CHAPTER IV

DEPOSITION MEASUREMENTS USING GUMMED-PAPER COLLECTORS

 Gummed paper was used to measure fallout deposition at locations throughout the United States during the 1950s and 1960s (Eisenbud and Harley [1953, 1955, 1956, 1958; Harley et al.](#page-18-0)

1960). Deposition was measured daily at 40 to 95 locations during major periods of nuclear weapons testing at the Nevada Test Site (NTS) ([Beck et al.](#page-18-0) 1990; [Hoffman et al](#page-18-0). 1992). Most of these data have been retrieved and compiled for use in an assessment of radioiodine doses to thyroids of persons in the continental United States from weapons testing at the NTS [\(Wachholz 1](#page-19-0)990).

Radionuclides in air are subject to deposition by processes such as gravitation and washout by precipitation.

 Gummed-paper collectors were used periodically to measure radioactive fallout at and around the Rocky Flats Plant (RFP) during the years 1954−1972. The earliest references to fallout monitoring were found in 1954 site survey reports, although no data were reported. Fallout monitoring is not mentioned again in the site survey reports until May 1963. Data were found for the following periods: (a) May 1963 through June 1964, (b) May 1965 through December 1965, and (c) January 1970 through June 1972. Although continuous measurements are not available for the entire time period of interest, the available data indicate some temporal and spatial trends for those years (see Chapter I).

DOW MONITORING LOCATIONS AND METHODS

Early Tests (1954−**1955)**

 The first reference to fallout monitoring can be found in the *Site Survey Monthly Progress Report* for May 1954. This report states,

A prototype fallout tray was installed at a location near 44 bldg on May 14. Two weeklong samples of Whatman No. 1 filter paper have been collected and subjected to autoradiograph.

The *Site Survey Monthly Progress Report* for June 1954 notes

Another sample was collected from the fallout tray near 44 building. A gummed paper was used to collect this sample. The fallout tray was then moved to a location about 1,000 feet southeast of the 71 building stack. One sample of Whatman No. 1 paper has been collected from this location. Autoradiographs of these papers have all shown a slight deposition (less than 10 particles per square foot) of radioactive particles.

Additional onsite locations were monitored at different times during the remainder of 1954, including areas near Building 23, northwest of Building 71, near the Building 81 parking lot, and at some air sampling stations. Autoradiographs were used to detect any radioactive particles.

Particles were detected only once, during a special study conducted at the $NO₃$ pond (the original solar evaporation pond) area. According to the *Site Survey Monthly Progress Report* for December 1955,

Fallout paper was used to pick up dust particles that originated from the $NO₃$ pond on November 10th. The average wind velocity during the work hours was approximately 40 mph from the west.

A pulse-height analysis showed the presence of ²³⁵U (0.3 pCi), ²³⁸U (0.9 pCi), and ^{239,240}Pu (0.05 pCi). Before 1957, the releases of long-lived alpha activity from uranium areas at the RFP were larger than those from plutonium areas. For further discussion of this issue, see "Apportioning Total Long-Lived Alpha Count into Specific Radionuclides" in Chapter III.

Routine Monitoring (1963−**1965)**

 There is no further mention of fallout monitoring being performed until the May 1963 *Site Survey Monthly Progress Report*. According to that report, a series of 10 locations for plutonium fallout monitoring were set out east and southeast of the Building 71 stack, between that point and the east cattle fence, which was the boundary of the RFP at that time. The June 1963 *Site Survey Monthly Progress Report* notes that nine additional fallout trays were placed in service to the west and south of Building 71 stack. The 19 monitoring locations are shown in [Figure IV-1](#page-2-0) and listed in T[able IV-1.](#page-2-0)

 The August 1964 *Site Survey Monthly Progress Report* states that the station located 1417 m (4650 ft) east-southeast of the Building 71 stack (Station 3 in [Table IV-1\)](#page-2-0) was abandoned because of construction of a new road. The monitoring program was modified in 1965 to include more

The abbreviation dpm stands for disintegrations per minute, which is a measure of radioactivity. There are 2.22 dpm per picocurie (pCi).

locations east-southeast of the Building 71 stack and to exclude those locations where plutonium was rarely detected. Although the monitoring network was changed, 11 of the 19 stations were located at or near positions monitored in 1963−1964 (T[able IV-2\).](#page-3-0) Thus, comparison of measurements over the entire period from 1963 through 1965 was possible for some locations. There were no offsite areas monitored at

this time. Results during 1963 through 1965 were reported as plutonium deposition rate (disintegrations per minute per square foot per week). We have not found a description of the methods used during this time period.

