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Simulations of atmospheric krypton-85 to assess the detectability of clandestine nuclear reprocessing

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Abstract.

The results of this study were achieved in the project "Simulation of Atmospheric Noble Gas Concentrations to Assess Sampling Procedures for the Detection of Clandestine Nuclear Reprocessing" (IAEA GER 1643) in the joint programme of IAEA and Federal Government of Germany. In the first year of the project the detectability of additional krypton-85 sources was investigated using atmospheric transport modelling. Krypton-85 is released into the air during reprocessing of spent nuclear fuel rods. Therefore the krypton-85 signature can possibly be used for the detection of undeclared plutonium separation.

First, the global krypton-85 background produced by known reprocessing facilities from 1971 until 2006 was simulated with the atmospheric general circulation model ECHAM5 using annual emission data. The model results were evaluated by extensive comparison with measurements performed by the German Federal Office for Radiation Protection. Of particular interest for an assessment of the detectability of unknown sources is the background variability. The variability of concentrations is very high over central Europe, where the large reprocessing plants La Hague and Sellafield are located, and it is very low on the Southern Hemisphere, where no nuclear reprocessing takes place. The analysis of concentration time series on various time scales allows partly a distinction between fluctuations caused by the variability of the sources from variations due to atmospheric dynamics. Furthermore the detection sensitivity to a set of arbitrarily specified source locations is analysed with a Lagrangian particle dispersion model. This, in combination with the location specific background variability, is giving first benchmarks on the capability of using krypton-85 for IAEA Safeguards based on the Additional Protocols foreseeing environmental sampling.

1. Introduction

In spite of the broad range of well approved measures for safeguarding known nuclear facilities and materials, there are still deficits in detecting undeclared activities. In the Additional Protocol, environmental sampling is generally foreseen. While location specific sampling is already applied, wide area environmental sampling is not approved so far.

Krypton-85 is a radioactive noble gas which is built up during reactor operation in the nuclear fuel rods. With its half-life of 10.8 years it remains captured until the fuel rods become destroyed and dissolved during reprocessing. Then it is released to the atmosphere. Therefore the krypton-85 signature is potentially suitable for the detection of clandestine nuclear reprocessing activities. The suitability has already been shown in case studies around the Karlsruhe pilot reprocessing plant [1] and the Tokai reprocessing facility [2]. The retention of the ^{85}Kr from the off gas stream is only possible with a huge technological effort. Nevertheless, earlier assessments disfavoured the use of krypton-85 signature for long range detection ([3],[4]). This project is making use of advanced atmospheric transport models and does not aim for a permanent measurement network covering the whole globe. The sampling scenarios under consideration are regionally and temporally limited sampling networks, spot sampling wherever inspections take place, or flexible sampling anywhere in the country ("catch the plume"-approach).

The feasibility and costliness of the inspection scenarios is investigated in other parts of the project. A new measurement technique for the determination of the ^{85}Kr content of small air samples is under development at the Centre for Science and Peace Research at University of Hamburg, the atomic trap trace analysis (ATTA). The first project year of the task GER 1643 (2008) and this article focus on the assessment of the general detectability of additional krypton-85 sources in three major steps:

1. Simulation of the global background distribution caused by known sources
2. Investigation of the location specific variability of the background concentrations in order to develop benchmarks for the necessary strength of additional signals to be detectable
3. Quantification of detetectability of additional sources modelling hypothetical release scenarios in various regions.

This is a short overview article with special focus on the detectability results. The detailed results of the study with full evaluation of the global background simulations can be found in the first year project report [5] and the related PhD thesis [6].

2. Global Background

For the assessment of the detection potential looking for the ^{85}Kr signature of a clandestine facility the knowledge of the ambient background concentrations should be as precise as possible. For this study a long term simulation of the global background from 1971 to 2006 was performed. The model results were evaluated by comparison with measurements and the variability of the background concentrations was investigated.

