the distribution of released activities of the four CTBT relevant xenon isotopes reported for U.S. PWRs. The plots and the following preliminary analysis of the various release modes are based on about half of the available data. In particular, quarterly effluent release reports for reactors in the United States are used for the years 1999 and 2003. The data for 2000, 2001, and 2002 are available but not yet incorporated into the database.

1. Gaseous effluents, continuous mode

The gaseous effluents in continuous mode are dominated by Xe-133. On average, the annual discharge amounts to 1,500 GBq per reactor. About 5% are due to Xe-135. The meta-stable isotopes are below detection limit in every second report. Nevertheless, in average 20% of all releases are Xe-131m. The contribution of Xe-133m is three orders of magnitude below Xe-133.

2. Gaseous effluents, batch mode

On average, 120 batch releases per year are reported for a single reactor, i.e. about one every third day. The average duration is five hours with a typical range between one and thirty hours. Fig. 2 shows the distributions of the batch release frequency and of the duration of batch releases.

The average activity released per batch is 5 GBq. In single cases, ten times as much may be released in one batch. Almost all of the average activity—about 95%—is due to Xe-133, the shorter lived isotopes Xe-135 and Xe-133m have almost equal shares and the Xe-131m activity is almost two times as high as the Xe-133m activity. Cumulated over the year, the average total release in batch mode is 700 GBq.

3. Liquid vs. gaseous effluents

Fig. 3 gives a comparison of average activity release rates for the liquid and the gaseous phase of the four CTBT relevant xenon isotopes. The activities in liquid releases are lower than in gaseous effluents by one to two orders of magnitude (see Table 1).



Fig. 2: (a) Distribution of batch release frequency



(b) Duration of batch releases



Fig. 3: Comparison of total gaseous vs. liquid activity release rates for the four xenon isotopes averaged over all data sets available irrespective of reactor type and reporting period.

	Xe-131m	Xe-133	Xe-133m	Xe-135	total
gas, continuous	9.02E+04	3.42E+05	1.17E+04	2.15E+04	4.66E+05
gas, batch	3.99E+03	1.57E+05	2.14E+03	2.31E+03	1.65E+05
liquid, batch	3.40E+02	4.57E+03	1.34E+02	3.05E+03	8.09E+03

Table 1: Activities [MBq] in three release modes averaged over all quarterly reports for LWRs in the USA.

4. Comparing continuous and batch mode of gaseous releases

The total activity integrated over a longer period (three months) is three times higher for the continuous release mode in comparison to batch releases (see Table 1). Fig. 4 illustrates the time pattern of continuous vs. batch releases.

Xe-131m is the isotope with the longest half-life of the four isotopes considered here. Its fraction is particularly larger in continuous releases than in the batch mode. This indicates that many of the batch releases have shorter delay times within the facility as compared to the continuous emissions. The latter can be assumed to have contributions from air masses of various ages including gas that is of a comparatively older age after being held back in the decay line gas tanks. In contrast, batch releases are expected to have small variances in residence time and to be typically very fresh as a result of a prompt pressure vessel plenum gas release upon opening a valve or even the lid.



Fig. 4: Illustration for average time pattern of continuous vs. batch releases.

For Xe-135, the isotope with the shortest half-life, the fraction found in continuous mode is significantly higher than in the batch mode as well. Since Xe-135 has the shortest half-life of all four isotopes (9.1 hours), these relative activities indicate that continuous releases are comparatively fresh, while batch releases are containing gas that has been collected for some time inside the reactor containment. The high fraction of Xe-135 in continuous releases may to some extent be due to changes of the neutron flux, especially during shutdown and start-up of the reactor.

Comparison with Other Data and Preliminary Conclusions for CTBT Monitoring

The isotopic activity ratios of the xenon isotopes carry important information about the source. Plots of activity ratios for one pair of isotopes vs. another pair of isotopes in logarithmic scale can be used to characterize the source of the emission and in particular to discriminate between nuclear reactors and nuclear explosions (Kalinowski 2005; Kalinowski et al. 2005). The green separation line in Fig. 5 has a slope that is determined by the half-lives of the respective isotopes. With time progressing, the entry for a certain gas sample moves along a straight line with the same slope towards the lower left corner of the plot due to the radioactive decay.