Routine Monitoring (1969−**1972)**

 Monthly site survey reports could not be located for the years 1966 through 1969, so the evolution of the deposition monitoring program during this interval is unknown. Data documented in the *Semiannual Environmental Survey Report* for July to December 1969 shows that monitoring of 12 offsite locations (Arvada, Boulder, Broomfield, Coal Creek, Denver, Eastlake, Golden, Lafayette, Marshall, Superior, Wagner Station, and Westminster) began in July 1969. This report provides cumulative plutonium deposition rates (picocuries plutonium per

Table IV-1. Locations of Deposition Measurements on the Rocky Flats Plant Site during 1963−**1964**

 \triangle - GUMMED PAPER MONITORING STATION

Figure IV-1. Gummed-paper monitoring stations during 1963−1964.

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square meter per month) at each location, estimated from weekly measurements, for the periods from July to December 1969.

 The *Rocky Flats Environmental Monitoring Results, January 1 through March 31, 1970,* provides biweekly data (picocuries plutonium per square meter) for the 12 offsite locations. Three onsite locations (at air monitoring stations S-5, S-8, and S-50) were added in May 1970 ([Figure IV-2\).](#page-4-0) The onsite network was expanded to include air monitoring station $S-51$ in July and station S-1 in September 1970. Remote locations were also added in October 1970. The control measurements were generally alternated between Berthoud, Castle Rock, and Alamosa, Colorado. Collection frequency for measurements made during 1970–1972 varied from 10 days to 3 months. In the August 1972 report, it was noted that the program was discontinued. The report provides the rationale in the following statement:

Pu conc. in dustfall samples will not be shown in this or subsequent reports due to their uninterpretable nature. Samples will continue to be taken as modifications of sampling and analysis techniques are being evaluated.

 Specific methods used in the early 1970s were not located. However, handwritten notes on the June and July 1971 *Rocky Flats Monthly Environmental Reports* say "sticky paper" and "A =

0.07 m²," indicating that gummed paper, with an exposed area $7/100$ m², or approximately 10 in.², was used. Also, where a value is missing in the monthly report data table, typically there was a comment of "no sample" due to "wind." This perhaps indicates that the paper was not

Figure IV-2. Gummed-paper monitoring stations during 1970−1972.

anchored in a holder typically used in other deposition monitoring programs. The analytical method used for determining the plutonium content of the samples may have been a liquid scintillation counting method documented in [Bokowski \(](#page-18-0)1974). This method was used for other environmental samples collected at Rocky Flats in the early 1970s.

Additional Deposition Measurements

 A laboratory analysis sheet was found that indicated three papers, located along the perimeter fence, were collected in October 1974 and analyzed for plutonium isotopes and for gamma-emitting radionuclides (D[ow 1](#page-18-0)974). In addition, a precipitation collector (a 55-gal drum lined with polyethylene) was placed on Niwot Ridge from late December 1975 to June 21, 1976, to obtain a background measurement for ^{90}Sr , ^{238}U , $^{239,240}Pu$, ^{241}Am , and tritium ([Cleveland et al.](#page-18-0) 1976). The results were presented, in terms of activity per unit area, along with the comment that:

Plans call for continuous collection of precipitation and analysis at intervals of approximately six months. In this way it is hoped that a bank of useful data on atmospheric fallout in the Front Range area of Colorado may be established.

No subsequent reports on deposition monitoring by the RFP contractor were found.