2.1 Emission Inventory

The main emitting sources of ^{85}Kr are nuclear reprocessing plants. The emissions of normal reactor operation are negligible small. For the global background assessment an emission inventory was used. The inventory published by Winger [7] was extended and updated by Ahlswede [8]. The data can also be found on the website www.igse.net. The global ^{85}kr background increased from 1945 on. Figure 1 shows the total activity amount in the atmosphere and the yearly emissions according to the inventory.

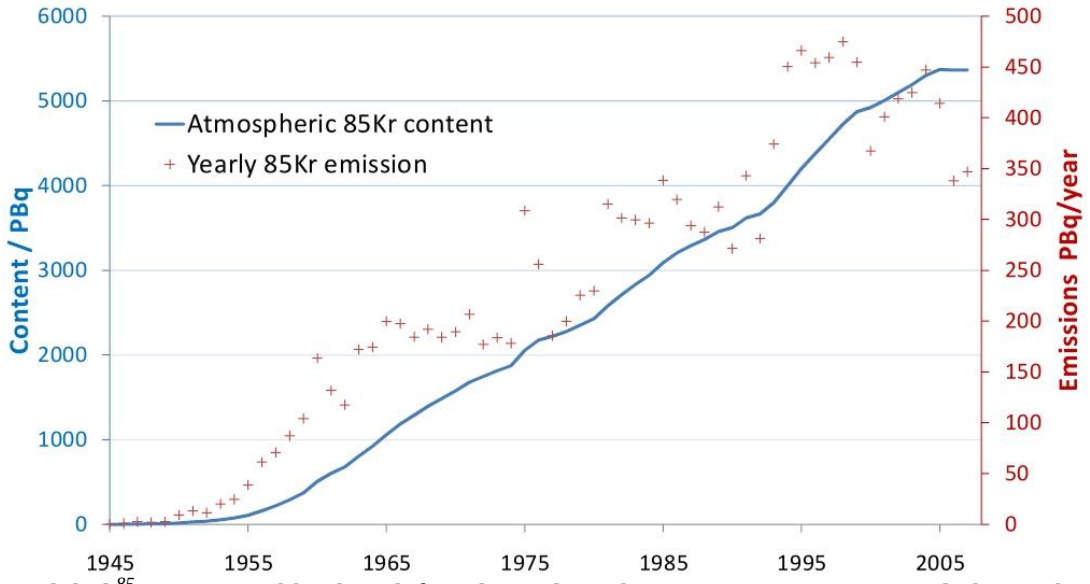

Figure 1 Global ^{85}Kr content (blue line, left scale) and yearly emission rate in PBq (red plus, right scale)

2.2 Global Background Simulation

For the simulation of the global background, the general circulation model ECHAM 5 was used in a modified version guaranteeing tracer mass conservation. The reprocessing facilities listed in the emission inventory were implemented as point sources to the model. The emission strength was assumed to be constant in time in lack of knowledge of exact emission time patterns. At least for the industrial facility La Hague this assumption is justified, as they operate rather continuously [9]. As best compromise between accuracy and computational effort a spectral resolution of T63 with 31 vertical levels was chosen. This corresponds to an horizontal grid width of ca. 200 km. The model time step was 12 minutes and the model was "nudged" with ERA-40 reanalysis data in order to constrain the large scale dynamics to the real weather situations which occurred in the simulated period from 1971 to 2006. The initial distribution was derived from the model results in [7]. Figure 2 shows two examples for the simulated annual mean concentration at surface. While in 1976 the main emissions came from the SU, in 2006 the reprocessing plant La Hague in France is the by far strongest source and the general concentration level increased.

