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An Initial Analysis of ^{85}Kr Production and Dispersion from Reprocessing in India and Pakistan

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Reprocessing of nuclear reactor spent fuel to recover plutonium is accompanied by release of krypton-85 (^{85}Kr), a fission product. Monitoring for ^{85}Kr might form a part of the verification regime of a possible fissile material cut-off treaty and, in the interim, a South Asian moratorium on reprocessing for military and civilian plutonium. Estimates are made of plutonium and ^{85}Kr generated in the spent fuel of India's CIRUS and Dhruva plutonium production reactors, and Pakistan's Khushab reactor, and of the release of ^{85}Kr from subsequent reprocessing activities at India's Trombay reprocessing plant and Pakistan's facility at Nilore. The reprocessing of nuclear power reactor spent fuel at India's Tarapur and Kalpakkam facilities is also examined since India reportedly conducted a nuclear weapons test in May 1998 using non-weapons grade plutonium.

The atmospheric dispersion of krypton from reprocessing activities in South Asia is modeled using both a simple Gaussian plume model and a more sophisticated code available for download or to run on the web, the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT-4), developed by the U.S. National Oceanic and Atmospheric Administration's Air Resources Laboratory and Australia's Bureau of Meteorology.

Preliminary results suggest that it would be difficult to detect the release of krypton from nuclear weapons related reprocessing activities in India and Pakistan at distances of more than around a hundred kilometers, assuming representative atmospheric

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conditions. This would make it difficult for mutual independent verification of a moratorium on reprocessing of military and civilian plutonium.

A possible way forward is offered by claims that the U.S. may have monitored ^{85}Kr emissions from Pakistan's Nilore reprocessing facility. The results presented here seem to support the possibility that the U.S. embassy could have been used as an air sampling station, in keeping with an apparently long standing U.S. program, "Operation Bluenose," to monitor krypton emissions associated with reprocessing. An India-Pakistan moratorium could be based on a similar capability, if India were to use its embassy in Islamabad as a monitoring station and allowed Pakistan to open a consulate in Mumbai, which would be able to detect reprocessing emissions from Trombay and Tarapur, and to open a consulate in Madras for detecting possible reprocessing activity at Kalpakkam.

INTRODUCTION

A longstanding nuclear disarmament proposal has been an indefinite ban on the production of plutonium as part of a larger fissile material cut-off agreement.[1] Having produced large stockpiles of fissile materials for their nuclear arsenals, the major nuclear weapons states (the United States, Russia, China, France, and the United Kingdom) have ended or suspended the production of highly enriched uranium and plutonium for nuclear weapons. However, Israel, India, and Pakistan continue to augment their relatively smaller stocks of fissile material. India, and Pakistan, in particular, seem intent on expanding their fissile material production capacity. An agreement between them to ban further production of fissile material would contribute significantly to India and Pakistan being able to limit their nuclear arms race, and constitute an important step towards nuclear disarmament.

Monitoring atmospheric ^{85}Kr, a radioactive noble gas produced as a fission product in nuclear reactors and released as effluent when spent nuclear fuel is reprocessed to recover plutonium, may help verify such a ban.[2] The importance of detecting reprocessing activity was recognized in a secret United States program, "Operation Bluenose," started in the late 1940s as an effort to monitor plutonium production around the world through tracing released fission product gases, especially ^{85}Kr, by both airborne sampling and a global land-based network including U.S. embassies equipped with ^{85}Kr samplers.[3] The data was used apparently to produce detailed weekly maps of global ^{85}Kr.

India has an extensive nuclear reprocessing program, with reprocessing plants at Trombay and Tarapur, and a large, new facility at Kalpakkam. Spent fuel from India's two plutonium production reactors, CIRUS and Dhruva, is

used to make nuclear weapons (a third production reactor has been proposed), while that from the 14 power reactors is supposed to go towards its Fast Breeder Reactor program (in which a mixture of uranium and plutonium is used as reactor fuel, with a view to breeding more plutonium than is consumed. The longer term goal of the program is to use fast breeder reactors with a thorium blanket to breed uranium-233 and eventually create a ^{232}Th-^{233}U fuel cycle in thermal reactors).[4] There are ^{85}Kr emissions associated with the reprocessing activity.

Pakistan has traditionally relied on uranium enrichment to produce its nuclear weapons material.[5] On March 16, 2000, CBS News reported that "recent air samples prove that Pakistanis have begun reprocessing the fuel rods . . . into weapons grade plutonium. The samples, taken secretly in Pakistan, contain traces of a gas called Krypton 85, which to scientists is a dead giveaway that plutonium is being reprocessed."[6] The claim that Pakistan has started reprocessing spent fuel at its New Labs facility to recover plutonium fits with reports that its Khushab plutonium production reactor started operating in 1998, providing Pakistan a source of unsafeguarded spent fuel (Pakistan's other reactor, the Karachi nuclear power plant, is under IAEA safeguards and its spent fuel is monitored). The first batch of fresh spent fuel from Khushab may have been cooled through 1999, and became available for reprocessing in 2000.

A South Asian ban on producing fissile material would have to include a halt to uranium enrichment and reprocessing to recover plutonium, along with appropriate verification procedures. This article describes briefly plutonium production and reprocessing and estimates the production and release of ^{85}Kr from reprocessing activities in India and Pakistan (Figure 1 shows the location of these facilities). Both a simple Gaussian plume model and the U.S. National Oceanic and Atmospheric Administration's Air Resources Laboratory Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT-4) code are then used to assess the dispersal and detectability of released krypton from reprocessing facilities in the two countries. The important question of how to monitor an agreed end to uranium enrichment is not discussed here.

PRODUCTION AND RELEASES OF ^{85}KR

The atmosphere contains about 1.14 ppm by volume of krypton, and natural krypton consists of a mixture of six stable isotopes: krypton-84 (57%); krypton-86 (17.3%); krypton-82 (11.6%); krypton-83 (11.5%); krypton-80 (2.25%); and krypton-78 (0.35%). Atmospheric krypton also contains radioactive ^{85}Kr, which is largely the result of reprocessing activities especially by the U.S. and former U.S.S.R. over the past nearly 60 years.[7] After reprocessing ends, this

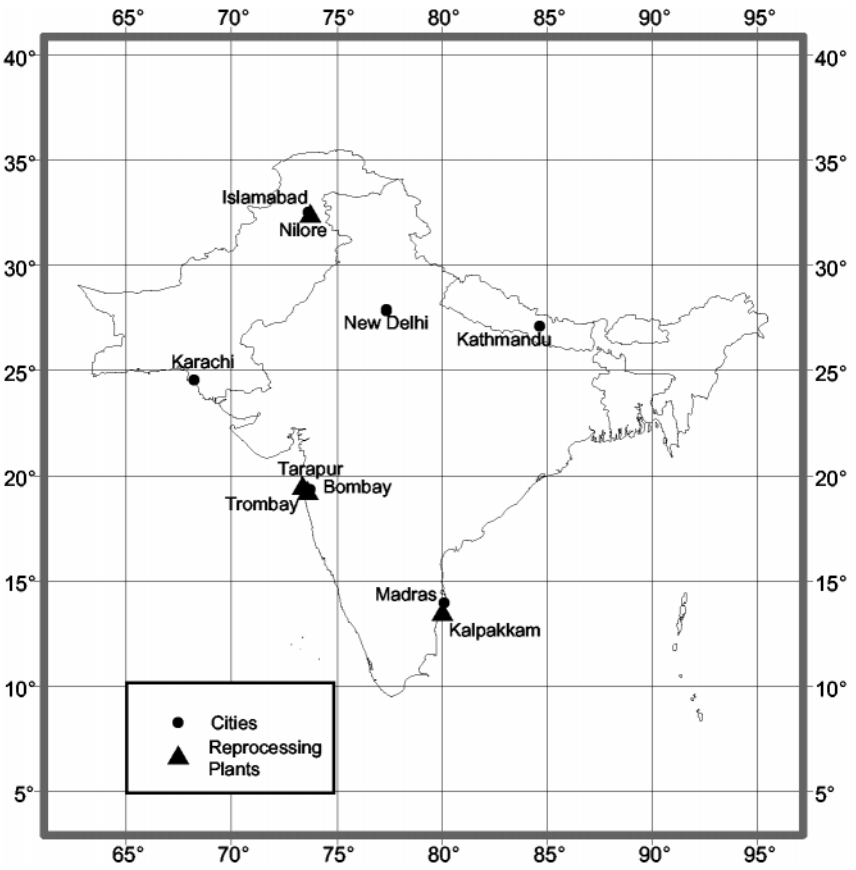

Figure 1: Outline map of India and Pakistan showing the location of reprocessing plants.