Fig. 5 shows the xenon ratio vs. ratio plots with logarithmic average values for the three release modes (gaseous and liquid batch releases as well as gaseous continuous effluents) in context with simulation curves showing typical trajectories for reactor operations and nuclear explosions. The locations of the values of the three release modes relative to the other data in these plots allow the following conclusions to be drawn.

The gaseous release in continuous mode is found in all five diagrams very close to the point of equilibrium that is reached after a few weeks of continuous reactor operation. In particular, the isotopic ratios do not indicate a move away from the equilibrium point along the line of radioactive decay. As a result, it can be concluded that the continuous releases are always fresh releases that reach the stack from the reactor core within one or at most several hours. Also, the batch releases can be characterized as clean gas samples not mixed with any contribution from a second reservoir.



Fig. 5: Xenon ratio vs. xenon ratio plots with average values for continuous gas, batch gas and batch liquid effluents from PWRs, put in perspective with other data.

The values of gaseous releases in batch mode are found further towards the center of the simulation results that represent the xenon activities as they change with the reactor power cycles. In comparison to the entry for gaseous continuous releases the batch releases are found shifted away from the line of clean sources and moved down along the line of decay. This can be understood by the fact that the batch effluents result from opening valves to release gas that was collected from various air streams from different reactor compartments and with different residence times. In particular, the batch releases contain a significant mix-in from the decay line that allows highly active air masses to decay for tens of days before being released through the stack. The distance from the point of equilibrium is equivalent to a residence time of two days.

The liquid release in batch mode is located closer to the gaseous batch release than to the gaseous continuous release. Obviously, its pathways through the reactor follow similar patterns as the gaseous batch releases. The location of the entries in the various plots indicate a little longer residence time and a bit more mixing of contributions from various sources for the liquid batch releases in comparison to the gaseous batch releases.

Since continuous releases are fresh and clean reflecting the stable and fairly well known reactor equilibrium concentrations, the detected isotopic ratios can be used for release dating as long as the activity release is large enough to be detected and as long as the air mass is not mixed with other air masses that contain xenon from another recent release still having activity concentrations with at least the same order of magnitude. However, it is more likely that detection results from a batch release because those lead to higher atmospheric concentrations.

PART TWO

Reactor Parameters for Estimating Radioxenon Emissions

Goals

The goal is to get the best estimate for radioxenon emissions from all relevant sources for which no effluent reports are available. To achieve this, the relationship between certain reactor parameters and the reported radioxenon effluents are analyzed. The most useful relationship shall be selected for scaling the estimated continuous emissions from a suitable reactor parameter.

Data Sources for Reactor Parameters

There are two major data sources for nuclear power plants:

1. The Power Reactor Information System (PRIS) of the International Atomic Energy Agency (IAEA) can be accessed online at http://www.iaea.org/programmes/a2/. This database contains basic reactor parameters like reactor type and design power for all 440 nuclear power plants in operation worldwide. The focus of PRIS is on reactor performance parameters like energy availability or unit capability factor. Special design parameters are currently available only for a limited number of power reactors.

2. The World Nuclear Industry Handbook (Verley et al. 1993) contains a large number of technical data for all power reactors and large research reactors. It is not available online or in electronic form.

For research reactors, the main source is a table of research reactors and their characteristics compiled from openly available sources at the Nuclear, Plasma, and Radiological Engineering Department of the University of Illinois at Urbana-Champaign (Jones 2005).

Search for the Best Parameterization of Radioxenon Emissions

The following parameters are selected for the first approach:

- Number of reactors for which aggregated effluent reports are available
- Power [MWe]
- Fuel inventory [t]
- Average linear fuel rating [kW/m]
- Peak linear fuel rating [kW/m]
- Maximum cladding temperature [°C]
- Cladding thickness [mm]
- Period between shutdowns for revision and reloading the core [months]

These parameters are selected for the first approach, because they are design parameters that are likely to influence the amount of fission gas generated and released, but they do not change during normal operation. A more realistic approach will be taken at a later stage. It will consider actual operational conditions, mainly the real power generation expressed by the capacity factor and the real times of reactor shutdowns and startups.

