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Detecting clandestine plutonium separation activities with krypton-85

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Abstract

Krypton-85 is released from nuclear reprocessing plants during the separation of plutonium, which has produced a global atmospheric Kr-85 background. Due to this background, it may be difficult to use Kr-85 to detect emissions from clandestine separation activities. In order to draw meaningful conclusions from measured atmospheric levels of Kr-85, it is necessary to know the history of the local variability in atmospheric Kr-85 concentrations. In this work, atmospheric transport modeling is applied to simulate the emissions from known reprocessing plants to determine the worldwide background and variability of Kr-85 concentrations. A sampling methodology and detection technologies are presented, along with simulation results for three scenarios involving hypothetical undeclared facilities. Possible policies and techniques for increasing the usefulness of atmospheric Kr-85 detection are also discussed.

1 Introduction

This paper presents a computational model that estimates the magnitude and variability of global atmospheric krypton-85 (Kr-85) background concentrations. Kr-85 is a radioactive fission product that is released from spent fuel rods as they are disassembled during routine nuclear reprocessing activities. As Kr-85 is a noble gas, it is technically difficult to separate and contain. As a result, it is typically released into the atmosphere through stacks at the reprocessing facility.

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Nuclear reprocessing facilities are of particular importance to safeguards, as these plants can be used to separate plutonium from spent nuclear fuel. This plutonium can be either reused in reactor fuel, or possibly used as material in a nuclear weapon. The potential misuse of reprocessing technology can take place either at a safeguarded facility that has been declared to the International Atomic Energy Agency (IAEA), or at an undeclared facility. In accordance with the IAEA's generic safeguards objectives that apply to all states with comprehensive safeguards agreements, the IAEA aims to: 1) detect the diversion of nuclear material at declared facilities, 2) detect any undeclared production or processing of nuclear material at declared facilities, and 3) detect any undeclared nuclear material or activities in a given State [Iae14]. Furthermore, the need to verify the absence of undeclared reprocessing facilities may redouble in the future, if a Fissile Material Cut-off Treaty (FMCT) is concluded.

Undeclared reprocessing facilities could be very challenging to detect. While large-scale reprocessing plants are taking years of planning, building, and testing, experts have noted that existing industrial facilities (such as a winery) could be remodeled to function as "quick and simple" reprocessing plants [Fer77].

Monitoring Kr-85 concentrations via atmospheric sampling, however, could be of use in verifying the absence of undeclared reprocessing facilities. The IAEA already has the ability (given Board of Governors approval) to deploy wide-area environmental sampling (WAES) regimes in most states that have concluded an Additional Protocol to their safeguards agreements. The IAEA has also studied the use of WAES for nuclear safeguards in the past. A three-year study undertaken in the late 1990s by the IAEA and six Member States concluded that atmospheric sampling appeared to be most promising WAES technique (versus soil, aquatic and vegetation sampling), in terms of probability of detection per sample, and that atmospheric concentrations of Kr-85 were a relevant signature of reprocessing activities [Wog10]. While the study's authors cautioned that the cost of operating a WAES network could be high, they also recommended steps that could be taken prior to deployment of a WAES program. According to a summary of the IAEA report, these steps included "further refining evaluation of the variability in background levels of target signatures" [Wog10], which this paper aims to address.

Since publication of the IAEA report, Kalinowski et al. have argued that Kr-85 is the most promising wide-area signature for detecting the undeclared separation of plutonium, based on atmospheric samples collected at various distances from the Karlsruhe Reprocessing Plant between 1985 and 1988 [Kal04]. Furthermore, a joint German-IAEA effort has used atmospheric transport modeling simulations to benchmark detection probabilities for undeclared reprocessing facilities, and characteristics of Kr-85 background levels [Ros10].

