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An evaluation of 1957 to 1966 data to assess residual radiation from nuclear weapons sites in the U.S.

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by

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ABSTRACT OF THE THESIS

An evaluation of 1957 to 1966 data to assess residual radiation

from nuclear weapons sites in the U.S.

by

Chih-Jen Cheng

Master of Science in Civil Engineering

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Professor Michael K. Stenstrom, Chair

Between 1950s and 1960s, active bomb tests produced many radionuclides and exposed radiation to the environment. This thesis addresses the radioactivity caused by strontium-90 (^{90}Sr), a beta-only emitter and the health concerns it has caused. Through the bomb detonations, radioactive compounds were carried by atmospheric transport globally. This research is conducted to analyze the pattern of distribution and the inventory of ^{90}Sr. Data from a total of 53 sites were collected from public records and seven, four and eight sites were further analyzed for gross beta in milk, air and raw surface water, respectively. Monthly

concentrations of ^{90}Sr and the values of gross beta from 1957 to 1966 are presented in multiple figures, and shows that radionuclides are not transported in predictable directions. By comparing the concentrations of ^{90}Sr to the gross beta detected in the local ecological systems, a relation between gross beta and ^{90}Sr is examined, and eventually concluded to show a weak linkage between them. Distance to the bomb test sites is not proportional to the amount of ^{90}Sr detected at the inspection stations. The results also show a 2-year atmospheric residence time before ^{90}Sr settles down. It is suggested that precipitation (wet deposition) may play a more significant role of enhancing gravity settling of ^{90}Sr in different areas, and more factors should be considered for the analysis.

Keywords: radionuclides, bomb tests, ^{90}Sr, gross beta, atmospheric transport, precipitation, wet deposition

1. Introduction

Nuclear power developed in the 20th century due to military needs, and the first bomb test was conducted in the United States in 1945. The most significant and frequent nuclear activity was the nuclear bomb testing, which have been documented in various reports and literatures. Both of the underground and atmospheric tests have been examined and their ability to release radioactive fallout (Herranz et al., 2011) has been confirmed. From 1945 to the present, 2054 bomb tests have been conducted globally, in which 1030 and 715 tests were conducted by the United States and Russia, respectively (Arms Control Association, 2016). Fig. 1 shows the number of bomb tests conducted by the eight nuclear weapons-processing countries from 1945 to the present. Atmospheric detonations were mainly experimented from 1951 to 1962, with the yield ranging from less than 20 kilotons (kt) to several megatons (Mt). After July 1962, the U.S. ceased atmospheric testing because of the concern over the spread of the radiation, and limited testing to underground tests. The US government conducted its last test in 1992. Since 1945, 904 weapons tests were conducted at the Nevada Test Site, approximately 100 km northwest of Las Vegas (Nevada Operations Office, 2016). It has been pointed out that the low-level radiation exposed from the 26 Nevada sites from 1951 to 1958 had affected the whole U.S., especially those regions located at the downwind of Nevada

(Beck et al., 1983) (Lyon et al., 1979). Following the example of the United States, the Soviet Union, France, the U.K., China and later India, Pakistan and North Korea all demonstrated their capabilities and continued with their tests (Arms Control Association, 2016). The first non-US test occurred in 1949 in Semipalatinsk, Kazakhstan. Fallout from the bomb tests entered the atmosphere and was transported world-wide often in unanticipated directions.

The U.S. government implemented bomb testing in its territories across the Western Hemisphere. Operation Argus, was the only operation conducted in the Atlantic Ocean, and was kept secret. Test sites in the Pacific Ocean include: Bikini Atoll, Enewetak Atoll, Johnston Island, Christmas Island, and the Central Pacific Ocean. Other sites distributed inland of North America include: Nevada, San Diego, Alaska, and Mississippi. Besides Nevada test sites, which had greatest intensity of tests in 1960s, other sites inland only had one test each. Fig. 2 shows the monthly yield each site. Every bomb test involved complex reaction and released active radionuclides, including polonium-210 (^{210}Po), cesium-137 (^{137}Cs), strontium-90 (^{90}Sr), uranium-238 (^{238}U), etc. to the environment, during and after detonations (REMM, 2016). High-level radiation is believed to generate carcinogenic effects, such as leukemia and thyroid cancer, in humans and animals. Internal and external exposure