All reported effluent data are fitted to the parameters of the relevant reactors in order to determine the most suitable relationship for scaling. Least-square linear, quadratic, and cubic polynomials are fitted to the log-transformed activity release data. This is done for all eight parameters listed above and for all four CTBT relevant xenon isotopes (see Fig. 6). For each of the four isotopes of interest, the best relationship will be selected for scaling the estimated continuous emissions from a reactor parameter. Two criteria are applied to guide this selection. First, a good fit of reported effluent data to the reactor parameters is desired. Second, a strong dependence of the effluents on the respective parameter is required in order to have the reactor parameter

maintain a strong predicting capability when data are not available. A prospective candidate for being a good scaling parameter is the reactor power because the generated xenon quantity is approximately proportional to the number of fissions, which in turn is proportional to the energy release. Figs. 7, 8, and 9 provide a preview of the fittings on a limited set of data.



Fig. 6: Matrix of reactor parameters under investigation for possible scaling (rows) CTBT relevant xenon isotopes (columns).



Fig. 7: Linear fit of xenon effluent data to eight reactor parameters.



Fig. 8: Quadratic fit of xenon effluent data to eight reactor parameters.



Fig. 9: Cubic fit of xenon effluent data to eight reactor parameters.

PART THREE Estimation of the Global Radioxenon Emission Inventory

For the first time, a complete dataset will be put together that provides an estimate of radioxenon emissions for each relevant source. It is based on reported effluent data for those reactors for which these are available (see Part One). It assesses the emissions based on certain reactor parameters for those reactors for which no effluent reports are available (see Part Two).

Goals

Xenon isotopes are the most likely observable radioactive signatures of underground nuclear explosions. However, these isotopes will very frequently be detected because there are more than 500 nuclear facilities in the world that regularly release radioactive xenon during their normal operation. Therefore, proper methods are required to distinguish source types to facilitate the verification of compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

It has been expected that the xenon activity ratios would be useful for source discrimination (see e.g. Bowyer et al. 1998; Finkelstein 2001; Carman et al. 2002). However, first experience with atmospheric measurements (Heimbigner et al. 2002) and a detailed analysis (Kalinowski et al. 2005) show that the original concepts are insufficient. In particular, nuclear reactor emissions may have larger isotopic activity ratios than previously assumed, some as high as ratios that were thought to be unique indicators for nuclear test explosions.

In this paper, a best estimate for radioxenon emissions from all relevant sources is prepared. All nuclear power plants of the world are included as well as all research reactors in the southern hemisphere that have a thermal power of more than 100 kW_{th} as well as all research reactors with more than 10 MW_{th} in the northern hemisphere (see Fig. 10). The goal is to feed these data into a global atmospheric transport model in order to characterize the full range of possible radioxenon concentrations and isotopic signatures that might be expected from nuclear reactors at any of the atmospheric noble gas monitoring sites (see Fig. 15).



Fig. 10: Map showing all relevant radioxenon sources included in the global emission inventory.

Steps for Creating the Global Radioxenon Emission Inventory

Fig. 11 shows the flow-chart illustrating the process to generate the global radioxenon emission inventory. The reactor related input data are: reported emissions (continuous and in pulsed form), certain design parameters, geographic locations, and scenarios for reactor operation. The results of processing these data are: emission statistics (see Part One), relationships between emissions and reactor parameters (see Part Two), and the global radioxenon emission inventory.

In Part One, the analysis of the reported effluents is described. From the emission statistics important lessons are learned for establishing a reasonable global emission inventory. In particular, the batch release frequency and average duration is calculated for all relevant reactor types (PWR, BWR, HWR). Further, the relationships between continuous emissions and batch releases are drawn from the emission data analysis. For each reactor type and for each of the four xenon isotopes of interest, a factor is derived for the relationship between the activities released by continuous emissions and batch releases. Since there are three different reactor types, there are twelve factors.

In Part Two, the relationship between certain reactor parameters and the reported radioxenon effluents are analyzed. These are least-square linear, quadratic, and cubic polynomials to log-transformed activity release data. For each of the four isotopes of interest, the best relationship is selected here for scaling the estimated continuous emissions from a reactor parameter. Selection criteria are a good fit of reported effluent data to the reactor parameters and a strong dependence of the effluents on the respective parameter.



Fig. 11: Flow-chart of the process to generate the global radioxenon emission inventory. Blue boxes represent data files, and purple boxes indicate Matlab scripts for data processing.

The combining script uses the best available estimates for each reactor. It takes the average of all relevant reported data, if these are available. If emission reports are not available, the continuous releases are determined individually for each isotope by a scaling equation based on the reactor parameter providing the best relationship. If that reactor parameter is not available, the next best relationship is applied for an available reactor parameter. The next step is to determine the annual activity released by pulsed emissions from the continuous release by multiplying them with the respective factors described above.