will in time dissipate since ^{85}Kr has a half-life of 10.76 years, and decays by β-decay and γ-decay. A tiny fraction of global ^{85}Kr is produced in the atmosphere by krypton-84 neutron capture and in rock from the decay of uranium and thorium. A somewhat larger amount is the legacy of nuclear weapons testing and also leakage from nuclear power plants (reactor effluent contributes only about 1%).

^{85}Kr is a radioactive noble gas produced as a fission product in nuclear reactors.[8] This ^{85}Kr is largely trapped inside the spent fuel rods, along with other fission products. Prior to reprocessing, spent fuel is cooled in ponds to allow for a reduction in the activity from short-lived fission products. Minimum cooling times for the U.S. reprocessing plant at Hanford, Washington, which operated from 1945–1989 and was finally closed in 1992, is given as 200 days

for natural uranium fuel with a maximum burn-up of 1500 MWd/tHM, while spent fuels originally enriched in uranium-235 reprocessed at other facilities with much larger burn-ups had shorter cooling times, of the order of 120–160 days.[9] The reprocessing plants at Marcoule and La Hague in France, and at Sellafield in the United Kingdom, have minimum cooling times for natural uranium fuel of 120–140 days.[10] Since ^{85}Kr has a half life of 10.76 years, a small fraction of it decays during this cooling time.

Reprocessing is a complex and hazardous mechanical and chemical process for plutonium recovery from spent nuclear fuel. The problems arise from the intense radioactivity of the remaining short-lived fission products in the spent fuel (even after it has been cooled for up to a year) and the use of nitric acid in the PUREX process, which is now standard, to extract the plutonium (and uranium) from the fuel matrix. The process produces large amounts of liquid high level radioactive waste.[11] The radioactivity and corrosiveness of the materials involved in reprocessing lead to significant requirements for plant maintenance; to allow for this, a typical reprocessing plant may actually operate only for 150–200 days a year.[12]

Early U.S. plutonium production reactors used uranium metal fuel in an aluminum cladding, similar to that believed to be in use at Dhruva and CIRUS, and Khushab, which was declad for reprocessing by chopping the fuel rod into pieces several centimeters in length and dissolving the aluminium in hot, aqueous sodium hydroxide solution (which does not attack the uranium metal).[13] Some reprocessing plants undertake mechanical decladding, in which the aluminium sheath is removed before dissolution; the process takes about 45 minutes.[14]

Both chemical and mechanical decladding release about 10% of the radiokrypton, as well as some of the other volatile fission products.[15] The bulk of the ^{85}Kr (90%) is released during the subsequent dissolution of the small pieces of the declad fuel rods in hot nitric acid.[16] The dissolution must take place in small diameter tanks, made of materials resistant to corrosion and containing neutron absorbers such as boron or gadolinium to prevent criticality.[17] The total residence time in the dissolver may be of the order of about 4 hours.[18] It is reported that at the 0.17 tonnes of heavy metal per day (tHM/day, i.e., mass of uranium, plutonium and reaction products as spent fuel processed per day) Karlsruhe pilot commercial reprocessing plant in Germany the acid treatment lasts typically 5–8 hours, and the krypton is released in bursts of about 5.5 hours.[19] The dissolution time depends, however, on the nature of the fuel and its burn-up, with uranium metal fuel requiring shorter times than oxide fuel normally used in power reactors. In either case,

it is reasonable to assume that almost all the remaining krypton would be released at this stage. As an inert gas, krypton is not easily captured and most of the ^{85}Kr is released as an effluent from the reprocessing plant chimney stack.[20]

Krypton Detection

There are a number of techniques used to measure ^{85}Kr abundance in air. They typically rely on several cubic-meter-sized air samples being collected and the krypton being isolated using cryogenic and adsorption onto chromatographic columns, with the ^{85}Kr activity of the purified Kr gas measured by scintillation counting of beta activity.[21] It has also proved possible to collect and store krypton, after it has been separated, with an efficiency of about 80% by passing it through a cold metal coil filled with a molecular sieve that is a barrier to air molecules. This allows air samples to be collected, the krypton to be extracted and stored, and the krypton samples sent elsewhere for analysis.

The diversity of techniques for collecting and measuring krypton yields a range of accuracy, determined largely by measurement uncertainty in the mass of recovered krypton (determined for example by measuring its temperature and pressure in a fixed volume) and the ^{85}Kr count rate, which are typically less than 10%.[22] The uncertainty decreases with increases in mass of the sample and higher count rate.

Nonetheless, the krypton background and excess can be now determined at many locations with considerable accuracy. For instance, a recent study of the reliability of ^{85}Kr emission monitoring compared measurements of atmospheric ^{85}Kr with data on the actual releases of the Karlsruhe reprocessing plant in Germany and found a ^{85}Kr background of 1.1 Bq/m^3 (30 pCi/m^3), with a measurement error of $\sim$3%, i.e. $\sim$1 pCi/m^3.[23] This amounts to 7.46×10^8 atoms of Kr-85 per m^3. Measurements of ^{85}Kr from 1979–1998 at Gent, Belgium, often downwind from the British and French commercial reprocessing plants, found peak levels of ^{85}Kr in 1998 as high as 39 Bq/m^3 and a background value (in October 1998) of 1.34 Bq/m^3, with an uncertainty of 2–3%.[24]

The present ^{85}Kr background varies most significantly with the proximity to reprocessing facilities, latitude, longitude, altitude, and global atmospheric mixing processes, and it has been possible to produce good tropospheric scale models.[25] But to first order, it is reasonable to assume that the ^{85}Kr background is constant over much of the Northern Hemisphere and, in regions far from very large reprocessing facilities, the temporal variability is low.[26] It is assumed that ^{85}Kr concentrations in a plume from a reprocessing plant greater than about twice the krypton background measurement error (i.e., greater than 2 pCi/m^3)

would be reliably detected and constitute a reasonable indicator of reprocessing activity.

Krypton is not the only fission product released to the atmosphere during reprocessing that has been detected at long distances. Traces of Iodine-129 produced by the La Hague and Sellafield reprocessing plants and released mostly as methyl iodide have been detected in rain water (typically of the order of 10^7 I^{129} atoms/L) and in mosses and lichens in the continental United States.[27] It has also been detected at levels 10 to 20 times higher in rain and snow in Sweden.[28] Although much more dependent on meteorological conditions, Iodine-129 may offer an independent check on inferences concerning the presence and sources of reprocessing activity.