to radioactive materials cause penetration and accumulation of radiation in the tissues (Mettler and Voelz, 2002). Though radioactive fallout has not been confirmed to have the same impacts as high-radiation does, a risk still remains and the potential threat is concering (Lyon et al., 1979). Several reports show that the residence time and distribution of the fallout, from above ground tests are influenced by the bomb yield and the height of the detonation. Warneke et al. (2002) indicates that when the explosive yield of the tests are lower than 100kt, which is categorized as small scale, the radioactive debris can reach the troposphere, and reside for at most 70 days in the atmosphere. For a relatively large test, which generates energy more than 500 kt, radioactive compounds can be injected into the stratosphere, transporting the radionuclides worldwide. The Chernobyl and Fukushima accident, demonstrated that fallout can distribute globally.

In order to show a link between distribution of fallout and the occurrence of the bomb tests, data were assembled to track radionuclides in soil, fallout samples, water, and milk. Databases were acquired from the U.S. government and various official firms, and present the comparison of regions among the U.S.A.

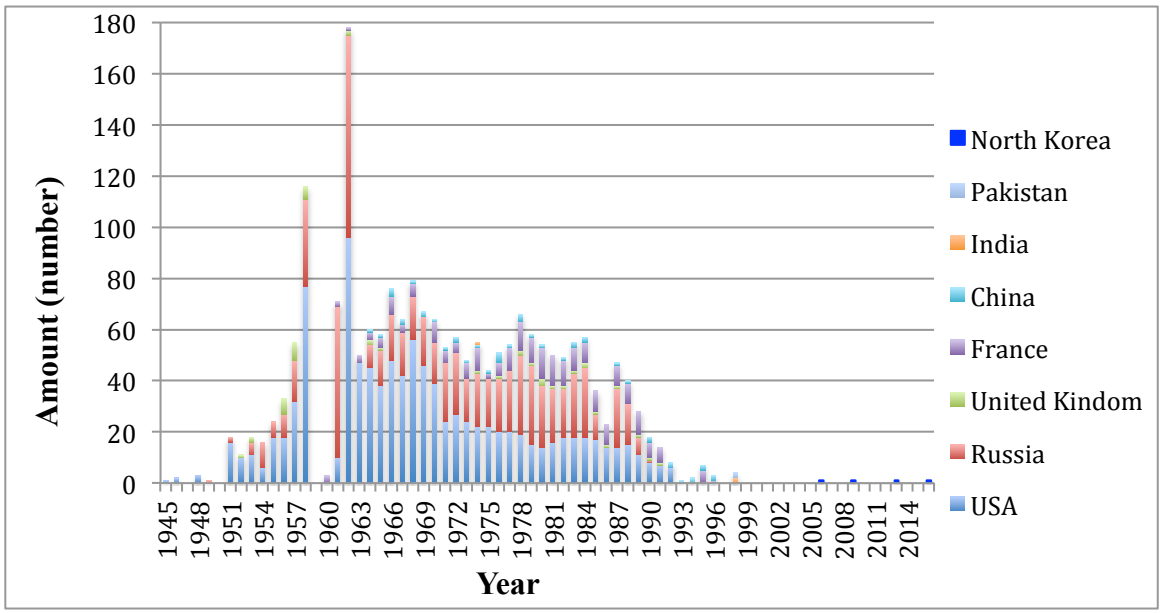

Fig. 1. Accumulated bomb test in the world from 1945 to 2016 (Arms Control Association, 2016)