The output file that will serve as input to the atmospheric transport model contains the following information (see sample file in the Appendix). It will have some aggregated information in the first few rows showing the total global radioxenon release activity by reactor type. The bulk of this file contains one line for each reactor in the world. The following columns will be provided:

- 1- Reactor identification number
- 2- Location in latitude and longitude
- 3- Continuous emissions (GBq/year)
- 4- Batch releases (GBq/year)
- 5- Frequency of the pulsed emissions (1/y)
- 6- Duration of the pulsed emissions (h)

Open Issues and Future Work

The selected scenario defines the quality of the data. In the first phase of this project, a generic year is used that makes use of as much information as possible. A better approximation can be achieved by selecting a specific year. This can only be implemented at a later stage in the project work, once sufficiently many data are entered into the database from effluent reports in order to have the years of interest be covered in the database. A future year can be selected, if a forecast of nuclear power operation is available. It is intended to implement an update of the fifty-two year scenario established by Cliff Singer on regional and global installed nuclear generating capacity (Singer 1998).

Further refinements planned are making use of even more realistic data. Instead of using the installed reactor power as a parameter for scaling, the real power generated will be applied by using the annual capacity factor as reported to the Power Reactor Information System (PRIS) of the IAEA. The PRIS also contains data on reactor shut-downs, which will be utilized at a later stage as well, as many observations of elevated levels of radioxenon concentrations are due to exceedingly large emissions lasting for several hours or even days associated with the shutdown or restart of a power plant. With all data mentioned in this paragraph, the emission inventory can be prepared for specific years with high accuracy and good time resolution.

Naval reactors on both nuclear powered aircraft carriers and submarines contribute to the radioxenon inventory. The data for these sources is not yet available but shall be considered in future refinements. It is assumed that these vessels contribute to the radioxenon inventory by batch gaseous discharge while in port and liquid batch discharge while at sea. The discharge while in port during maintenance will have the greatest impact on the radioxenon inventory as it will be a gaseous batch discharge, and because reactor maintenance is only carried out at a few shipyards for all nuclear vessels, it will involve the discharge of several reactors in a localized area. Currently there are approximately 160 nuclear vessels operated worldwide. The United States operates half of those with the remainder being operated by Russia, England, France, and China.



Fig. 12: Locations of reprocessing plants, the sources of krypton-85.

Similar Work Done before on Krypton-85

In order to provide an idea of what kind of results should be achieved with the xenon emission inventory as input to a global atmospheric transport model, a similar study done on krypton-85 is summarized here (Winger et al. 2005).

A global-scale krypton-85 emission inventory has been compiled covering the period from the beginning of anthropogenic emissions in 1945 until the year 2000. For a map of all global sources see Fig. 12. Annual emissions for each reprocessing facility have been gathered from the literature. Estimates were made where no release data were available. According to this data set, 10,600 PBq of krypton-85 had been emitted globally until the end of 2000. Taking radioactive decay into consideration, the global atmospheric inventory at the end of the year 2000 can be assessed to have been 4,800 PBq.

These emission data have been used as input into an atmospheric general circulation model (ECHAM4) as point sources to calculate the global krypton-85 distribution for the years 1945–1998. The simulated concentration was studied at the sites of atmospheric krypton-85 monitoring (see Fig. 13). In order to study the impact of specific emitters on the krypton-85 distribution, different tracers were used for different emissions. To lower the number of tracers some emissions were combined within a specific region. The resulting monthly mean atmospheric krypton-85 concentrations (see Fig. 14 for two examples) have been compared to measurements taken at the observation sites. At each observation site the temporal evolution of the observed and calculated concentrations as well as the contributions from different emitters and the variability of the concentrations have been analyzed. High variability has been considered as an indicator for influence of sources close to the receptor site. Based on this comparison the quality of the inventory has been assessed. The calculated concentrations are found to give reasonably good agreement with the observations, indicating that the krypton-85 emission inventory is reliable and realistic.

In a similar way as for krypton-85, the xenon emission inventory will be used to simulate the background concentration at the radioxenon monitoring sites identified for CTBT verification. First observations are available from the International Noble Gas Experiment (INGE) to allow for a comparison between simulations and measurements.