RESULTS

 Ideally, these monitoring data can be used to estimate deposition rates of plutonium from airborne effluents. For example, uranium deposition data collected, using gummed paper, around the Fernald Feed Material Production Center were used in the Fernald Dosimetry Reconstruction Project to validate the source term and deposition models used ([Killough et al. 1](#page-19-0)998). However, quantitative calculations require knowledge of the methods used and a calculation of the collection efficiency of the paper, which is highly variable, particularly during precipitation. Figure IV-3 shows collection efficiency plotted versus precipitation rate (millimeter during a 24-hour period) as reported in [Beck et al. \(1](#page-18-0)990). The data were obtained through experimental simulation, using naturally occurring 7 Be in rain as an analog for fission product fallout; through use of results collected following the reactor accident at Chernobyl, Ukraine; and the 1957 PLUMBOB test series conducted at the NTS. Because the three datasets indicate a similar dependence on precipitation intensity, they were combined to make estimates of the average collection efficiences for various precipitation intensities. The estimated collection efficiency for dry deposition is 20% and it ranges from $7-30\%$ for high (>25 mm) and low (<0.75 mm) precipitation amounts in a 24-hour period.

These results are consistent with those reported in H[offman et al. \(](#page-18-0)1992). Collection efficiencies for two soluble radionuclides, ^{131}I and ^{7}Be , estimated using simulated low (19.7 mm h⁻¹ [0.78 in. h⁻¹]) and high (104 mm h⁻¹ [4.09 in. h⁻¹]) rainfall intensities under field conditions ranged from about 30% at 2.5 mm [0.10 in.] of rain to 4−6% at 20 mm [0.79 in.] of rain. The results for the low intensity study, more typical of rainfall events at Rocky Flats, are plotted in [Figure IV-4.](#page-6-0) There is little difference between the collection efficiencies for 131 I and 7 Be, which were also reported in **Beck et al.** (1990).

Figure IV-3. Estimated collection efficiency of gummed paper (from [Beck et al. 1](#page-18-0)990). The estimated uncertainty for each efficiency estimate is approximately $\pm 25\%$ of the values given (1 standard deviation).

[Hoffman et al. \(1](#page-18-0)992) noted that rain intensity seemed to have little effect for these radionuclides and that the measurements reported in [Beck et al. \(](#page-18-0)1990) over a 24-hour period are more appropriately considered as total rainfall amount rather than rainfall intensity. The Hoffman et al. report concluded that collection efficiency of these soluble nuclides on gummed paper was simply a matter of rapid saturation and runoff. These particles settle readily into the surface of the gummed paper, and they are not easily removed by additional rain. The portion of the soluble particles that does not settle on the gummed paper was found in the standing water present on the paper. Thus, considerable bias could be introduced into the results if the standing water is discarded before analysis.

[Hoffman et al. \(](#page-18-0)1992) also examined the retention of large insoluble particles, simulated using three size classes of polystyrene microspheres $(3, 9, \text{ and } 25 \text{ µm})$, which were marked with radioactive tracers. Average values of the collection efficiency for the microspheres ranged from 20−50% for the 3- and 9-µm microspheres to about 50 to 80% for the 25-µm microspheres. [Figure IV-5 s](#page-7-0)hows the results for the low intensity (19.7 mm h⁻¹ [0.78 in. h⁻¹]) rainfall study. With the exception of the 3-µm particles, the influence of the amount and intensity of rain on the collection efficiency was essentially negligible. The authors concluded that the larger particles rapidly settled onto the surface of the gummed paper and that, once settled, they were not readily removed by additional rain. Analysis of the standing water present on the gummed paper indicated that nearly all of the 25-µm particles (94%) and most of the 9-µm particles (>70%) had settled on the gummed paper. By contrast, only 50% of the 3-µm particles, 20% of the ^{131}I activity, and 10% of the 7 Be activity were attached. The rest of the retained activity was actually in the standing water.

Figure IV-4. Average values ($n = 2$) of the collection efficiency of ¹³¹I and ⁷Be by gummed paper for rain applied at low (19.7 mm h⁻¹ [0.78 in. h⁻¹]) intensity (from [Hoffman et al.](#page-18-0) 1992). Trendlines are power function fits to the data.

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Figure IV-5. Average values ($n = 2$) of the collection efficiency of 3, 9, and 25-µm microspheres by gummed paper for rain applied at low (19.7 mm h^{-1} [0.78 in. h^{-1}]) intensity (from [Hoffman et al. 1](#page-18-0)992). Trendlines are power function fits to the data.