2 Atmospheric Background of Krypton-85

Kr-85 has a half-life of 10.7 years, and is estimated to be released at a rate of at least 16 TBq per kg of separated plutonium [Sch15]. Over the last seven decades, a Kr-85 background has built up in the atmosphere due emissions from continuous reprocessing

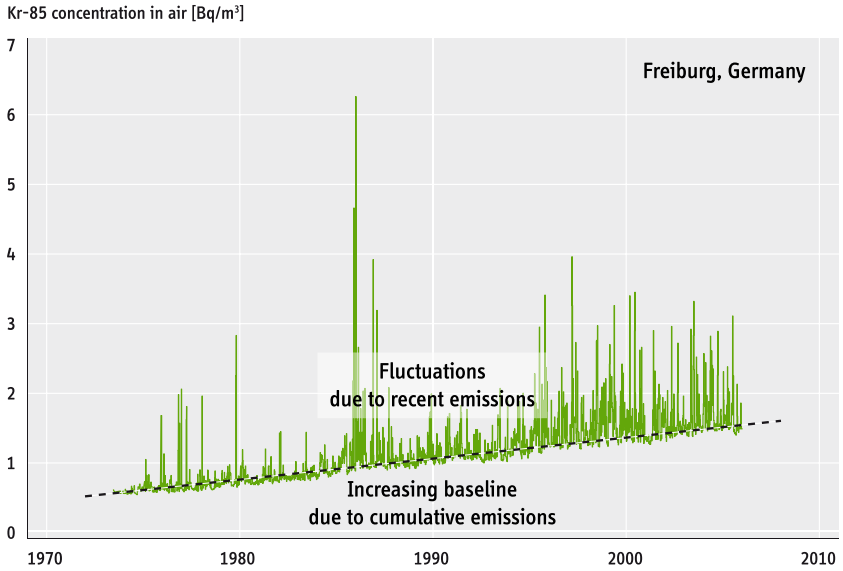


Figure 1: Time series of detected krypton concentration at Freiburg (Germany). [Gfm07] The baseline is due to the long-term global emissions that have accumulated in the atmosphere over decades. The fluctuations from sample to sample are due to recent emissions. In this example the fluctuations are most likely caused by releases from La Hague (France) and Sellafield (UK).

activities, and its long half-life. The long-term development of the background has been simulated from 1945 to 2006 [Ros10]. The average background for the year 2006 has been estimated to be 1.5 Bq/m^3 for the Northern Hemisphere, and about 1.3 Bq/m^3 for the Southern Hemisphere, with strong fluctuations downwind from active reprocessing facilities . The background in the Southern Hemisphere is generally lower than in the Northern Hemisphere, because almost all reprocessing has taken place in the Northern Hemisphere and the exchange mechanisms can take up to a few years to transfer significant air masses between the hemispheres.

Over the last four decades the average baseline increase has been estimated to be 0.035 Bq/m^3 per year [Gfm07], as shown in Figure 1. Reprocessing plants are still separating plutonium today, mostly for civilian nuclear fuel, and thus emitting krypton-85. Once in the atmosphere, the gaseous krypton is dispersed by prevailing winds and gets diluted over time and space. As this distribution takes place, it gets increasingly difficult to distinguish the recently emitted krypton against the background. However, downwind from active reprocessing facilities, the krypton concentration can still show large variability, depending on the size of the releases and the movement of the plume. When taking an air sample to check for unusual krypton concentration, the composition of the sample could be traced back to known reprocessing plants. If time-dependent stack emissions of Kr-85 are known, then expected atmospheric concentrations of Kr-85 at sampling sites could be estimated. If these source terms are unknown, the variability can only be estimated based on average annual emissions.

Table 1: Locations and estimated Kr-85 emissions from active nuclear reprocessing plants for the year 2010. [Ahl09, Gfm10, Gfm11]

Facility	Country	Latitude	Longitude	Kr-85 emissions per year
Lanzhou	China	36.2	103.5	2.24E+14 Bq
La Hague	France	49.4	−1.5	2.26E+17 Bq
Kalpakkam	India	12.3	80.1	1.12E+16 Bq
Trombay	India	19.0	72.6	8.00E+15 Bq
Dimona	Israel	31.0	35.1	5.76E+14 Bq
Tokai	Japan	36.3	140.4	1.00E+15 Bq
Nilore	Pakistan	33.4	73.2	1.92E+14 Bq
Mayak RT-1	Russia	55.4	60.1	4.86E+16 Bq
Zheleznogorsk	Russia	56.2	93.4	1.00E+16 Bq
Sellafield	UK	54.3	−3.3	4.53E+16 Bq

Ten reprocessing facilities were active in the year 2010; their Kr-85 releases have been estimated either based on the throughput of spent fuel or the plutonium separation rate [Gfm10, Gfm11], or on previous estimates [Ahl09]. These simulations and analyses can easily be redone for a different set of sources, different source terms or another time period.