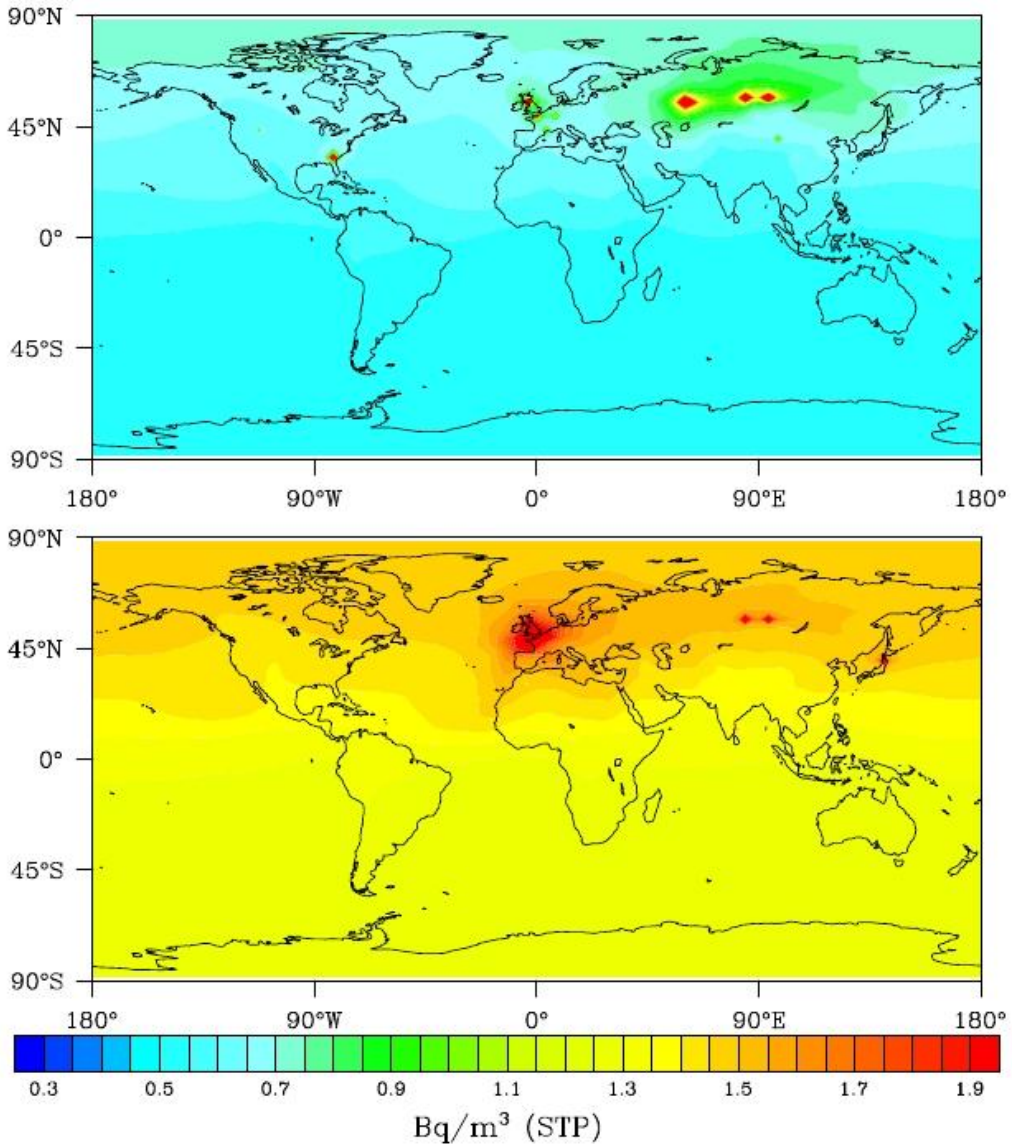

Figure 2 Annual mean concentration of krypton-85 at surface for 1976 (top) and 2006 (bottom)

2.3 Evaluation of model results

The results of the global background simulation were evaluated by comprehensive comparison with available measurements. The German Federal Office for Radiation Protection operates a network with stations observing ^{85}Kr concentrations in weekly samples all over the world. As example figure 3 shows the comparison of weekly measurements with corresponding model results for Vienna. The baseline concentration level is well represented and even the variability is in the right order on the weekly scale in this case. The observed values (crosses) exceeding the simulated concentration range are probably caused by fresh plumes not fully resolved in the coarse global model. Overall the model results were compared at 14 observation stations and with several ship and balloon measurements. The statistical evaluation has shown errors in the range of few percent and stay within the accuracy of the emission data. Only in the direct vicinity of sources the global model predicts to low concentrations as the tracer is immediately distributed over the whole grid box and therefore the numerical diffusion is stronger than that of the real plume. With sources located exclusively on the Northern Hemisphere, ^{85}Kr is an ideal tracer to study the air mass exchange between the hemispheres. Results on the investigation of interhemispheric exchange can be found in [6]