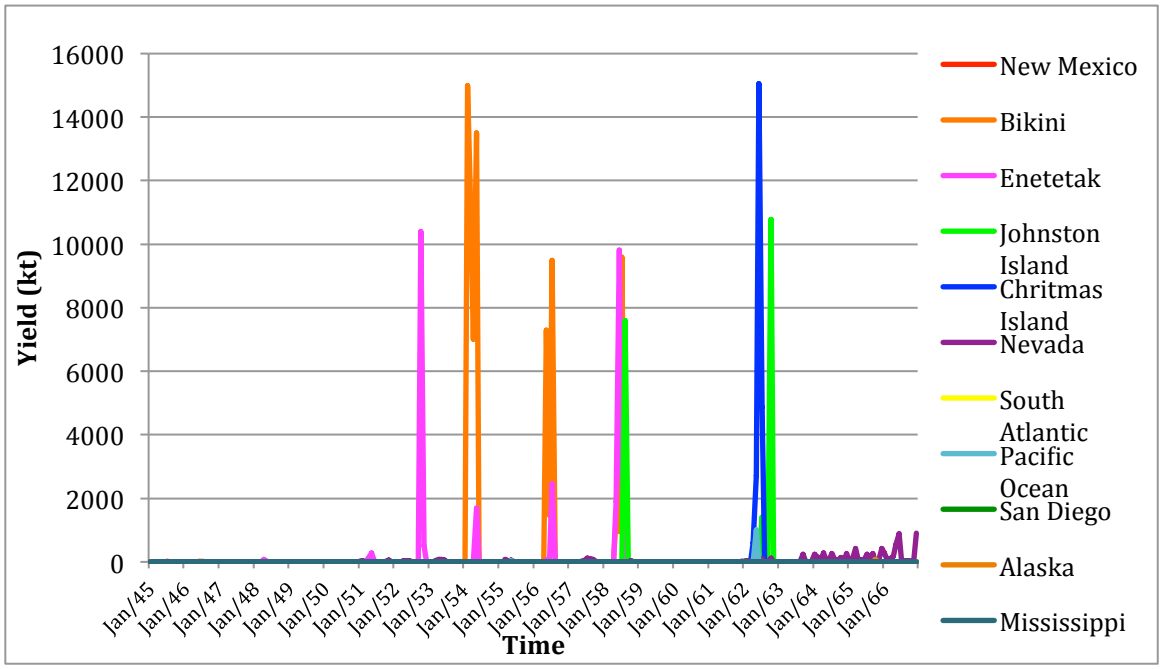

Fig. 2 Monthly yield of bomb tests in various sites from January 1945 to December 1966 (United States Department of Energy, 2015)

2. Establishment of the database

Radioactive materials can be categorized as alpha emitters, beta emitters, and multiple-radiation emitters. Each radionuclide has different half-life; therefore, the residence time it remains active and damage it caused to the environment is different. Furthermore, the residence time can also be affected by the yield of the nuclear explosions based on a research done by Warneke et al. (2002). For instance, explosions which generated more than 500 kt of yield can cause radionuclides to penetrate into the stratosphere, and keep them settling out for years. Consider the radioactive effects induced by the half-life and bomb yield, we chose to focus on the following two types of radionuclides or radiation emitter:

Strontium-90 (^{90}Sr)

Gross beta radiation

^{90}Sr was chosen due to a monitoring program conducted by EML (Environmental Measurements Laboratory) which collected a large database and is available for analysis. As ^{90}Sr can only release beta radiation, an investigation of the beta radiation is needed and used to correspond to the effect of ^{90}Sr in this thesis.

The time period chosen in this thesis is 1957-1966, which can be further divided to three time spans: 1957-1958, 1959-1960, and 1961-1966. The first time span represents a period of

intense testing; the second period was a voluntary cessation of testing by both the U.S. and Russia; the final period consists of a redevelopment of tests and the beginning of underground tests.

2.1 Introduction of radiation

Before the analysis, details of the radionuclides and different radiations have to be clarified. The following section provides an overview.

Alpha radiation: Alpha particles are the particles acquiring relatively heavy and positive charged nuclei. An alpha particle has two protons and two neutrons. By releasing an alpha particle, which is known as the form of alpha radiation, the element is transferred to another element due to loss of protons. Alpha particles, while highly energetic do not penetrate because they are charged and large. They lose energy quickly and easily through air, so they can barely penetrate a sheet of paper; they are retained in front of the paper. While alpha particles cannot penetrate skin, research has shown that if the alpha emitters are ingested, they can be retained, releasing alpha particles, and create significant damages to tissues and organs. Thus, alpha radiation is regarded as a threat and cause of cancer occurrence (Cheney, 1996; Arnold, 2007).

Beta radiation: Beta radiation occurs while a beta particle is released from the nucleus. Each beta particle can be regarded as an electron, because it has one negative charge. Once a radioactive atom releases beta particles, the atom will be changed to a new element. Because beta particles are lighter, smaller, and move faster than alpha particles, they penetrate deeper and can travel farther. High-energy beta particles are able to penetrate human skin and travel inside the human body; however, because of their low energy, external exposure to beta particles tends to have less negative effects on human health (Cheney, 1996; Arnold, 2007).