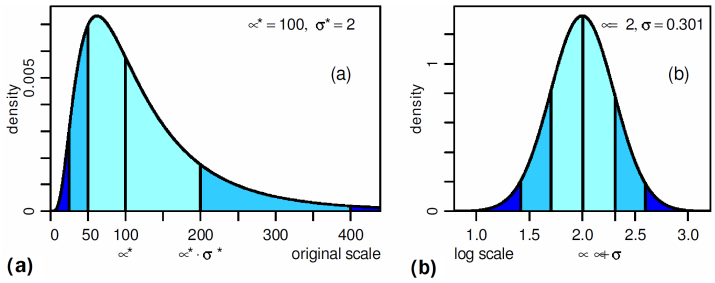
In the following, it is assumed that the Kr-85 background for any given location is composed of two components. The first part is the background baseline that has resulted from decades of reprocessing activities (see also Figure 1). This baseline is assumed to be constant during one year for a given location, though it might be offset between Northern and Southern Hemispheres. On top of this baseline is the local concentration, which varies on a day-to-day basis, depending on recently emitted plumes from active facilities. Emissions from active facilities are dispersed in the atmosphere over time and are considered in the simulation until the dilution makes the concentration insignificant in comparison to the background baseline. In other words, recently formed plumes only have to be considered until they are washed out in the background.

3 Simulation of the Kr-85 Background

For the simulations, constant emissions have been assumed for the aforementioned ten reprocessing plants. Daily emissions from all ten reprocessing plants have been simulated for the year 2010. Each emission was tracked in the simulation for four weeks after release, at which point it was assumed that the plume was dissolved into the background. The atmospheric transport was simulated with the Lagrangian transport and dispersion model Flexpart 8.23, applied together with NCEP meteorological data in 0.5 degree latitude x 0.5 degree longitude resolution. It is noted that taking into account only the recent Kr-85 emissions cannot reflect the meteorological processes on longer time scales, such as the exchange of air masses between the hemispheres, which

Log-normal distributions and confidence intervals

It has been shown previously that concentrations of particles in the atmosphere over time usually do not follow a Gaussian normal distribution [Lim01, Sch14]. This is due to the multiplicative nature of the the dilution process. Processes that involve adding or subtracting tend to lead to normal distributions (Central Limit Theorem), whereas processes that involve multiplicative factors (such as dilution) tend to lead to log-normal distributions. Gaussian distributions can be described by their *arithmetic* mean and the standard deviation, where the standard deviation has the same dimension as the mean and adding/subtracting it to/from the mean gives the known confidence intervals of 68.3%, 95.5% and 99.7% of data around the mean. Similarly a log-normal distribution is described by its *geometric* mean and its standard deviation, where the standard deviation is dimensionless, and the confidence intervals are given my multiplying/dividing the mean with/by the standard deviation. This behavior is shown in the figure below for (a) log-normal and (b) its transformed normal distributions [Lim01]. When setting a one-sigma threshold for identifying unusually high atmospheric concentrations by multiplying the geometric mean with the dimensionless standard deviation, the result (below in the left figure a value of 200) covers about 84.1% of the data.



takes place on a time scale of one year and longer. The feeding of Kr-85 enriched air masses into the southern hemisphere is therefore not reflected in these simulations. This would only be possible, if the emissions and the movement of plumes were simulated over multiple years or even decades. This has been accomplished before for the years 1971-2006 [Ros10], but requires a high input of computational resources and/or a reduced resolution in space and time. The approach presented in this work, on the other hand, is to be understood as a more efficient method that starts from the established baseline and involves less computational resources to estimate the global variability for any choice of source configurations.

For each location on the globe, the concentrations from these recent emissions were stored for every time step of the year. This yields a time series for each latitude and longitude coordinate pair. From this time series the geometric mean and the multiplicative standard deviation were calculated for each location. The resulting confidence interval is plotted on a global map in Figure 2. For more details on the geometrical mean and the multiplicative standard deviation of log-normal distributions, refer to the box on this page. Since all active reprocessing plants are located in the Northern Hemisphere, also the strongest local fluctuations as well as highest differences from region to region are visible there. The effects of the European reprocessing facilities La Hague and Sellafield are clearly visible on the map. Also, the location of the two Russian and two Indian reprocessing facilities can be seen on both maps.