2.3 Background variability

For the detection of additional signals caused by unknown sources the knowledge of the background variability is crucial. In this study the location specific variability of the ^{85}Kr concentrations was derived from the global simulation results. As measure for variability the standard deviation of the 6-hourly values

was determined for each grid cell. Figure 4 shows this standard deviation for the month December in 2005. The variability is very high close to the sources and very low on the Southern Hemisphere, where no known reprocessing facilities are operating. This kind of background variability map was generated for each month in 2005 and is used in the following detectability study for defining the threshold for an additional signal to be detectable.

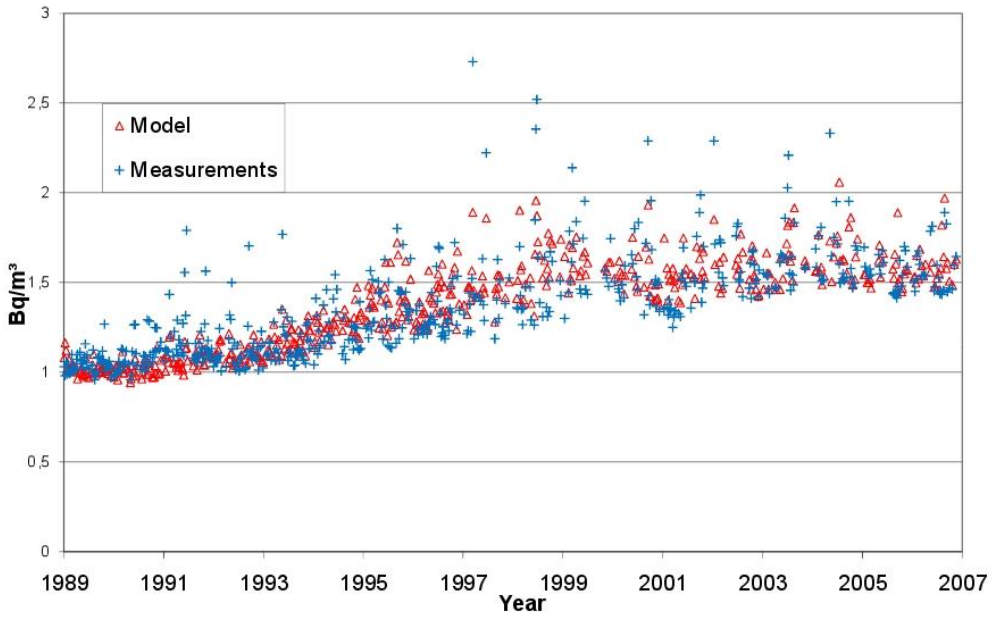

Figure 3 Comparison of weekly means measured in Vienna with the corresponding simulated concentrations