Multiple authors have pointed out that the biological impacts caused by ingesting alpha radiation can be as much as one-hundred times more severe than beta radiation although the danger of beta radiation is not ignorable. Warren (1980) indicated that while many radionuclides can release both of the alpha and beta particles, some can only release beta particles and still impact their surroundings.

^{90}Sr: Strontium has four stable isotopes and sixteen radioactive ones. ^{90}Sr has the longest half-life, which can be approximately 28.9 years, while others have less than 65 days half-life. The decay of ^{90}Sr transforms the radionuclide to form ^{90}Y (Yttrium-90, stable) by releasing one beta particle. The yield of ^{90}Sr produced through nuclear fission is approximately 6%,

which means six atoms of ^{90}Sr are generated from one hundred fissions (ANL, 2001). Among many radionuclides generated from the bomb testing, ^{90}Sr has a relatively long half-life and its high yield which makes it one of the more important fission byproducts. For these reasons, several environmental and health organizations to include it in their surveillance systems. Because strontium is similar to calcium in biological functions, it is easily uptaken and retained by the humans. It can affect the food chain and be retained in humans (Paatero et al., 2010).

2.2 EML (Environmental Measurements Laboratory) databases

EML is wellknown for its outstanding success in performing radiation monitoring. Its predecessor was the Atomic Energy Commission (AEC), which was established in 1946, after the World War II. As the U.S. government began the launch of the Manhattan Project to develop nuclear weapons, EML was designated to monitor radioactivity of fallout from the bomb tests by extending the research to global levels. This paper only focuses on the territories of the U.S., which include the North America but also to US territories in the Atlantic Oceans and the Pacific Oceans. The strontium concentrations and distributions are

presented in the Analysis section. The procedures manual was obtained from EML's website (http://www.wipp.energy.gov/NAMP/EMLLegacy/index.htm)

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2.2.1 ^{90}Sr from the fallout samples

The US EPA has a welldefined assemblage of radioactive materials, which enter the atmosphere thorough different paths, including the detonation of nuclear bombs. Fallout normally contains multiple radionuclides and can remain in the atmosphere for years. Some most commonly detected radionuclides are americium-241, cesium-131, iodine-137, and strontium-90. Through precipitation or natural deposition, fallout can return to the ground surface, or flow to another places (EPA, 2016).

^{90}Sr in the fallout samples has been documented by EML since the mid 1950's to June 1976. The project was implemented globally and collected samples from 52 countries. The analyses were completed by various procedures, beginning with sampling, preparation, and continuing with separation, determination, calculation, and recording. Different sources of samples required specific reagents and different steps of process.

2.2.1.1 Sampling

Fallout samples collected in the environment were obtained from monthly atmospheric deposition. EML utilized three types of devices to collect radioactive debris from the atmosphere, which are pot, ion-exchange and wet/dry deposition collectors. It is noted that the former two types are the primary collectors used by EML. In order to obtain adequate sample mass, collectors are normally located in a public area without surrounding shade, obstructions and interferences. All of the collectors were placed outside on the first day of each month and removed at the end of the month. The ideal location for placing the equipment is on a flat roof of a multiple building.

Pot collectors are made of stainless steel, with an area of 0.076 m^3. Water in the pots is evaporated until approximately 200 ml of the solution is left in the collectors. Afterwards the samples are moved to a 1-L polyethylene bottle. In order to transfer the solids from the pots to the bottles completely, distilled water mixed with the same volume of HNO_3, is added in to the pots. The filled pots are scrubbed, and have a slurry at the bottom that must be transferred to bottles, and the process is repeated multiple times until no solids remain in the pots. This process should be conducted prior to the retrieval of the samples in heavily rainy months, to

assure the samples are not diluted. The bottles with the slurry are labeled with site information, including location, collecting date, and monthly precipitation.

Ion-exchange collectors are used in remote locations lacking facilities and personnel. They are located on roofs in a fashion similar to pot collectors. This type of the collector contains a polyethylene ion-exchange column, a funnel with an area of 0.72 m^2, and a leveling device. Inside the ion-exchange column, 4-cm paper pulp, a 17-cm glass wool plug, and 50 ml Dowex 50W-X12 cation-exchange resin are installed at the bottom of the column. The standard type of filter paper is either Whatman No. 41 filter paper or S&S No. 289 filter paper. The column only works efficiently when it is water-saturated, thus tap water or distilled water is added in constantly when the saturation dropped.