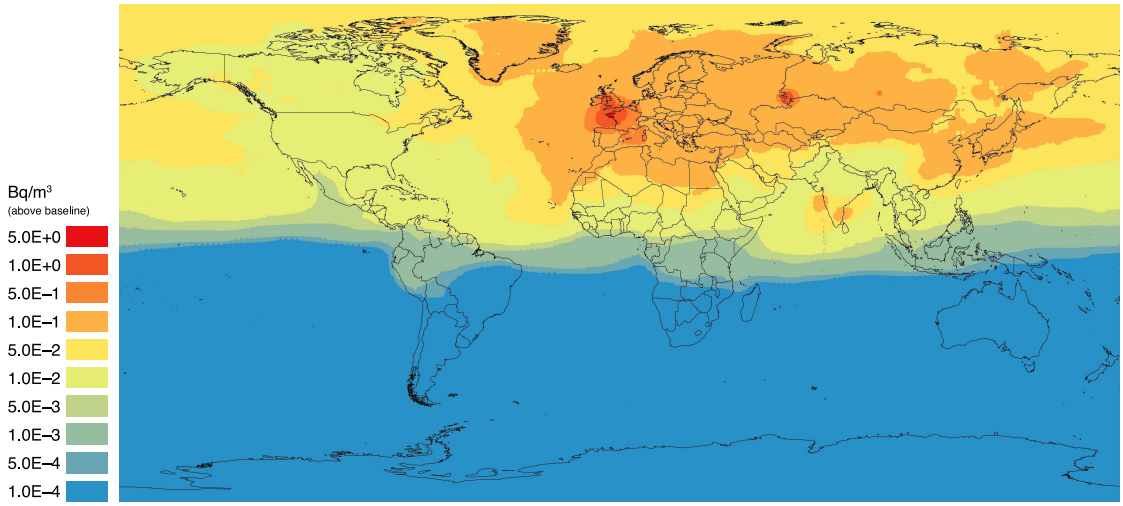


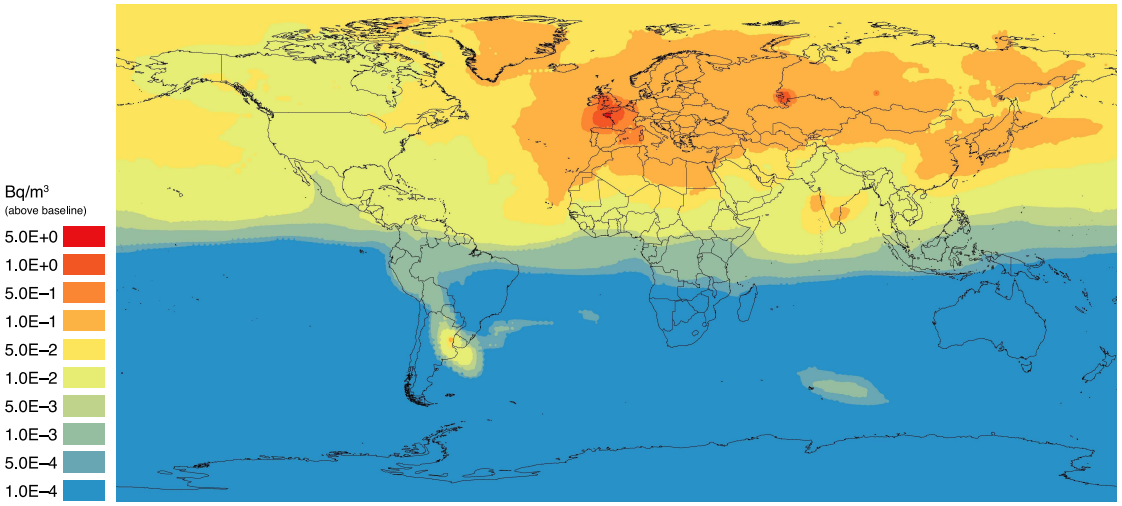
Figure 2: Global map of Kr-85 standard deviations above the local baseline for the source configuration as estimated for the year 2010.

In the mid-latitudes, it takes air masses in the order of one month to circumnavigate the world. The simulation time of four weeks is enough to show the effects of European and Asian reprocessing plants on the Northern Hemisphere including North America. It is apparent that in Western and Central Europe, short-term variations in Kr-85 concentrations constitute a significant share of the total Kr-85 concentrations. In large parts of Europe and Asia the variations would still be detectable above the background baseline. In North America, in regions closer to the equator and especially in the Southern Hemisphere, the variations are significantly smaller compared to the baseline, which increases the vulnerability of clandestine reprocessing activities to detection.