Stable noble gas isotopes of krypton and xenon are also released from stacks of reprocessing plants. It has been proposed that in samples taken directly in the stack, stable isotope ratios could be measured with sufficient precision to identify the burn-up and type of fuel being reprocessed, i.e., to distinguish between power reactor fuel subject to high burn-up and low burn-up plutonium production reactor fuel.[29] This very intrusive sampling regime would presumably form part of the strict verification provisions accompanying an international agreement that banned only reprocessing for weapons-grade plutonium. The problem with safeguarding against the possible use of non-weapons grade plutonium for nuclear weapons would remain.

PLUTONIUM AND ^{85}Kr PRODUCTION IN INDIA

India has a large nuclear energy complex and a smaller, but interlinked, dedicated nuclear weapons complex. There are presently 14 power reactors, with a combined, nominal installed capacity of about 2.7 GWe, although a number of these reactors have had their nominal capacity reduced.[30] India has two plutonium production reactors, CIRUS and Dhruva, and four very low power research reactors: Apsara, Zerlina, Purnima-I, and Purnima-II. India also has a 40 MWth Fast Breeder Test Reactor fueled with plutonium-uranium-carbide. The plutonium fuel for this reactor, and for the planned 500 MWe Prototype-Fast Breeder Reactor, has been produced by reprocessing spent power reactor fuel. The plutonium for India's nuclear arsenal is derived from the two production reactors.

CIRUS

CIRUS, originally named the Canada-India Reactor (CIR), located at the Bhabha Atomic Research Center at Trombay, a suburb of Mumbai (formerly

Bombay), is a Canadian designed and built 40 MWth plutonium production reactor. It is similar to the Canadian NRX reactor. While construction was completed in 1960, CIRUS only commenced full power operation in 1963. It has natural uranium metal fuel, and is moderated by heavy water but cooled by light water.[31] Weapons-grade plutonium produced in CIRUS was used in India's first nuclear explosion in May 1974. For the purposes of this exercise, it is reasonable to assume a capacity factor for the plant of about 70%, and using a typical burn-up used for optimizing weapons grade plutonium (i.e., Pu-239) production of about 1000 MWd/tHM, and a standard plutonium production rate of 0.9g Pu per MWth-d of output, then CIRUS could produce about 9 kg of weapons-grade plutonium a year.

Dhruva

India's other nuclear weapons plutonium production facility is the Dhruva reactor, a 100 MWth (thermal) reactor, also located at BARC and fueled by natural uranium metal, clad in aluminum and cooled and moderated by heavy water.[32] It has been in operation since August 1985.[33] Soon after start-up, the reactor was closed for a year due to vibration problems caused by the design of the fuel, and it resumed operation at low power in October 1986.[34] The reactor was almost immediately shut down again until December 1986 for further changes in the fuel design, and there were reported to be other obstacles to operating the reactor at its design capacity.[35] It was not until January 1988 that Dhruva was reportedly able to operate at full power.[36] At 70% capacity, Dhruva would produce about 21 kg of weapons-grade plutonium per year.[37]

The combined plutonium production at BARC's Trombay facilities could be about 30 kg per year, assuming reasonable capacity factors for its production reactors. This may be sufficient to add several nuclear weapons worth of weapons-grade plutonium to its nuclear stockpile each year.[38] Plans have been announced to build a new reactor similar to Dhruva, most likely at BARC, to be operational by 2010.[39] This would almost double India's weapons-grade plutonium production capability.

Reprocessing in India

India has three reprocessing plants, at Trombay, Tarapur, and Kalpakkam. The reprocessing plant at Trombay, built in the 1960s, is apparently dedicated to the spent fuel from the two plutonium production reactors at that site, i.e., it is used for recovery of weapons-grade plutonium. It had an original design capacity of 0.1–0.15 tHM/d, or about 30 tHM/year (comparable in size to the one

at Karlsruhe, Germany).[40] The facility was shut down between 1974 and 1983–1984, for decontamination and rebuilding, and now has a nominal capacity of 50 tonnes of heavy metal per year, significantly larger than what is required for the two production reactors on the site.[41] This excess capacity may be used to accommodate spent fuel from the proposed Dhruva II production reactor mentioned earlier.

The Power Reactor Reprocessing Plant (Prefre) at Tarapur, about 200 km up the coast from Mumbai, was completed in 1975 and began operating in 1982.[42] It has a design capacity of 150 tHM/year, and each reprocessing campaign lasts "six to eight months to about one year."[43] It has been used to reprocess spent fuel from the CANDU reactors, Rajasthan-1 and 2, and Madras-1 and 2 (in Tamil Nadu).[44]

India's third and largest reprocessing plant is the new Kalpakkam Reprocessing Plant (KARP), some 80 km south of Chennai (formerly Madras). It is claimed to have two reprocessing lines each with a design capacity of 100 tHM/year, which can be upgraded to 125 tHM/year; the two lines will not be run in parallel, the first is planned to run for seven to eight years and then be decommissioned, at which time the second line would take over.[45] The facility became operational in 1998 and was reported as having "operated satisfactorily" in 1999 and 2000.[46] It is supposed to reprocess spent power reactor fuel to separate plutonium for use in India's fast breeder reactor program. Since Prefre and Kalpakkam are believed to be used for reprocessing spent power reactor fuel, their ^{85}Kr emissions will be considered separately from Trombay.

There is apparently a fourth reprocessing facility, of laboratory scale, at Kalpakkam which has been used for recovering U-233 from Thorium fuel.[47] This is not considered further.

TROMBAY KRYPTON RELEASE

The ^{85}Kr inventory in the fresh spent fuel from CIRUS and Dhruva can be estimated from two simple rules of thumb:[48]

1. For each gram of U^{235} that is fissioned, 0.405 Ci of ^{85}Kr are produced; for each gram of Pu^{239} fissioned, 0.177 Ci ^{85}Kr is produced.
2. In a plutonium production reactor, for each gram of weapons-grade plutonium in the spent fuel 1.09 g of U^{235} fissioned and 0.12 g of Pu^{239} also fissioned.

The production of weapons-grade plutonium in a CANDU reactor may be a few percent larger than in the U.S. water-cooled, graphite-moderated Hanford

plutonium production reactors and the heavy water-moderated production reactors at Savannah River (South Carolina, USA) from which the second of these estimates is derived.[49] Nonetheless, the above rule offers a conservative estimate of ^{85}Kr abundance in Indian plutonium production reactor spent fuel; 0.46 Ci of ^{85}Kr per gram of weapons-grade Pu.

Taking Dhruva and CIRUS together as producing about 30 kg of weapons grade Pu a year, there would be 13,800 Ci of ^{85}Kr in their fresh spent fuel discharges every year. If the fuel is cooled for one year before reprocessing, which is a conservative assumption, then about 6% of this ^{85}Kr will decay (recall ^{85}Kr has a half life of 10.76 years).

Assuming that the Trombay plant operates for 200 days each year and that the krypton release takes place over a five-hour period in each of these days, then the ^{85}Kr release rate from the Trombay reprocessing plant would be about 13 Curies per hour, or 3.6 mCi/s.

PLUTONIUM AND ^{85}Kr PRODUCTION IN PAKISTAN

Pakistan has a much smaller nuclear complex than India. It has two power reactors: the 30-year-old Canadian designed and built Karachi Nuclear Power Plant (a 125 MWe PHWR), and a new Chinese designed and built Chashma Nuclear Power Plant (a 300 MWe PWR). Both these reactors are under IAEA safeguards. Pakistan also has two very low power research reactors, PARR-I and PARR-II, which are also under safeguards. In addition, Pakistan has one unsafeguarded plutonium production reactor.