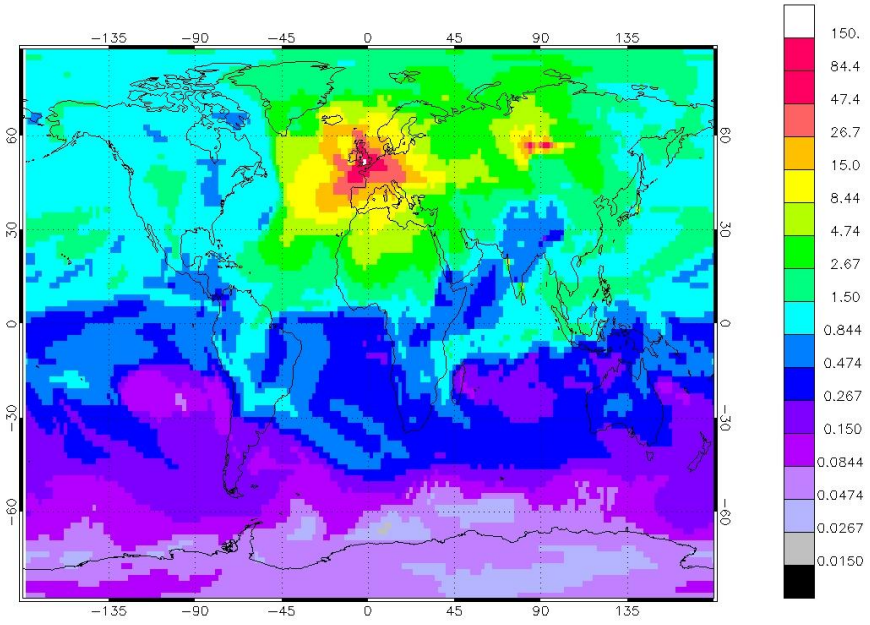

Figure 4 Standard deviation of the 6-hourly concentrations in percent of the monthly mean for Dec 2005

3. Detectability of additional sources

A source hypothetical location setup was defined comprising 58 stations globally grouped in 4 clusters: Atlantic/South America, Europe/Africa, Central-East, and Far East/Australia. Their geographical distribution is shown in Figure 5.

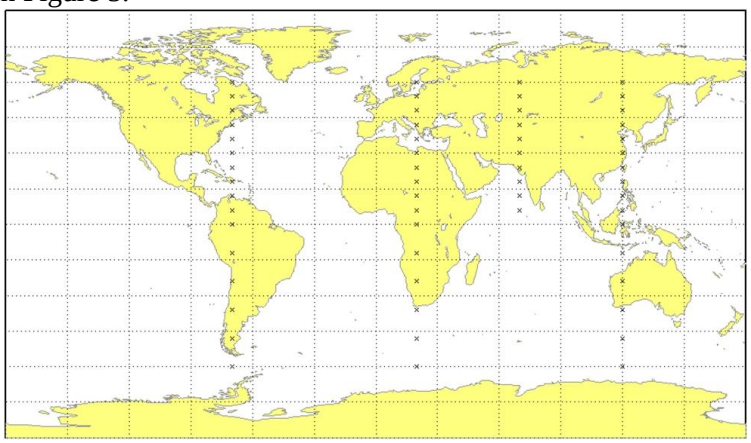

Figure 5 Geographical distribution of hypothetical release points in longitudinal clusters

3.1 Simulation setup

The Lagrangian particle dispersion and trajectory model HYSPLIT [10] is used for this study. As driving data, gridded meteorological data from the Global Data Assimilation System (GDAS) archive hosted by NOAA's Air Resources Laboratory for the year 2005 has been used. For each source location a constant ^{85}Kr emission signal of 6 hours has been assumed released at an elevation of 10 meters above ground. The propagation of the concentration plume for the subsequent 144 hours has been simulated. Every 6 hours a concentration output was stored, averaging concentration levels of the preceding 6 hours. The starting time of emissions was 9:00 a.m. at the 8th day of each month in 2005 except for the months January and March where the starting time was set to 9:00 a.m. at 15th because of missing meteorological data. A number of 16704 data fields have been generated; thereof 8532 were used for the analysis. The difference is due to plumes that left the model area within the simulated time because of high wind velocities. The first three days after release which are of particular interest for the detection of small sources are not affected by this effect of plumes leaving the model area.
For the assessment of detectability the concentration maximum in the plume was evaluated for each stored time step. The criterion for possible detection in this model study was that the concentration in question has to be at least three sigma of the background concentrations of the month in question. Thus, only if the potentially measurable signal exceeds three standard deviations of the background concnentration, the signal is assumed to be detectable. For the variability the standard deviations of the monthly means from ECHAM5 were taken. The values represent the variability of the instantaneous six hourly surface concentrations in the global model. As the specific time patterns of the emissions from reprocessing plants were not included in the global model, the variability may be higher in regions directly influenced by active background sources. In the HYSPLIT runs, standard emission strength of 1 unit was assumed. It is easily possible to scale the source strength linearly according to a specific expected concentration. For that, the quantity of the minimum detectable release was defined: The minimum detectable release (MDR) is three times the location specific standard deviation of the global background divided by the local concentration output of HYSPLIT and corrected for a scale factor accounting for the emission pulse length.
That way of definition of MDR implies that low sensitivity leads to high minimum detectable releases and high sensitivity For the analysis three thresholds for the minimum detectable release are of special interest for the application: A release of