<u>180°</u>	150°	120°	90°	<u>60°</u>	<u>30°</u>	በ የ	30°	<u>60°</u>	90°	120°	150°	_180°
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Fig. 13: Location of ground-based stations for atmospheric krypton-85 monitoring.

Possibility for Comparison of Simulations with Observations of Radioxenon

The radionuclide monitoring network for the CTBT will consist of two types of sensors: eighty particulate stations and forty noble gas systems collocated with particulate stations (Kalinowski and Schulze 2002). Their locations are marked on the map in Fig. 15. Though particle filter gamma-spectroscopy is a well established method, its drawback is the very low probability of particulates being emitted from underground nuclear explosions. It is much more likely that gaseous effluents are released, radioxenons in particular (Kalinowski 2005).

The number of forty noble gas systems is a compromise after some delegations were hesitant during the Geneva negotiations to agree to this technique at all. Doubts were mainly due to the complicated measurement technology that had been applied in a few specialized laboratories but never in the field before. Another reason for questioning the feasibility of noble gas monitoring is the abundance of radioxenon in the atmosphere due to nuclear reactors that could potentially flood the system with an intolerably large number of false alarms. According to the Geneva compromise, experience to be gained at the selected forty sites will help to evaluate the capabilities of noble gas monitoring (Auer et al. 2004; Bowyer et al. 2002). After entry into force of the CTBT the Conference of States Parties will have to decide whether the number of noble gas systems should be increased to eighty.

Atmospheric radioactivity is the only possible evidence that a suspected explosion has in fact been a nuclear one. For the CTBT, the list of relevant particle-bound fission and activation products is long and includes barium-140, lanthanum-140, zirconium-95, as well as anthropogenic radioisotopes with other legitimate sources like cesium-137, iodine-131 and technetium-99m. There are four CTBT relevant noble gas isotopes, Xe-135, Xe-133m, Xe-133 and Xe-131m. The selection of these isotopes as indicators is based on their production rate in an explosion as well as on their half-life (De Geer 2001). The particulate and noble gas sensors are taking daily samples. A high sensitivity of 30 Bq/m³ for Ba-140 and 1 mBq/m³ for Xe-133 can be achieved. Isotopic activity ratios can be used to determine the explosion time and to discriminate between a nuclear explosion and nuclear reactor sources (Kalinowski et al. 2005).



Fig. 14: Simulated monthly mean atmospheric krypton-85 concentrations.

There is still no radioxenon monitoring station operational for the CTBT. However, the first experimental stations are undergoing testing as part of the International Noble Gas Experiment (INGE). These are located at Guangzhou (China), Ottawa (Canada), Schauinsland (Germany), Spitzbergen (Norway), Stockholm (Sweden), Papeete (Tahiti), and Yellowknife (Canada). Further stations are being set up and will soon start taking and analyzing air samples on a daily basis. These stations have a time resolution of eight, twelve, or twenty-four hours. This is much better than the current standard of taking weekly samples and it will be possible to resolve passages of fresh plumes with sharp concentration gradients. This time resolution will allow the application of methods for atmospheric transport modeling in order to determine the source location.

In the northern hemisphere today the Xe-133 atmospheric background activity concentration level is approximately a few mBq/m³. In the southern hemisphere the mean atmospheric activity concentration of Xe-133 is well below the detection limit of the existing systems of $< 1 \text{ mBq/m^3}$ (Stocki et al. 2005) at most locations. The other three isotopes have concentrations that are typically one order of magnitude lower than Xe-133 concentrations or even less. Since the minimum detectable concentrations of all isotopes are on the order of 1 mBq/m³, most of the time they are below the detection limit. However, it is not yet known how much lower they are and what kind of fluctuations they undergo. They are expected to be fairly dynamic due in large part to batch releases and the many point sources as well as due to the radioactive decay with relatively short half-lives. These are: 9.10 hours for Xe-135, 2.19 days for Xe-133m, 5.25 days for Xe-133, and 11.9 days for Xe-131m.



Fig. 15: Network of eighty radionuclide stations for the CTBT International Monitoring System. The forty sites selected for the testing period prior to entry into force are marked by blue squares. The other forty sites are indicated by orange circles.