Khushab

Pakistan's has a single dedicated plutonium production reactor, located at Khushab in the province of Punjab.[50] It is reported to be a 40–50 megawatts (thermal) natural uranium fueled, heavy water moderated reactor.[51] It is said to have gone critical in 1998.[52]

Assuming again a burn-up of 1000 MWd/tU, with about 0.9g of weapon grade plutonium produced per megawatt (thermal) day of output and that the reactor operates at 60–80% of its capacity, Khushab could produce about 10 kg of plutonium per year.

Reprocessing in Pakistan

Pakistan is believed to have established a reprocessing capability at its New Labs facility, located next to the Pakistan Institute of Nuclear Science and Technology at Nilore (just outside Rawalpindi and Islamabad), with a design

capacity of 10–20 tons of heavy metal per year.[53] However, it is reported that "due to technical problems, the New Labs facility is operated at a lower throughput than its design would otherwise warrant but the effective capacity factor. . . is high enough to handle the entire spent fuel inventory discharged from the Khushab reactor without generating any bottlenecks."[54]

If Khushab produces 10 kg of Pu a year, then there would be an attendant 4600 Ci of ^{85}Kr. Making the same conservative assumption as in the case of India, i.e., that the fuel is cooled for one year before reprocessing, then New Labs would receive spent fuel containing 4313 Ci. Assuming, again, that the reprocessing plant runs for 200 days a year and the ^{85}Kr release is spread over five hours during these days, then the estimated release rate is about 1.2 mCi/s.

With the Khushab reactor having gone critical perhaps only in early 1998, and with a full load of fuel being exposed in one year (about 9 tonnes), and allowing another year for cooling, reprocessing at New Labs may only have started in earnest in early 2000. This would be consistent with claims in March 2000 that ^{85}Kr has been detected by the United States in Pakistan. A possible site for U.S. monitoring of ^{85}Kr releases would be the U.S. embassy in Islamabad, a distance of 10–20 km. As mentioned earlier, the use of U.S. embassies in other states for monitoring reprocessing activities through ^{85}Kr detection apparently has a long history. The krypton dispersal calculations in the next section allow for an assessment of the viability of such detection in the case of Pakistan.

A summary of the estimates for the average annual rates of plutonium production at Cirus and Druva in India and at Khushab in Pakistan, along with the rates of ^{85}Kr release associated with reprocessing during runs at the Trombay facility in India and and at the New Labs plant in Pakistan is given in Table 1.

DISPERSAL MODELS

The two models that are considered here for atmospheric transport are the simple, first order, Gaussian plume model and the far more sophisticated

Table 1: Estimated plutonium production and ^{85}Kr releases from nuclear weapons activities in India and Pakistan.

	Plutonium (kg/year)		Krypton-85 (Curies/hour)
India	Cirus 9	Dhruva 21	Trombay 13
Pakistan	Khushab 10		New Labs 4.3

but user-friendly HYSPLIT-4 package developed by the U.S. National Oceanic and Atmospheric Administration's Air Resources Laboratory and Australia's Bureau of Meteorology. Each model is briefly described below and then applied to the reprocessing facilities in India and Pakistan. More extensive descriptions and the results of these models are given in Appendices 1 and 2, respectively.

The Gaussian Plume

A Gaussian plume is often used to provide a first order estimate of the concentration of atmospheric contaminants from point sources.[55] The model requires as input only limited local meteorological conditions (windspeed and solar insolation), a value for the release rate of the airborne contaminant, and a height of release. All of these are assumed constant in the model. Comparison of predictions from a Gaussian plume model with measurements of ^{85}Kr releases from the Savannah River Plant (South Carolina, USA) at distances of about 10 km found the model to be in agreement with observations to within a factor of two; the predictions were also systematically larger than the data.[56] This model can provide useful insight into krypton dispersal from weapons-grade plutonium reprocessing plants in South Asia at distances of tens of kilometers.

Dispersal from Trombay and Nilore

The monthly averaged wind data for Mumbai (Trombay is a suburb to the northeast) was inferred from the Global Gridded Upper Air Statistics database, which uses measurements from 1980–1995 to describe the atmosphere for each month of the year with a spatial resolution of about 100 km in the middle latitudes. This suggests a monthly averaged surface wind speed around Mumbai of 3.5–6.2 m/s. The other assumptions about atmospheric conditions are given in Appendix 1. The results, in figure A1.1 (Appendix 1), suggest that, depending on the atmospheric conditions, the ^{85}Kr releases from reprocessing activities at Trombay (at a rate of 13 Curies per hour, or 3.6 mCi/s) could be detectable at distances of 50–150 km.

The atmospheric conditions assumed here, including the effects of reflection from the atmospheric mixing layer would increase the predicted ground level concentration at distances greater than about 10–20 kilometers from the stack (the distance at which the top of the plume meets the mixing layer, taken to be at a height of about 1 kilometer, and is reflected back towards the ground). A taller stack also reduces predicted ground level concentration close to the source but not at large distances. Finally, for emissions at night time, the downwind concentration at large distances would also be larger than predicted here.

But even with less conservative assumptions than those made here concerning the rate of plutonium production and associated ^{85}Kr in the CIRUS and Dhruva reactors, the relatively long one-year cooling time for the spent fuel, the short chimney or stack height of the reprocessing plant, and day-time emissions, it appears very unlikely that any useful observations could be made by a monitoring station located in Pakistan, the nearest point in which is about 700 km away.

A similar calculation for ^{85}Kr released from reprocessing at Pakistan's New Labs facility suggests it would apparently be detectable at distances of the order of 100 km, which is somewhat larger than the distance from Nilore to the border with India. Again, the effect of including reflections from the mixing layer would increase the predicted concentration at ground level. Given the conservative assumptions made here, it may be feasible for India to monitor from its own territory these ^{85}Kr emissions, when the wind is in the right direction.

The Gaussian plume calculations therefore show a significant asymmetry between India and Pakistan in the possibility of using their national territory to usefully monitor reprocessing associated with plutonium recovery for the other's nuclear weapons program. While India may be able to undertake such monitoring, Pakistan cannot.

"Bluenose" in South Asia

The reported U.S. detection of ^{85}Kr in Pakistan in 2000 and the existence of "Operation Bluenose," the U.S. program to clandestinely monitor reprocessing activity in other states including through the use of detectors located at its embassies, offer an interesting application of the Gaussian model. The Gaussian plume equation was used to calculate the expected ^{85}Kr concentration near ground level at a distance of 10–15 km from the New Labs plant, comparable to that of the U.S. embassy in Islamabad. The results suggest that the excess ^{85}Kr concentration at those distances would be 15–30 pCi/m^3. This would be significantly above the background and thus could be reliably detected in air collected in rooftop air samplers. This would indicate that the U.S. embassy in Islamabad could have detected the release of ^{85}Kr from the onset of reprocessing at New Labs.

The use of an embassy as a krypton monitoring station offers a solution to the asymmetry noted earlier between the India and Pakistan's ability to monitor each other's krypton releases. If it were suitably equipped, the Indian embassy in Islamabad could monitor the New Labs at Nilore as part of a bilateral India-Pakistan agreement to halt reprocessing. As part of such an agreement, India could permit Pakistan to reopen a consulate in Mumbai, closed

since March 1994, which could feasibly monitor the end of reprocessing activity at the Trombay plant, located in the northeast of the city. However, there are some complications created by the activity of India's Tarapur reprocessing plant which are discussed below.