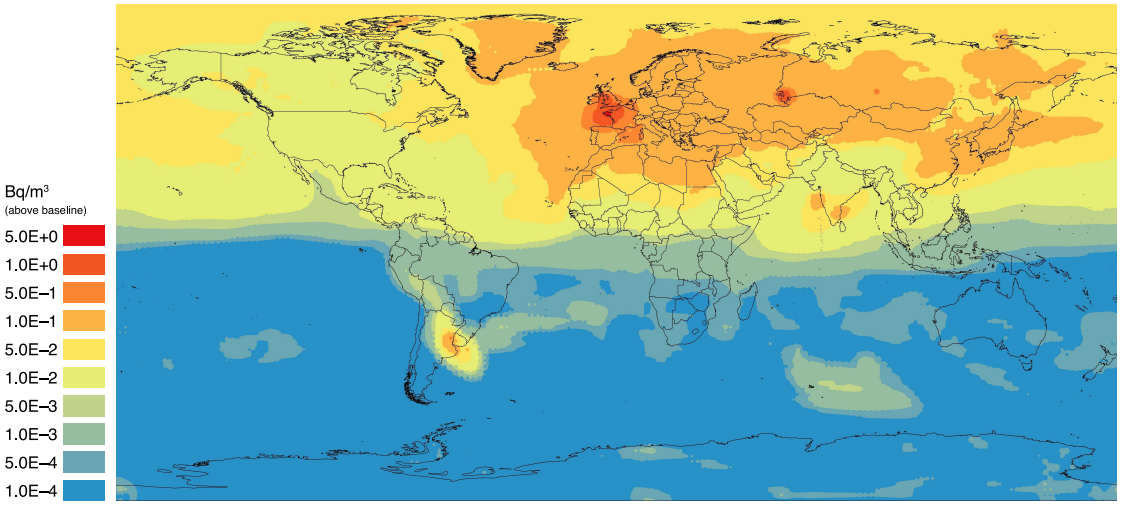
4 Impact of the Global Variability on the Detectability of Undeclared Reprocessing Plants

The global variability of Kr-85 concentration is shown here in a map of its one-sigma confidence interval (see Figure 2, and compare with the explanation in the box on page 5). This data in this map - or alternative versions of it with different source configurations - is the basis for detecting reprocessing activities at undeclared sites.

An approach to detecting undeclared reprocessing facilities could involve taking a number of air samples from random locations at random times, and measuring the Kr-85 concentration of the sample. This idea aims to use mobile sampling instead of ground-based monitoring stations and would require small, transportable sample sizes and short sample processing times. A detector that fulfills these requirements is currently under development in form of an optical trap that can detect Kr-85 down to the single-atom regime [Koh14]. Such samples could be collected airborne with planes or drones, and be analyzed in a laboratory that processes all samples from a region. After a Kr-85



(a) 1 SQ per month.



(b) 1 SQ per week.

Figure 3: Fictitious scenario with an additional reprocessing plant in South America to demonstrate its influence on the Southern Hemisphere. The upper figure shows the effects from a facility that reprocesses one significant quantity per month, the lower figure the effects from a facility with one significant quantity per week. Comparable sources would not be detectable in most parts of the Northern Hemisphere, due to ongoing emissions from existing reprocessing facilities.

concentration is measured, it can be compared to the calculated confidence interval at the sampling location in order to determine the expected likelihood that the Kr-85 originated from known reprocessing activities. Additionally, a backward atmospheric transport simulation could be applied to determine the composition of the collected air.

To demonstrate this approach, three scenarios with fictitious clandestine reprocessing plants have been simulated. First, a reprocessing facility in a low-background region (South America) has been assumed to separate a) one significant quantity per month, and b) one significant quantity per week. The effects on the background are shown in Figure 3. The affected area with a confidence interval at least 10% above the background is calculated to be a =175,000 km^2. Compared with the Earth's total landmass area A =149 million km^2, it is possible to calculate the probability to detect the clandestine facility. When taking n random samples the probability to get a sample with elevated Kr-85 concentration from the clandestine facility is $p = 1 - \left(1 - \frac{a}{A}\right)^n$. For 1000 samples the probability is 69.1%, and for 100 samples it is still 11.1%. However, these calculations are based on a scenario with low background fluctuations.