 Plutonium deposited on gummed paper at Rocky Flats has two predominant sources: stack effluents and airborne contamination from the 903 Area. It can be assumed that the oxide form, which is highly insoluble, predominates in the environment. In a study of plutonium particle sizes in surface soil within the buffer zone east of the site, [Hayden et al. \(](#page-18-0)1994), using autoradiographic techniques, demonstrated that these two sources could be distinguished in the field. The particle size distribution showed an overall mean particle size of 0.08 μ m (i.e., physical diameter). It was observed by the authors that this was similar to the 0.09 micron mean size measured in the stack effluent from 776 building and that this inferred that stack effluent was the prime source of the widely distributed particulates observed in the buffer zone. However, an additional group of particles directly east of the plant (i.e., downwind of the 903 Area) were superimposed on this distribution. The larger particles found here $(0.5 \mu m PuO₂)$, having an equivalent unit density size of about 1.7 µm) could not have effectively penetrated high-efficiency particulate air filters and, thus, probably came from the contaminated 903 Area. The authors note that particles measured within the 903 Area had been found as large as 2 to 3 μ m, and a mean particle size of 0.3 μ m was measured on one sample excavated from the area.

 The range of plutonium particle sizes measured in the field is much smaller than that used in the collection efficiency study documented i[n Hoffman et al. \(](#page-18-0)1992). It could be assumed that the collection efficiencies of the smaller particles approach those of the more soluble species, 131 I and ⁷Be. In addition, precipitation at Rocky Flats is typically <0.1 mm d⁻¹ [0.00394 in. d⁻¹], while the Hoffman study focused on rainfall rates >2.5 mm [0.10 in.] per collection period. For these reasons, it was decided that the collection efficiencies estimated in [Beck et al. \(](#page-18-0)1990) were more appropriate estimates for the gummed paper used at Rocky Flats.

 Precipitation data obtained at Boulder by the National Oceanic and Atmospheric Administration (NOAA) for the periods from 1963–1965 and 1970–1972 were used to estimate the collection efficiencies of the gummed paper employed in the Rocky Flats deposition monitoring program. We obtained the data from the NOAA web site on the Internet. Using the data from B[eck et al. \(1](#page-18-0)990), shown in F[igure IV-3, w](#page-5-0)e assigned a collection efficiency to each day in the measurement period based on the precipitation rate for that day. We then calculated average collection efficiencies for the specific period of collection (typically 1 to 2 weeks) and used the measurement for that period.

 Although the Boulder precipitation rates are not strictly equivalent to Rocky Flats precipitation rates, we determined, through comparison of Rocky Flats and Boulder data from later years, that the error associated with using collection efficiencies estimated with Boulder data is small. Precipitation was measured at Rocky Flats in 1972, 1973, and 1974 by the U.S. Geological Survey (USGS) ([USGS](#page-19-0) 1976) and in 1990, 1991, and 1992 by EG&G Rocky Flats, Inc. ([EG&G 1](#page-18-0)993). We estimated weekly average collection efficiencies using these data and the NOAA Boulder data. We estimated the ratio of the weekly average collection efficiency calculated for Rocky Flats to the weekly average collection efficiency calculated for Boulder for the two datasets (i.e., 1972–1974 and 1990–1992). A ratio equal to 1 means that the two datasets are equivalent. A ratio >1 indicates that the Boulder data underestimate the Rocky Flats value. A ratio <1 indicates that the Boulder data overestimate the Rocky Flats value. Plots of the frequency distribution of the ratios indicate a normal distribution of values around a mean value of 1.0 for each dataset (Figures [IV-6 a](#page-9-0)nd I[V-7\).](#page-9-0) The small range of values represented by the 95% confidence interval for each data set shows that the bias introduced by using the Boulder data is small. The bias decreases further when applied to monthly averages. Thus, the longer the period of collection, the smaller the error associated with using the Boulder data for estimating the collection efficiency. We applied the collection efficiencies estimated using the Boulder data to both onsite and offsite locations.

 Plutonium deposition rates measured at the onsite locations monitored from 1963–1965 and corrected for gummed-paper collection efficiency are presented for each month in Figures [IV-8,](#page-10-0) $IV-9$, and $IV-10$. The uncorrected results, as reported by Dow, are presented in Appendix C. The highest result (12 nCi m⁻²) was observed at station 16, located 0.8 km (0.5 mi) south-southeast of the Building 71 stack during May 1964. There are no strong temporal trends in the data, although the deposition rates appear to be lower in the fall of 1963 and higher in the winter of 1963–1964 and in the spring/summer of 1964. Spatially, the stations located in the southeast quadrant, relative to the Building 71 stack, seem to have higher deposition rates than those located in other directions. An evaluation of the relationship between deposition rate, direction, and distance shows that the deposition varies as a function of the latter two factors.