HYSPLIT-4

HYSPLIT-4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) is the latest in a series of programs developed over the past two decades by NOAA's Air Resources Laboratory and Australia's Bureau of Meteorology to compute trajectories, dispersion, and deposition of airborne pollutants. It offers a far more sophisticated tool for tracking dispersal of ^{85}Kr than the simple Gaussian plume. HYSPLIT-4 is on-line, along with a detailed users guide, and can be run on the Web from the NOAA website or the executable code and meteorological data can be downloaded to a PC.[57]

HYSPLIT-4 has previously been applied to modeling ^{85}Kr dispersion. A comparison of measurements of ^{85}Kr made at Gent, Belgium, with HYSPLIT-4 predictions from reprocessing activity at La Hague, France, based on hourly emission data from the plant, suggested the model appeared systematically to underestimate the observations by a factor of 2 to 4.[58] This caution should be borne in mind in assessing the significance of the following results of applying HYSPLIT-4 to ^{85}Kr releases in South Asia, along with the assumptions that have been made about ^{85}Kr production and release.

HYSPLIT-4 was run for a five-hour krypton emission from Trombay and the dispersal tracked over a 48-hour period starting 6 AM, March 26, 2001. The HYSPLIT-4 results (Figure A2.1) suggests that the ^{85}Kr plume drifts eastwards, away from Pakistan, for the prevailing weather conditions. After 12 hours, the Kr concentration contour of 1 pCi/m^3 extended out to about 170 km from the source. Between 12 and 24 hours after the emissions, the ^{85}Kr concentration in the plume from Trombay has fallen below the measurement errors of the krypton background and is essentially undetectable.

This can be compared with results from the Gaussian plume model. The Gaussian results suggest that the 1 pCi/m^3 contour reaches a maximum extent of about 190 km downwind. However, the models diverge significantly in the crosswind direction. The Gaussian shows a maximum crosswind spread over about 30 km at downwind distances of about 100 km, while HYSPLIT-4 has a contour width of about twice this.

HYSPLIT-4 was also run for Nilore for a smiliar period and time of year. The results (Figure A2.2) show a more circular, slower moving dispersal pattern than in the case of India. The ^{85}Kr concentration falls below the detection threshold after about 12 hours.

The calculations were repeated for Nilore for another time of year (July), and even though the wind directions were different, the overall dispersal pattern remained the same. It may be that the lower average windspeeds and a temperature inversion that is often found in the Rawalpindi area combine to produce less advection and thus a weaker plume, giving instead the more symmetrical, diffusion dominated dispersal pattern which is found in the calculations. This suggests that even though the plume from Nilore may at times drift over India, it would not be reliably detected.

Tarapur and Kalpakkam

The discussion so far has been concerned about the releases and subsequent dispersal of Kr from the nuclear weapons reprocessing plants in India and Pakistan. While Pakistan has only one reprocessing plant, and one source of unsafeguarded spent fuel, India has two reprocessing plants for power reactor spent fuel (almost all unsafeguarded), at Tarapur and Kalpakkam. These facilities are much larger than the plant at Trombay, and could be used for reprocessing weapons-grade plutonium, should India wish to do so.

These facilities are also important because India could seek to use non-weapons grade (and possibly reactor-grade) plutonium for its weapons given that nuclear weapons expert have made clear that the difficulties are "not appreciably greater" in building nuclear weapons from reactor-grade plutonium than from weapons-grade.[59] This is particularly significant since on May 11, 1998, as part of its series of nuclear tests India conducted an explosion using non-weapons grade plutonium, and it has been suggested reactor-grade plutonium or a mixture of reactor-grade and weapons-grade plutonium may have been used.[60]

Assuming that during normal operation, the Tarapur and Kalpakkam plants reprocess spent power reactor fuel from India's 12 PHWR's, then it is possible to estimate the associated ^{85}Kr emissions. PHWRs are fueled with natural uranium, containing 0.72% U-235 (i.e., 7.2 kg U-235 per tonne of fuel) and have a typical burn-up of 7000 MWtd/t. There is about 2.3 kg of U-235 per tonne in the spent fuel at this burn-up. Along with the 4.9 kg of U-235 that fissions, about 3 kg of the plutonium that is produced (assumed to be all Pu-239) also fissions. With about 0.405 Ci from each gram of U-235 that fissions and 0.177 from each gram of Pu-239 that fissions, then there are about 2500 Curies of Kr-85 in each tonne of PHWR spent fuel.

During a one-year cooling period, the Kr-85 abundance will decrease by about 6% and thus about 2350 Curies of Kr-85 per tonne of spent fuel reaches

Table 2: Reprocessing plants and estimated Krypton-85 releases in India.

	Trombay	Tarapur	Kalpakkam
Design capacity (tHM/year)	50 (weapons-grade)	150 (power plant fuel)	100 (power plant fuel)
Kr-85 releases (Curies/hour)	13	350	235

the reprocessing plant. Tarapur's 150 tHM/year design capacity spread over 200 days gives it a reprocessing rate of 0.75 tHM/d. Supposing five hours of release on each day, the ^{85}Kr release rate will be 350 Ci/hour. For Kalpakkam, with a reprocessing capacity of 0.5 tHM/d, the ^{85}Kr release rate will be 235 Ci/hour. These are over 20 times the release rate from the Trombay facility. Table 2 summarizes the reprocessing capacity and the estimated ^{85}Kr releases for the three plants in India.

HYSPLIT was run for a 48-hour period in March 2001 for emissions from Tarapur (Figure A2.3 shows the dispersal after 12 hours). The results suggest that the concentrations of 10 pCi/m^3 extend over 150 kilometer from the site, and thus there may be mixing of the Kr plumes from Trombay and Tarapur within about 12 hours. The concentrations from Tarapur are, of course, much larger. A similar result can be seen in Figure A2.4 which shows the dispersal from all three Indian reprocessing plants simultaneously for a 12-hour period in August 2000.

Plume mixing could make it difficult to consistently separate a ^{85}Kr signal from Tarapur with suspected emissions from Trombay. This would become an issue in verifying any reprocessing ban that was restricted to only the presently dedicated weapons-grade plutonium recovery operations (i.e., to Trombay in India and New Labs in Pakistan). It would be easier to verify a blanket ban on reprocessing, one that included explicitly recovery of weapons-grade plutonium and civilian reprocessing of power reactor spent fuel (reactor-grade plutonium also can be used to make nuclear weapons).

The results indicate that the proposed monitoring station in Mumbai could possibly be used to detect ^{85}Kr from both Trombay and Tarapur. Although, the detection threshold seems to be reached at distances comparable to that of Mumbai from Tarapur, this should not be read as a very strong constraint. Conservative assumptions have been made about the rates of krypton production and release and a previous application of HYSPLIT-4 to tracing krypton dispersal and comparing its predictions based on actual release data with field measurements found that it typically underestimates krypton concentrations

by a factor of 2 to 4. Taken together, it seems feasible to use a monitoring station in Mumbai to detect reprocessing activity at both Trombay and Tarapur when the wind is blowing toward Mumbai.

The releases from Kalpakkam suggest a more serious problem. Kalpakkam, the largest source of krypton, is located at the greatest distance from Pakistan, and as the figure shows, the winds seem to blow the plume further eastwards away from Pakistan and into the Indian Ocean. Even if the wind were to blow towards Pakistan, there seems no feasible means to detect such emissions at a station on Pakistan's national territory or from a monitoring station at Mumbai. Alternatives might be to locate another local monitoring station in the nearby city of Madras, or to use a sea-based platform.

CONCLUSIONS

India and Pakistan, along with Israel, continue to produce fissile material for their nuclear weapons, thus laying the basis for larger arsenals. India has a long-established program for reprocessing spent fuel from its dedicated weapons-grade plutonium production reactors, and may also have exploded a nuclear weapon based on non-weapons grade plutonium in its May 11, 1998 tests. Pakistan has recently initiated plutonium production at its Khushab reactor and begun recovery of plutonium through reprocessing at its New Labs facility. This is in addition to the much larger uranium enrichment program Pakistan established over the past several decades to produce weapons-grade uranium for its nuclear arsenal.