Emissions from a second fictitious plant located in Central France were simulated, but the effects on the background fluctuations are negligible and not visible on the global map due to ongoing reprocessing activities nearby. A third fictitious plant was placed in the Great Plains in North America. Although there is no active reprocessing plant nearby, the simulations showed that the effect on the background is very small, and Kr-85 emissions would most likely stay hidden in the background due to the strong and steady winds that are typical for this area.

Furthermore, this quick way of calculating the global variability can be used to determine categorization thresholds for monitoring of known facilities. For a given ground-based monitoring station that has been set up to detect emissions from a specific reprocessing plant, the influence from other reprocessing plants must be estimated. For such a case, the declared facility would be taken out of the source dataset in order to determine the local variability resulting from all other known reprocessing plants. The local confidence intervals can then be used to categorize the samples according to the likelihood that the Kr-85 concentration has originated from the monitored plant.

5 Effects of Ongoing Reprocessing

The ability to detect emissions from clandestine reprocessing plants is impeded by both the accumulated background of atmospheric Kr-85 and short-term fluctuations from recent emissions. The accumulated background significantly reduces the range at which emissions are still distinguishable against the noise. The only way to overcome this is to stop further Kr-85 emissions and wait until the existing background decays. Fluctuations in the Kr-85 concentration from recent emissions additionally impede the detectability of clandestine facilities. Downwind from active reprocessing facilities, it is difficult to attribute a measured concentration to declared or undeclared activities. The availability of stack emission data from known facilities would mitigate this issue. However, smaller emissions from clandestine reprocessing plants could still go unrecognized in a sample.

In view of the current Kr-85 background, it is assumed that the effect of an active, upwind facility is successfully detected if the local concentration is 10% above the baseline background. Thus, areas in which the concentration is regularly more than 10% above the background due to known facilities, the ability to detect clandestine facilities is severely inhibited. The total area of landmass in which the variability of Kr-85 is 10% above the baseline is about 60 million km^2. With reference to the total surface area of the Earth, about 149 million km^2, this means that on 40 % of the Earth's landmass the detectability of clandestine reprocessing is not only affected by the high baseline, but also by recent emissions.

6 Conclusions and Outlook

The separation of the background into two components enables the simulation of the global krypton-85 concentration in a more efficient way with less computational resources involved. This allows the comparison of scenarios with different source configurations.

For the detection of emissions from unknown facilities, knowledge of the variability of the background due to known sources is key. The current absolute level of the worldwide Kr-85 background and the fluctuations in the Northern Hemisphere, especially in Europe and parts of Asia, makes detection of clandestine facilities in these regions extremely difficult. Even in areas with low variability, samples would still have to be taken with a high density in space and time to detect clandestine reprocessing in meteorological favorably conditions. In areas with high fluctuations, the detection of small reprocessing facilities via their Kr-85 emissions is currently virtually impossible. At the moment, any approach to detect clandestine reprocessing via Kr-85 emissions must be hand-tailored to the scenario, i.e. the location, the regional meteorological conditions and suspected emission rates. In particular, ongoing reprocessing is not only increasing the total atmospheric content of Kr-85, but spikes from recent emissions also further handicap the detectability of clandestine reprocessing activities.

The following two options would increase the vulnerability of undeclared reprocessing plants to detection:

1. Daily 'mailbox' declarations of stack emission data from (declared) reprocessing plants. Such declarations would significantly improve the value of atmospheric modeling in correlating detected peaks with declared emissions and identifying peaks from unknown facilities.

2. Stop krypton-85 emissions into the atmosphere. This could be accomplished through cryogenic removal of krypton-85 prior to emission from declared reprocessing plants. Krypton retention would stabilize the global krypton-85 inventory and be followed by quick die-away of fluctuations in the baseline. Technologies for efficient krypton removal exist [Soe13], but they are expensive and would add another economic penalty to reprocessing. Licensing future reprocessing plants may require krypton removal based on more stringent radiation protection principles. Alternatively, of course, cessation of reprocessing would have the same effect.

Both options are complementary, and both could be pursued in parallel. This would then greatly increase the detection probability for undeclared facilities, along with the maximum distance from which they could be detected. Our future work will further quantify the significance of each.

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