• 3.2 TBq according to the emission scenario for one dissolution campaign out of fifty campaigns during one year for retrieving one significant quantity plutonium (8 kg).
• 10 TBq assuming a more active plant.
• 100 TBq corresponding to the emissions of an industrial facility.

For comparison it has to be considered that daily emissions of the reprocessing plant La Hague exceed occasionally 1000 TBq and have daily mean values of several hundred TBq (e.g. 650 TBq per day in 2007).

3.2 Results on Detectability

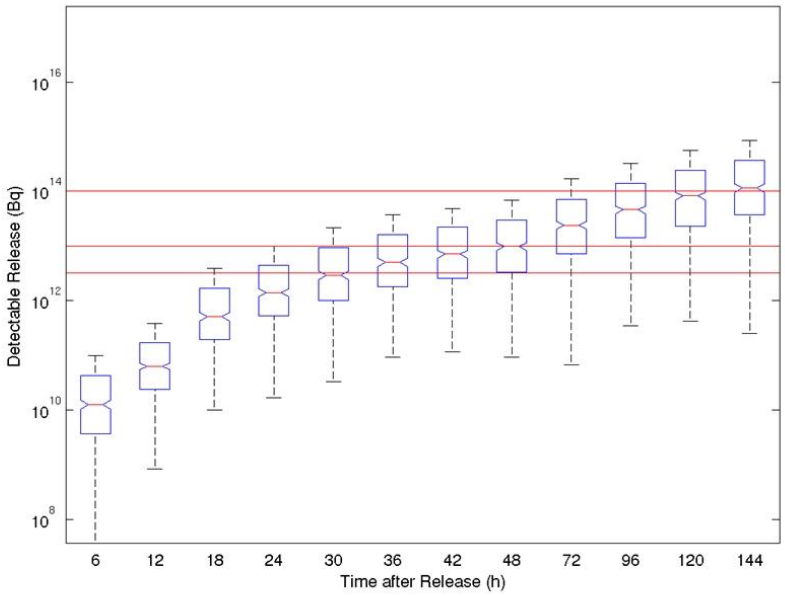

Figure 6 Evolution of minimum detectable release in time

The box plot in Figure 6 shows the evolution of the minimum detectable release in time. The median and the lower and upper quartile are indicated by the boxes. Thus, for example a release of 3.2 TBq was detectable in nearly half of the cases after 30 hours. About 25% of the 3.2 TBq releases are still detectable 48 hours after release. The time passed after release is clearly a critical factor for the detection as after 72 hours the releases have to be bigger than 3.2 TBq to remain detectable. Figure 7 shows the detectable release in dependence of the distance of the plume centre, i.e. the maximum concentration in the plume, from source. The vertical lines are at 100 and 500 km distance.