A simple Gaussian plume model has been applied to trace the dispersal of emissions of ^{85}Kr from the reprocessing plants in India and Pakistan. The results show a clear asymmetry between Pakistan and India with regard to possible monitoring from their respective national territories of the other's ^{85}Kr emissions. Under some weather conditions the release gases from Pakistan's sole reprocessing plant can blow into the Indian territory, but it would appear Pakistan may not be able to detect sufficient concentration from the main Indian weapon related reprocessing plant. This poses a problem for any suggestion for a bilateral South Asian moratorium on fissile material production or a global fissile material production cut-off agreement that relies only on national capabilities for verification.

The United States was reportedly able to monitor the onset of reprocessing activity in Pakistan in 2000. The Gaussian plume results show emissions from Pakistan's New Labs would be detectable in Islamabad. This result in combination with reports that the United States has had since the early 1940s an

effort ("Operation Bluenose") to clandestinely monitor reprocessing activities in other states, including through siting krypton monitoring at U.S. embassies, suggests the U.S. embassy in Islamabad could, in principle, have been used as the monitoring station.

Following this example, India could, in principle, use its embassy in Islamabad to detect ^{85}Kr emissions from reprocessing in Pakistan and permit Pakistan to reopen its former consulate located in Mumbai to monitor emissions from the nearby Trombay reprocessing plant. The technology for storing krypton samples in a metal coil allows air samples to be collected on site and the krypton extracted and stored and sent for analysis elsewhere, in this case Pakistan. This could offer a feasible first step in independent, mutual verification of a moratorium on the reprocessing of spent fuel for plutonium in the two countries.

The reprocessing of power reactor plutonium at India's unsafeguarded Tarapur and Kalpakkam plants poses some additional problems. Reactor-grade plutonium can be used to make nuclear weapons, and these facilities would have to be included in a fissile material cut-off moratorium or treaty. Further, the Tarapur reprocessing plant is about 200 km from Mumbai, and the HYSPLIT-4 simulation suggests that under certain circumstances its ^{85}Kr plume may mix with that from Trombay, making it more difficult to monitor the dedicated weapons-grade plutonium production facility there. A halt to reprocessing at Tarapur may possibly be verified by the station proposed to monitor activity at Trombay, although it is at the outer range of detectability suggested by the simulation.

Finally, the Kalpakkam facility, by far the largest reprocessing plant in India, is located far from Pakistan. There is no prospect of monitoring its activity from any station located in Pakistani territory. Moreover, the results shown here indicate the winds may at times blow its releases eastwards, further away from Pakistan. However, Kalpakkam is close to the city of Madras. Following the solution proposed for Trombay, a Pakistani consulate in Madras could feasibly be able to monitor the Kalpakkam plant by siting air samplers on its roof and either processing the samples on site for the ^{85}Kr content or by extracting and storing the krypton and sending the samples to Pakistan for further analysis.

NOTES AND REFERENCES

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production of plutonium and highly enriched uranium would limit the potential size of nuclear arsenals.

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11. For details of reprocessing and the PUREX process see, for example, Benedict et al., *Nuclear Chemical Engineering*, 457–564.

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APPENDIX 1: GAUSSIAN PLUME MODEL

The simplest Gaussian dispersion relations for the concentration Q (m^{-3}) at a point (x, y) in a horizontal plane can be written as:

$$Q(x, y) = \frac{Q_o}{\pi\, u\sigma_y\sigma_z} e^{-\frac{h^2}{2\sigma_z^2}} e^{-\frac{y^2}{2\sigma_y^2}}$$

where Q_o is the rate of release of the contaminant from the source, h is the effective chimney height (m), and wind speed (taken to be in the x direction) is u (m/s). The spread of the plume in the y and z directions is given by:

$$\sigma_y = a \cdot x^{0.894} \text{ and } \sigma_z = c \cdot x^d + f$$

where a, c, d, and f are empirical coefficients determined by the atmospheric stability conditions, which are in turn determined by the surface wind speed and solar insolation. The downwind distances x need to be in kilometers to give the Gaussian dispersion coefficients in meters. This appendix contains tables showing the stability classifications for different wind speeds and isolation and the associated values of the coefficients a, c, d, and f. The model neglects reflection from the atmospheric mixing layer, including this effect would increase the predicted concentration at large distances.

Tables A1.1 and A1.2 give the stability classifications and coefficients used in the Gaussian plume calculations. The tables are reproduced from G. M. Masters, *Introduction to Environmental Engineering and Science*, 1991, p. 320.

Trombay

The atmospheric stability conditions are taken as varying between type B and type C (i.e., with surface wind speed measured 10 m above the ground as 3–6 m/s, and strong to moderate solar insolation as would be expected from a sunny day). The mean monthly hours of sunshine at Trombay vary from 168 to 288, except for the two rainy months when there are only about 70–80 hours of sunshine.[61] The appropriate Gaussian dispersal

Table A1.1: Atmospheric stability classifications for different wind speeds and solar insolation. (Surface wind speed is measured at 10 m above the ground.)

Surface wind speed (m/s)	Daytime solar insolation		
	Strong*	Moderate**	Slight***
<2	A	A-B	B
2–3	A-B	B	C
3–5	B	B-C	C
5–6	C	C-D	D
>6	C	D	D

*Strong solar insolation corresponds to a clear summer day, with the sun higher than 60 degrees above the horizon.
**Moderate solar insolation corresponds to a summer day with a few broken clouds, or with the sun 35–60 degrees above the horizon.
***Slight solar insolation corresponds to a cloudy summer day, an autumn afternoon, or with the sun 15–35 degrees above the horizon.

parameters can be read from the table above. For the purposes of this analysis, it is assumed that the stack height is 30 meters.

As noted earlier, while current ^{85}Kr measurement accuracy is ~1 pCi/m^3 the application of the Gaussian plume to ^{85}Kr releases from Savannah River (South Carolina, USA) found the model to be reliable (to within a factor of two) but typically larger than the data for distances of about 10 km. That is, Gaussian predicted levels of 1 pCi/m^3 may not be reliably detectable at these distances and beyond. It seems reasonable, therefore, for the purposes of this calculation, to consider Gaussian model concentrations of 2 pCi/m^3 and 3 pCi/m^3 as indicative of levels that may be reliably detectable. The 2–3 pCi/m^3 ^{85}Kr contours from the Trombay plant, for atmospheric stability classes B and C, are shown below.

Nilore

Average daytime mean wind speeds are between three and four meters per second around Rawalpindi and Islamabad, the major cities close to Pakistan's New Labs reprocessing plant, from the same database as was used for Trombay. The solar insolation is taken as similar to Trombay, as is the stack height. The ^{85}Kr release rate from Pakistan's New Labs reprocessing rate was estimated earlier as about 1.2 mCi/s over each five-hour reprocessing period.

Table A1.2: Values of Gaussian coefficients for different stability conditions at dispersal distances greater than 1 km from the source.