Figure IV-6. Ratios of collection efficiencies calculated for gummed paper using precipitation data measured at Rocky Flats and Boulder in 1972, 1973, and 1974. The ratio is a measure of the possible bias associated with using Boulder precipitation data to represent Rocky Flats precipitation during periods of time when precipitation was not measured at Rocky Flats.

Figure IV-7. Ratios of collection efficiencies calculated for gummed paper using precipitation data measured at Rocky Flats and Boulder in 1990, 1991, and 1992. The ratio is a measure of the possible bias associated with using Boulder precipitation data to represent Rocky Flats precipitation during periods of time when precipitation was not measured at Rocky Flats.

Figure IV-8. Plutonium deposition rates measured using gummed paper at stations located in the north to east directions from Building 71 during 1963−1965. Correction has been made for collection efficiency.

Figure IV-9. Plutonium deposition rates measured using gummed paper at stations located in the southeast directions from Building 71 during 1963−1965. Correction has been made for collection efficiency.

Figure IV–10. Plutonium deposition rates using gummed paper measured at stations located in the south to northwest directions from Building 71 during 1963– 1965. Correction has been made for collection efficiency.

[Figure IV-11 il](#page-12-0)lustrates the observed dependence of the cumulative deposition on gummed paper with distance from the Building 71 stack during the period from 1963−1965. The figure demonstrates the source of plutonium (Building 71 stack) and the depletion of plutonium in air as a function of downwind distance. Note that only 9 of the 19 stations were included in this analysis because they were the only stations that had continuous measurements during this period. [Figure IV-12](#page-12-0) illustrates that cumulative plutonium depositions are a function of direction, as well as distance, from the Building 71 stack. Higher depositions can be observed in the east-southeast direction and at distances closer to the stack.

 Unfortunately, the onsite data obtained from the early 1970s are not extensive enough to assess any spatial trends. However, because offsite data are available from this period, it was possible to compare onsite with offsite measurements. An analysis of variance (ANOVA) of the data collected at the 12 offsite stations (Arvada, Boulder, Broomfield, Coal Creek, Denver, Eastlake, Golden, Lafayette, Marshall, Superior, Wagner Station, and Westminster) demonstrates that there are no statistical differences between these offsite stations. For this reason, the data from these stations were averaged to represent the offsite area in the vicinity of the RFP. Further statistical comparisons demonstrated that data collected at only two of the five onsite locations monitored during this period were statistically different from measurements made at the offsite stations. The two stations are S-5, which is located near the solar evaporation ponds, and S-8, which is located near the 903 Pad. [Figure IV-13 d](#page-13-0)emonstrates that 82% of the deposition rates measured at S-8 were higher than the offsite average rates.

Figure IV-11. Total plutonium deposition estimated from gummed-paper data from May 1963 through November 1964 as a function of distance from Building 71 stack. Data have been corrected for collection efficiency.

Figure IV-12. Total plutonium deposition estimated from gummed paper as a function of distance and direction from the Building 71 stack in 1963 and 1964.

Figure IV-13. Comparison of offsite average and maximum plutonium deposition rates with deposition rates estimated for stations S-5 (solar ponds) and S-8 (903 Area). Data have been corrected for collection efficiency.

Figure IV-14 summarizes the trend for deposition rates, averaged over the entire period. The higher deposition rates measured near the solar evaporation ponds and the 903 Area are most likely due to deposition of locally suspended contaminated soil.

Figure IV-14. Comparison of plutonium deposition rates, averaged over the period January 1970 through June 1972. Data have been corrected for collection efficiency.