Since longer time-series of daily measurements are becoming available for various INGE stations, it is now possible to superposition a large number of spectra to determine the average background. This has been successfully done for 295 Yellowknife spectra. As a result, evidence of Xe-131m and Xe-133 is found in a remote Arctic region with an average concentration still higher than one order of magnitude below the detection limit. If this can be confirmed by the planned atmospheric transport simulations with the radioxenon emission inventory, a strong argument can be made in favor of investing into reducing the detection limit by one order of magnitude.

Previous Comparison of Simulations with Observations

The only previous study of atmospheric transport modeling for estimated emissions of radioxenon was done in the late 1980s (Kunz 1989). This study was limited to the most abundant isotope, Xe-133. It was further restricted to a region in upstate New York. The reported annual or semiannual Xe-133 releases of all North American reactors were taken into consideration and regional as well as global dispersion modeling was applied to determine the surface concentration in Albany, NY. For one year (1975) the model estimate was a factor of 1.8 greater than the measured value. For the other years (1981, 1983, 1984) the estimates were less than the measured concentrations by a factor of approximately 1.5. Ambient air concentration of Xe-133 in Europe was reported to be about a factor of five less than at Albany.

Planned Activities

The following activities are planned with results of the global atmospheric model applied to the radioxenon emission inventory:

A.1. Time series comparison of daily measurements

The comparison will be done for each INGE station for which data are available (about ten by the end of 2006) and for each of the four isotopes. In addition, all measured isotopic activity ratios will be compared to the relevant simulated ratios.

A.2. Seasonal average comparison with observations

Ninety spectra will be superimposed for each INGE site to determine average seasonal concentrations. If the related spectral signals are still below the detection limit, longer integrating time frames will be used as appropriate. These will be compared to simulated seasonal averages and standard deviations.

After doing the comparison and improving the simulations as a result of any lessons learned from the comparison, the following results will be generated:

B.1. Global maps of surface concentration isolines for each season and each isotope (four times four maps).

These maps will help to get a visual impression of the impact of the global xenon sources on the regional distribution of xenon concentrations.

B.2. Predicted time series of activity concentration for all eighty CTBT radionuclide monitoring sites and for each isotope.

This will be used to determine typical concentration bands of all relevant xenon isotopes for the CTBT monitoring sites. These typical background concentration bands will be investigated for their potential use in categorizing a particular measurement as typical or anomalous. This is critical for avoiding a high level of false alarms because typical concentration measurements are considered as less sensitive than anomalous concentrations.

<u>B.3.</u> The simulated time series of isotopic activity ratios are entered as chronological paths in the five relevant activity ratio plots that are suitable for source discrimination between nuclear tests and reactor releases.

One example of the five isotopic ratio relationship plots used for source discrimination is shown in Fig. 16 (Kalinowski et al. 2005). Since the ratios will change with time (from one simulation time interval to the next, or from measurement to measurement), one can include the time series of two isotopic ratios as a zigzag curve through the plot. These chronological paths are expected to be found in the realm of reactor releases. The point of interest is to investigate the effect of atmospheric mixing of contributions from different sources and the discrimination margin provided by the distance of the predicted isotopic ratio combinations from the screening line, which can be used to discriminate between reactor emissions and nuclear explosions.

B.4. Simulating additional point releases from hypothetical nuclear explosions.

The question is to what extent the characteristic isotopic ratios of a nuclear explosion are preserved under mixing with ambient concentrations from multiple reactor releases. In the worst case, the background created by constant releases from nuclear reactors blends the characteristic signature of nuclear weapons emissions down into the reactor realm in the isotopic activity ratio plot. The probability of this happening should be assessed. For this purpose the use of "passive tracers" would make sense, helping to determine the impact of each emitter on the signal received at a receiver. Alternatively the calculation of source-receptor sensitivities could serve this purpose. Certain scenarios could be assumed, e.g. a release from Nevada with characteristics like the underground tests conducted in the 1960s to 1980s (Kalinowski 2005). Other scenarios of interest would be hypothetical releases from sensitive countries like India, North Korea, or Pakistan.



Fig. 16: Isotopic activity ratio plot for source discrimination adapted from Kalinowski et al. (2005). The blue curves are the isotopic ratios characteristic for nuclear explosions as they change with time. They never cross the red screening line. The green curves are following the changes of the isotopic ratios as generated in a nuclear reactor over the course of its three-year operation cycles. The green dots are atmospheric observations. One could connect them with a line along their chronological sequence. That produces a zigzag curve that would be interesting to study.