Stability	*a*	*c*	*d*	*f*
A	213	459.7	2.094	−9.6
B	156	108.2	1.098	2.0
C	104	61.0	0.911	0
D	68	44.5	0.516	−13.0

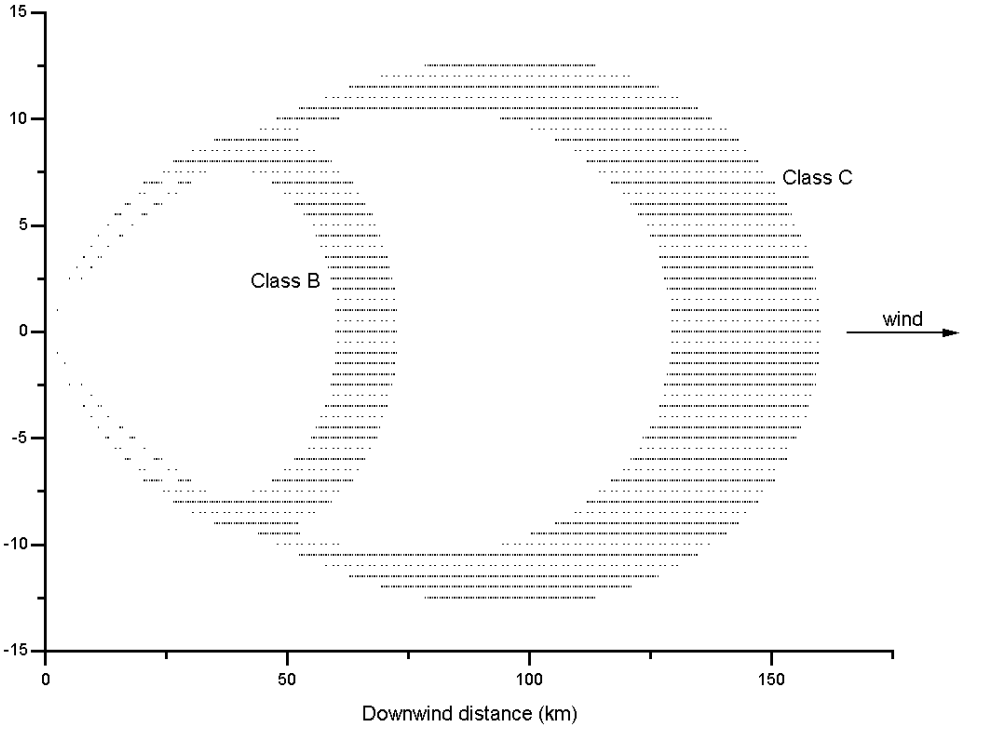

Figure A1.1: Gaussian plume calculations of the ^{85}Kr concentration at ground level from Trombay reprocessing plant for atmospheric stability classes B and C. The contours show concentrations in excess of the global ^{85}Kr background. The shaded areas show ^{85}Kr between 2 and 3 pCi/m^3.

APPENDIX 2: THE HYSPLIT-4 MODEL

HYSPLIT-4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) is the latest in a set of models developed by NOAA's Air Resources Laboratory and Australia's Bureau of Meteorology to calculate the dispersal of airborne pollutants. The user-friendly package consists of a modular library with programs for each specific application. Gridded meteorological data, in one of a number of possible map projections, are required at regular time intervals. This data, as archives or from forecast model outputs, is available on the website in a form already formatted for input to HYSPLIT. In addition, there are programs provided to convert NOAA, NCAR (National Center for Atmospheric Research) or ECMWF (European Center for Medium-range Weather Forecasts) outputs into a format appropriate for the HYSPLIT-4 model.

The details of the mathematical formalism underlying the different parts of the dispersion simulation in HYSPLIT-4 are set aside here in favor of a brief qualititative outline.[62] The model permits multiple, simultaneous pollutant species to be released from distinct sources The dispersion of each pollutant released in the form of a particle or "puff" is calculated by assuming either a Gaussian or "top-hat"(constant value inside a puff and zero outside) horizontal distribution within a puff or from the dispersal of a fixed

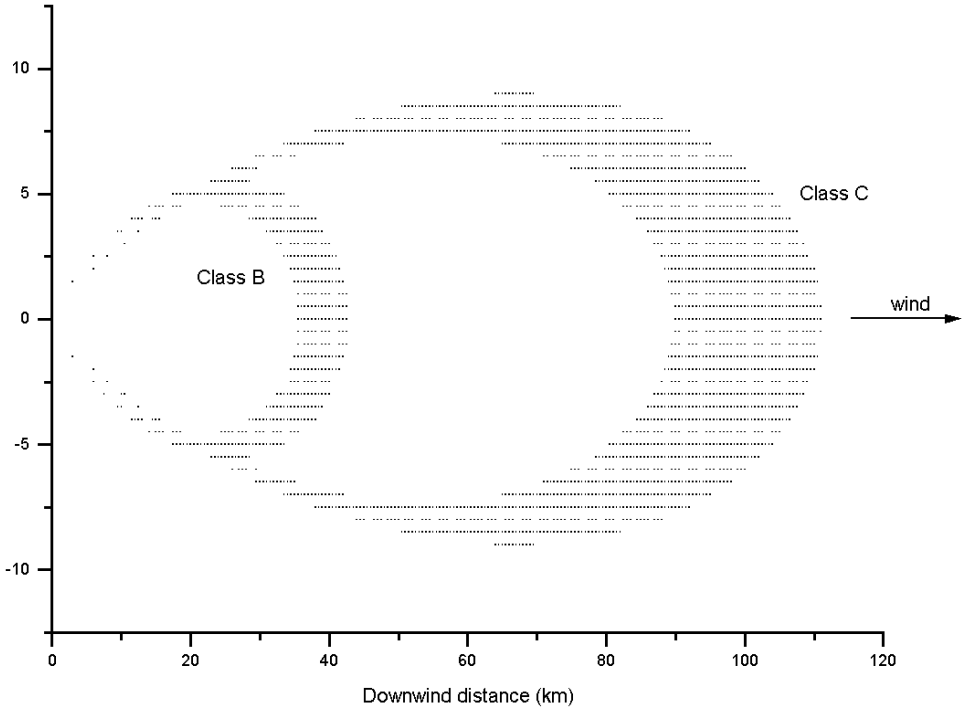

Figure A1.2: Gaussian dispersion contours of ^{85}Kr from New Labs, Nilore. The inner contour for both stability class B and C is for a ^{85}Kr concentration of 3 pCi/m^3, while the outer one is for 2 pCi/m^3.

number of particles. In the vertical direction, a puff distribution is always assumed to be "top-hat." A single released puff will expand until its size exceeds the meteorological grid cell spacing, and then it will split into several puffs each with some fraction of the initial pollutant mass. A top-hat puff splits horizontally into four equal puffs, and a Gaussian puff splits into five puffs with a large, center puff retaining 60% of the initial mass, and the remaining mass equally distributed among four smaller puffs. The problem of growing numbers of small puffs is dealt with by merging puffs which become sufficiently close to each other, with the new puff's dispersion coefficients set as a mass-weighted sum of the individual puffs. Puffs are also periodically sorted and those with less than 10% of the total initial mass are sorted by position and, where appropriate, merged.

Air concentrations are calculated at latitude-longitude intersection grid points for puffs and as cell-average concentrations for particles. The model contains gridded land use, roughness length, and terrain data, with a resolution of 1 degree in the Northern Hemisphere. The model allows for wet and dry deposition of pollutants, radioactive decay, and resuspension.

The basic model simulation input parameters are :

- starting time (year, month, day, hour),

- location (starting locations, as latitude, longitude, and height),
- start time and duration of the dispersion (i.e., run-time),
- pollutant characteristics (number of pollutant species, emission rates, emission duration),
- calculation grid size (including mixing height and height of each vertical level in concentration grid), and
- particle properties (diameter, density and shape, deposition velocity, molecular weight, surface reactivity ratio, diffusivity ratio, radioactive decay half-life, resuspension rate, etc.), output resolution, and sampling time.