 Figures C-10 through C-12 in Appendix C indicate no discernable patterns in offsite measurements from 1970 through 1972. The data are too erratic to make any conclusions about deposition of contaminants originating from the RFP onto offsite areas near the RFP during this period. Moreover, there are no statistical differences between the datasets collected offsite. If the RFP had an offsite impact during this time period, the locations closest to the site, which lie in the predominant wind direction, would be expected to demonstrate higher results than locations farther away and upwind of the predominant wind path. For example, the Wagner site located just west of Standley Lake (see Figure B-5), lies in an area that has been shown, through soil and sediment sampling, to have been contaminated by releases of plutonium from the 903 Pad in 1969 (see Chapters \underline{VII} and \underline{VIII}). Thus, higher deposition rates of resuspended plutoniumcontaminated soil might be expected at the Wagner site. However, using a single factor analysis of variance, there was no statistical difference between data collected from this site and data collected from sites farthest away and not in the predominant downwind path of Rocky Flats (e.g., Boulder, Marshall, and Lafayette). Figure IV-15 shows the cumulative monthly deposition results for these offsite locations.

 Based on the following observations, we did not perform a comparison of offsite and remote location data because we concluded that the latter measurements were unusable for this purpose. First, the remote location was rotated between the three sites (Alamosa, Berthoud, and Castle Rock) so that only one measurement was taken during each sampling period. Offsite data collected during the remote sampling period, from October 1970 through May 1972 represent 432 measurements taken at 12 locations, while data collected at remote locations incorporate only 31 measurements obtained at three alternating stations. Second, the Alamosa station, located in south-central Colorado, is probably not representative of the front range environment. Finally, many of the remote measurements were made for periods substantially longer (sometimes as long as 3 months) than the biweekly sampling period typical of the offsite program. No references could be found that address the impact of long collection periods on collection efficiency. However, longer periods of collection would be expected to decrease the collection efficiency because of an increasing dust load. A plot of the data versus the collection period appears to support this assumption (Figure IV-16). For these reasons, it was concluded that it would be inappropriate to compare remote station data with offsite data.

Figure IV-16. Remote location measurements as a function of collection period. Data have not been corrected for collection efficiency.

Ideally, a comparison could be made of onsite data collected during the early 1960s with those collected during the early 1970s. However, the change in onsite locations makes this difficult. Only one station in the later monitoring network was situated near an onsite station in the earlier monitoring network. Station S-5, located near the solar evaporation ponds, is close to gummed-paper station 8, which was used in the 1960s. Figure $IV-17$ shows the data from these two stations plotted as a function of time. Not surprisingly, the plutonium deposition rates measured in the early 1960s are generally greater than those measured in the early 1970s. This is not unexpected because relatively higher stack releases and global nuclear weapons testing fallout occurred during the earlier years. The S-5 air sampler location showed little airborne activity from 903 Area activity in 1970−1971. The only obvious peak in airborne contamination at that location was in June 1970 (Figure III-31). The gummed-paper activity does not appear to agree with the time trend in airborne activity for 1970−1971.

Figure IV–17. Comparison of plutonium deposition rates measured at an onsite area near the solar evaporation ponds from May 1963 to November 1965 and from February 1970 through May 1972. Data have been corrected for collection efficiency.

SUMMARY

 Gummed-paper data were obtained from monthly environmental monitoring reports from 1963−1965 and from 1970−1972. The results were corrected for incomplete retention of plutonium by the gummed paper using the collection efficiency factors developed by [Beck et al.](#page-18-0) (1990). The factors vary as a function of precipitation. Precipitation data measured at Boulder during the time periods of interest to estimate the collection efficiencies of the gummed-film collectors located at Rocky Flats and at offsite locations. The corrected results provide some information on spatial and temporal trends in plutonium deposition on the RFP. Specific observations about the data are

• The data from 1963−1965 show spatial patterns of deposition of plutonium onsite from releases from the Building 71 stack. The data could be used to an air disposition model, although the error associated with the results is largely unknown. Thus, the deposition

pattern may only be useful in a relative way to confirm patterns produced by model estimates.

- Elevated spring fallout from nuclear weapons testing was evident during the spring of 1963–1965.
- The data from 1970−1972 demonstrate localized sources of elevated deposition, specifically resuspension of contaminated soil from the 903 Area and the solar ponds, demonstrating that they exceed stack releases in relative impact during that period.
- No conclusions could be made about the possible impact of the RFP on the offsite area near the RFP during 1970−1972. This is because the offsite data are too erratic to make any conclusions, and data collected from areas far from the RFP (i.e., remote areas) were not usable for comparison with offsite data.
- The data do not appear to be useful for understanding 903 Area releases.

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