The next step would be to study real cases of detecting elevated levels of radioxenon concentrations at INGE sites.

<u>C.1. First the global modeling approach described above must be combined with a regional model having a sufficiently high spatial resolution.</u>

Validation of the model is possible with known krypton-85 release and observation data in Germany and Japan (Kalinowski et al. 2004; Igarashi et al. 2000).

C.2. Investigate single strong emission cases related to reactor shutdown or startup operations.

There are strong indications (e.g. by isotopic ratios, see Part One) that almost all big signals at INGE monitoring stations are caused by the high emissions resulting from reactor shut-down or start-up operations. These data are available from the PRIS. Atmospheric transport modeling in the inverse mode could be applied to determine possible source areas for the largest signals observed. These areas would be analyzed by correlating them with the location and time of known reactor shut-downs and start-ups. Two methods for confining the possible source region can be applied. Either the release time is estimated, i.e. the atmospheric transport time, from isotopic ratios. This is not very precise. Otherwise, one could determine by isotopic ratios which subsequent samples at the same station or samples at different stations are likely to sense the same plume, i.e. have the same source. One can then apply a correlation to the respective source-receptor sensitivity matrixes. This method is explained in Wotawa et al. (2003).

The project described here is the preparation of a global radioxenon emission inventory. The need for these data arises from the fact that atmospheric radioxenon monitoring will be used for the verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). It is the most important method to detect radionuclide signatures that would be required to determine whether an underground explosion has been a nuclear event. The challenge for this monitoring method is the presence of radioxenon that is emitted under normal operational conditions from more than 500 nuclear reactors in the world. All these sources are estimated here for the first time.

The emission inventory will draw wherever possible from a database of reported reactor effluents. For all reactors for which no reports are available, the estimated emissions will be scaled from reactor design parameters like nominal power or fuel inventory. Nine such parameters are under investigation to determine which one is best suited for predicting reactor emissions. Two selection criteria will be applied. First, the least square fit should be good; second, the dependence of the released amounts of activity should exhibit a strong dependence on the reactor parameter.

The global radioxenon emission inventory will be used as input to global atmospheric transport models to simulate the typical background concentration at the planned eighty sites for CTBT radionuclide monitoring stations. The results will be useful in determining typical vs. anomalous concentrations to limit the number of false alarms. A further concern is that the international community may overlook some future nuclear test, because the release from that test could be masked by mixing with ambient background from nuclear reactor emissions. The results from the global radioxenon emission inventory will be useful for the systematic study of how to prevent such an oversight.

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Sample Output for the Global Emission Inventory (draft, only first rows shown)