Trombay and Nilore

HYSPLIT-4 was run for a five-hour krypton emission from the Trombay reprocessing facility and the dispersal mapped every 12 hours over a 48-hour period starting 6 AM, March 26, 2001. The meteorological data was the weather forecast for that period. Figure A2.1 shows the results for the first 12 hours. The grid is latitude and longitude. The contours are for 1 pCi/m^3 (i.e., the current measurement error) reflecting that applications of this model to ^{85}Kr releases elsewhere show that it appears to systematically underestimate the observations by a factor of 2 to 4. This suggests a HYSPLIT-4 predicted level of 1 pCi/m^3 would be reliably detectable.

HYSPLIT-4 was run for a 48-hour period following release at Nilore for April 2001. The results are given (Figure A2.2) as a snapshot after 12 hours.

Tarapur

HYSPLIT-4 was run for a 48-hour period in March 2001 for emissions from Tarapur. Figure A2.3 shows the dispersal after 12 hours.

Releases from Military and Civilian Reprocessing

HYPSLIT-4 was run for a 48-hour period in August 2001 to investigate possible mixing of releases from the Trombay military reprocessing facility with the larger civilian reprocessing plants at Tarapaur and Kalpakkam (see Figure A2.4).

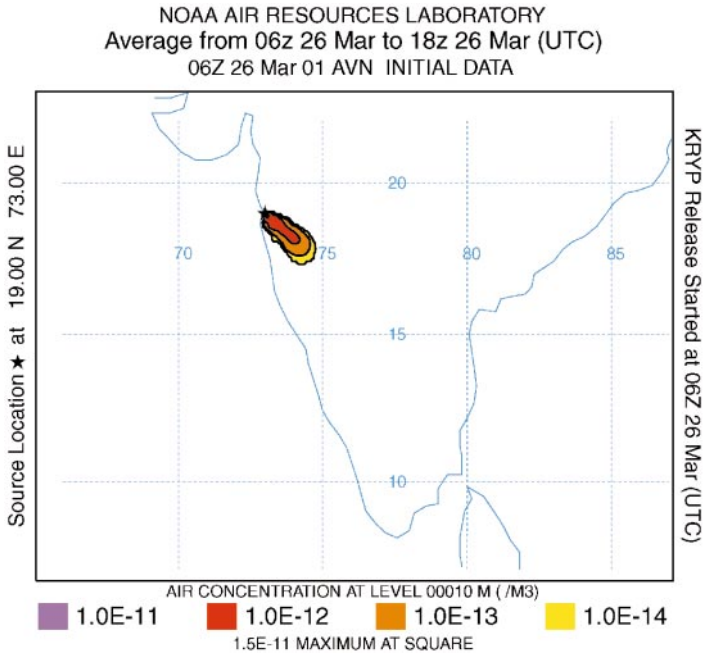

Figure A2.1: Dispersion contours of ^{85}Kr released from the Trombay reprocessing plant.

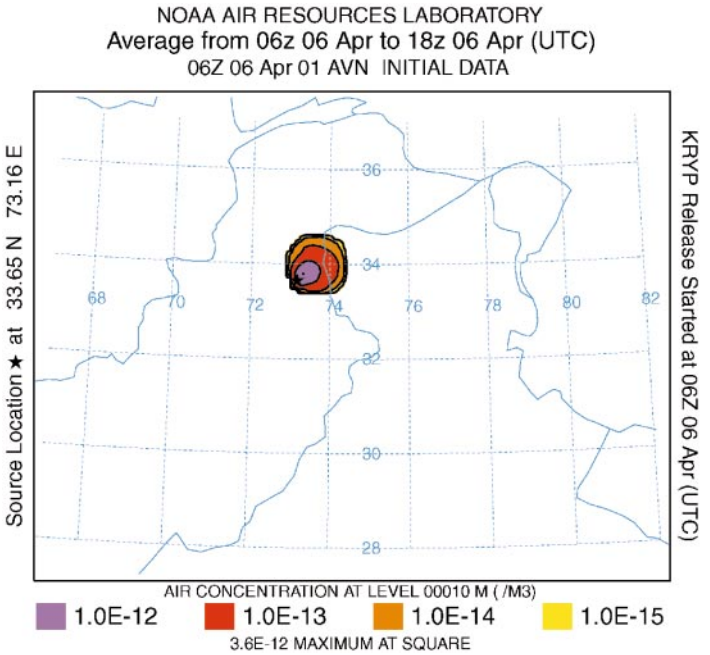

Figure A2.2: Dispersion contours of ^{85}Kr released from the New Labs reprocessing plant.

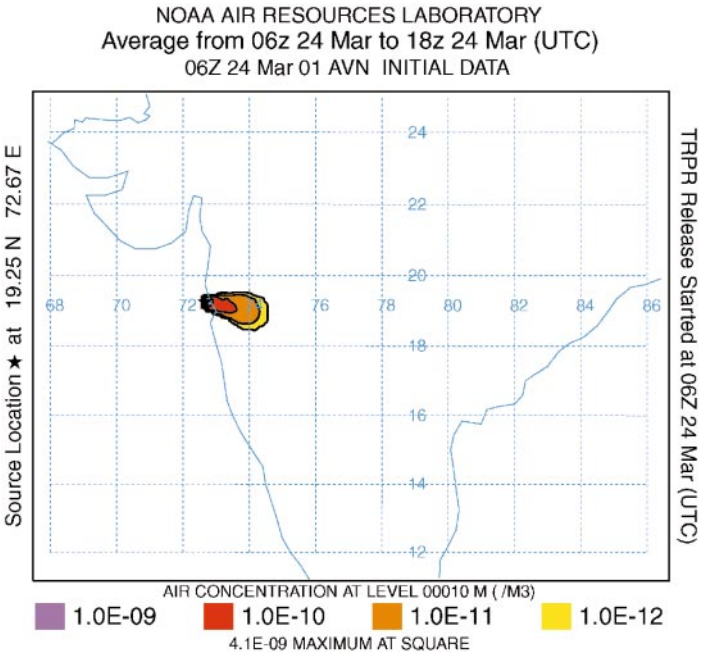

Figure A2.3: Dispersion contours of ^{85}Kr from the Tarapur reprocessing plant.

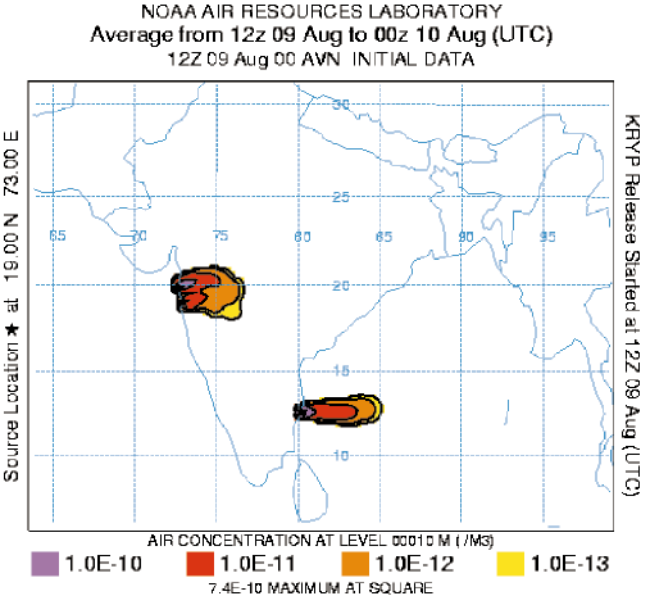

Figure A2.4: Dispersion contours of ^{85}Kr released from the Trombay, Tarapur, and Kalpakkam reprocessing plants.