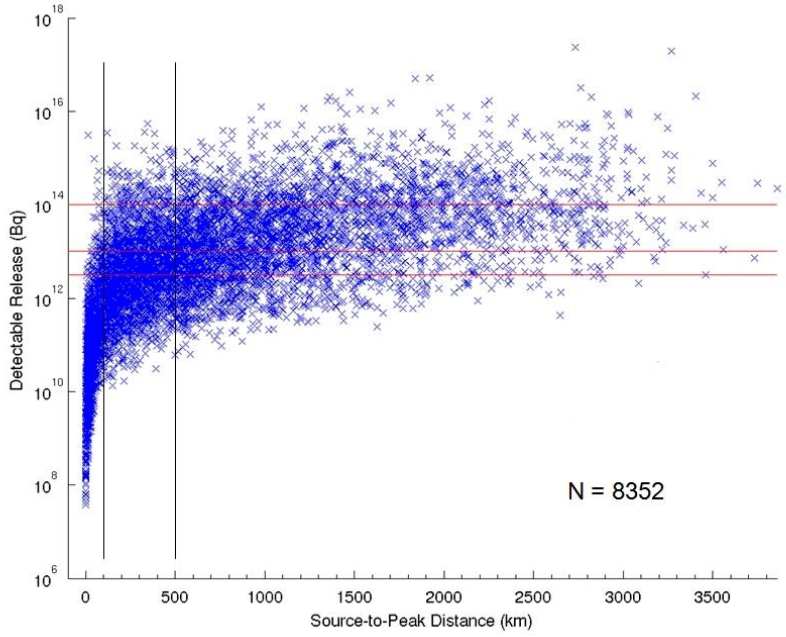

Figure 7 Minimum detectable release in dependence on plume travel distance from source

Under very suitable conditions, a plume can still be detectable at more than 1000 km distance from the source. For most cases, the detectability is limited to 500 km distance. The shape of the maximum of a plume is in general occurring as a rather broad peak so that small deviations from the plume center affect the detectability only marginally. Table 1 summarizes the detectability for the considered regions and emitted activities concerning detectability after 24, 48 and 72 hours after stop of release. As expected the results show large differences between the hemispheres and depend on the distance to western European background sources. The results on the Southern Hemisphere are quite promising with detection of even the small release of 3.2 TBq in 93% of the analyzed cases 24 hours after end of a 6-hour release and still more than 50 percent after 48 hours. However, at 20_East, Northern Hemisphere, only 100 TBq releases reach high detection probabilities. Remarkable is the reduction of potential detectable situations between 24 and 48 hours after release.

Table 1 Percentage of cases fulfilling the detection criterion for the three reference emission quantities 24, 48, and 72 hours after stop of release listed per region (100% corresponds to a sample 120 plumes on the northern and 72 plumes on the southern hemisphere).

LON	Time	3.2 TBq	10 TBq	100 TBq
Northern Hemisphere				
70°W	24 h	71%	93%	100%
	48 h	8%	50%	90%
	72 h	3%	19%	79%
20°E	24 h	23%	55%	97%
	48 h	3%	8%	77%
	72 h	1%	4%	45%
70°E	24 h	71%	94%	100%
	48 h	14%	48%	94%
	72 h	5%	24%	88%
120°E	24 h	72%	93%	100%
	48 h	20%	45%	95%
	72 h	10%	29%	81%
Southern Hemisphere				
70°W	24 h	89%	97%	100%
	48 h	54%	78%	93%
	72 h	39%	62%	90%
20°E	24 h	93%	99%	100%
	48 h	51%	75%	94%
	72 h	25%	54%	83%
120°E	24 h	93%	100%	100%
	48 h	49%	76%	97%
	72 h	25%	54%	90%

4. Conclusions

The simulation of the long term global background distribution was very successful as the model evaluation has shown. For the first time a location specific variability of ^{85}Kr concentrations has been derived. Concerning the detectability of unknown sources above that variability, there is a considerable chance to detect even small emissions when measuring in a plume quite soon after its emission. The results show that in particular for the detection of a 6-hour release of 3.2 TBq, the smallest reference release, the measurement should take place within 24 hours after the stop of release. On the Southern Hemisphere the chances of detection are higher because of the less variable background situation. The release on industrial scale of 100 TBq in 6 hours stays detectable for more than two days under most conditions. Although the probabilities for definitive detection of small releases are limited in the current situation, there would be a deterrent effect of the possibility of detection. Assuming a method with a detection probability of only p=5% for a single plume leads to considerable chances of detection when applied over a longer period. Thus in the reference scenario of 50 plumes the probability of at least one detection is $1-(1-p)^{50} = 92\%$. Of course this requires a very low false alarm rate. Overall the study has shown that state-of-the-art atmospheric transport models can support the detection of a krypton-85 signature. Their capability in terms of localizability of unknown sources is investigated in further parts of the project. Besides that, the costs of various inspection procedures have to be investigated.

Acknowledgments

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