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%XENON	COMB_DATA C	OUTPUT FILE		GENERIC	C YEAR							29-Jul-20	005
\$CODE LEGEND: -99 = DATA UNKNOWN ; -88 = PARAMETERS AND DATA UNKNOWN ; 999 = DATA IRRELEVANT ; 9900 = ID FOR ALL COMBINED REACTORS ; 9100 = ID FOR ALL BOILING WATER REACTORS													
8	9260) = ID FOR A	ALL PRESSURE W	WATER/RBMK REA	ACTORS ; 9345	= ID FOR ALI	L OTHER POWER	REACTORS ; 90	010 = ID FOR A	ALL RESEARCH	REACTORS ; -1	= PARAMETE	ERS KNOWN DATA UNKNOWN
8	% NUMBERS AT END MEAN THE FOLLOWING: 1 REAL EMISSIONS DATA/REAL BATCH DATA ; 2 ESTIMATED EMISSIONS/REAL BATCH DATA ; 3 REAL EMISSIONS DATA/ESTIMATED BATCH DATA												
90				6 EST	IMATED EMISSIC	ONS DATA/ESTIN	MATED BATCH DA	ATA ; POSITIVE	E NUMBERS IND	CATE ALL DAT	A WAS SPECIFIC	C, NEGATIVE	IF SOME WAS
				NONSI	PECIFIC								
8	LOCATION		CONTINUOUS H	MIS (Gbq/year	r)		BATCH EMIS	(Gbq/year)			(1/y)	(hours)	
%ID	LAT	LON	Xe-131m	Xe-133	Xe-133m	Xe-135	Xe-131m	Xe-133	Xe-133m	Xe-135	FREQUENCY	DURATION	
9900	999.000	999.000	7.302E+04	5.935E+05	2.251E+04	7.011E+05	2.894E+04	2.020E+05	3.868E+04	5.653E+04	132.4565	12.3582	999
9100	999.000	999.000	5.928E+04	3.487E+05	3.982E+03	6.521E+05	2.799E+02	3.321E+03	4.121E+01	2.939E+02	2.0000	142.6667	999
9260	999.000	999.000	1.268E+04	1.867E+05	1.461E+04	4.083E+04	2.481E+04	1.935E+05	3.325E+04	4.992E+04	138.3864	12.2726	999
9345	999.000	999.000	1.070E+03	5.811E+04	3.917E+03	8.173E+03	3.850E+03	5.148E+03	5.390E+03	6.316E+03	-1.0000	-1.0000	999
9010	999.000	999.000	1.500E+01	3.000E+01	4.500E+01	6.000E+01	7.500E+01	9.000E+01	1.050E+02	1.200E+02	-1.0000	-1.0000	999
2	39.810	-5.700	2.165E+01	9.157E+03	2.501E+01	3.053E+02	9.909E+00	7.079E+03	2.254E+01	1.960E+02	-1.0000	-1.0000	-3
4	-23.010	-44.470	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
8	35.230	-93.230	0.000E+00	0.000E+00	0.000E+00	0.000E+00	0.000E+00	7.062E+02	0.000E+00	5.565E-01	-1.0000	-1.0000	3
10	40.170	44.130	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
12	41.200	0.570	1.294E+01	4.708E+03	1.602E+00	2.706E+02	5.921E+00	3.640E+03	1.443E+00	1.738E+02	-1.0000	-1.0000	-3
14	-34.000	-59.170	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
16	51.920	47.370	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
22	55.750	12.920	8.964E+01	4.680E+03	3.316E+02	9.178E+03	3.600E-01	9.362E-01	3.317E-02	1.074E-01	-1.0000	-1.0000	-3
24	40.620	-80.430	3.625E+01	7.429E+02	0.000E+00	7.235E+01	9.308E+00	6.965E+02	1.306E+01	6.443E+01	41.0000	23.1837	1
28	47.510	2.880	1.218E+00	5.578E+02	1.336E-01	2.256E+02	5.577E-01	4.312E+02	1.204E-01	1.449E+02	-1.0000	-1.0000	-3
30	56.850	61.320	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
33	51.680	-2.500	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
35	47.570	8.230	0.000E+00	1.382E+03	0.000E+00	1.188E+03	0.000E+00	1.068E+03	0.000E+00	7.624E+02	-1.0000	-1.0000	-3
37	49.720	8.420	8.665E+01	1.110E+03	1.534E+01	1.241E+02	3.966E+01	8.584E+02	1.382E+01	7.970E+01	-1.0000	-1.0000	-3
39	45.360	-85.190	0.000E+00	0.000E+00	-1.0000	-1.0000	-3						
40	68.050	166.450	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
45	43.600	51.280	3.500E+02	7.000E+02	1.050E+03	1.400E+03	1.750E+03	2.100E+03	2.450E+03	2.800E+03	-1.0000	-1.0000	6
46	48.500	17.680	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
51	51.430	3.700	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
52	51.730	0.900	-8.800E+01	-8.800E+01	-88.0000	-88.0000	6						
54	41.240	-88.230	0.000E+00	1.309E+03	0.000E+00	0.000E+00	2.286E+00	2.127E+02	1.125E+00	4.160E+00	340.0000	1.6360	1
56	53.850	9.350	7.906E+01	1.758E+03	8.298E+00	1.179E+02	3.619E+01	1.359E+03	7.477E+00	7.571E+01	-1.0000	-1.0000	-3
57	34.700	-87.120	0.000E+00	8.832E+04	0.000E+00	5.200E+01	0.000E+00	1.767E+01	0.000E+00	6.085E-04	-1.0000	-1.0000	-3
60	44.330	-81.600	1.004E+02	1.768E+04	5.520E+02	9.720E+02	0.000E+00	0.000E+00	0.000E+00	0.000E+00	-1.0000	-1.0000	-3
68	53.920	9.120	3.000E+01	6.485E+02	1.294E+01	2.234E+03	1.205E-01	1.297E-01	1.294E-03	2.613E-02	-1.0000	-1.0000	-3