The ion-exchange column is first placed with its small end up, and then connected the short end with a J-shaped tube. Then, the column is inverted again, with its large end up, and screwed to the polyethylene funnel at the top. Afterwards, collectors are left at the observation sites at the first day of the month, and recycled at the end of each month. Dust and solids in the funnel should be fully transferred to the column, so the funnel is rinsed repeatedly and wiped by wet tissue, which is eventually added to the column as well. The

final procedure is to label the columns by writing the information that was presented on the pot collectors.

The third collector, the wet/dry deposition collector, is used to examine the physical and chemical characteristics. The system is a two-bucket design, one wet and the one dry. There is a movable peaked cover, which exposes the wet bucket and covers the dry bucket during the precipitation. The movable cover is controlled by using a set of two conductivity sensors on the roof. When the conductivity sensor detect rainfall, the cover moves to the dry bucket until rainfall ends. For the collected wet samples, there is a heater working beneath the sensors to melt snow or ice accumulated on the sensors, and enhance evaporation of the moisture in the wet bucket.

2.2.1.2 Ashing

Dry ashing is compulsory because strontium is not volatile. In EML's laboratory, dry ashing was implemented in two stages. The first stage heats the sample to 125°C in order to wholly dry the samples. The second stage gradually increases the temperature to 500°C over 8 hours. For larger samples the drying process can be extented to 16 or 24 hours as needed.

After ashing, the samples are weighed and then ground into fine powders, which can pass

a 40 mesh screen. The ash may have to be blended due to the inhomogeneity may happen after the samples are sieved.

2.2.1.3 Separation

Separation is applied differently to the samples collected in the steel pot or ion-exchange column. The collected samples retain strontium, but also calcium, other fission products, and natural radioactive elements; therefore, a separation process is first performed to isolate strontium. In this process, calcium and most of the ions are removed by using nitric acid (HNO_3). For radium, lead and barium, removal is performed by adding barium chromate. For tracers of other fission compounds, it is yttrium hydroxide that removes them from the samples. The concentrations of certain reagents and the standards of apparatus used in EML's experiment are listed in EML Procedures Manual in detail (www.wipp.energy.gov).

Separation is implemented after ashing, grinding and blending. In the ion exchange resin, samples are transferred to a 150-ml platinum crucible, and are heated by placing a heat lamp above the crucible to reach dryness. Afterwards, the samples are moved to a 550 ^{o}C muffle furnace to be ashed. The generated ashes are cooled to room temperature, and mixed with Na_2CO_3. The mixture is put back to the muffle furnace whose temperature is increased to 900

^{o}C until the samples are melted. The liquid samples are moved to a 400-ml beaker containing 300 ml of water inside, and stirred over a hot plate for an hour. Meanwhile, 1 ml of strontium carrier, 1 ml of calcium carrier, and 1 ml of strontium-85 tracer solution are added into the beaker. The stirred samples are filtered into a filter flask, and the residual carbonates on the filter are washed by water. After being washed, carbonates are dissolved on the filter with addition of hot 1:1 HNO_3. Next, the solution is transferred back to the original beaker with the dissolved carbonates and HNO_3. The solution is again heated and stirred until to dryness. The last step is to add 40 ml of water and 25 ml of HNO_3 at a concentration of 90% to dissolve solid particles in the solution. After stirring the solution, 115 ml of fuming HNO_3 is added to complete the whole separation process.

For samples collected in the stainless steel pot, 1 ml of strontium carrier and 1 ml of strontium-85 tracer solution are added into the collector. The mixed samples are firstly placed until evaporated to only 100 ml of the solution remains in the pot. Then, the solution is moved to an 800-ml beaker with water, and left to be dried again. The following procedures, starting from transferring the remaining matter to a 150-ml platinum crucible, are similar to those used to separate the ion exchange resin samples. At the end, the dried samples are

mixed with 60 ml of water and 25 ml of HNO_3 at a concentration of 90%, to dissolve solid matter in the beaker. Then, 195 ml of HNO_3 at the same concentration is added to the beaker.

2.2.1.4 Determination

Determination is the last process before doing calculation. First of all, HNO_3 has to be separated from the solution. By removing the beaker from the heat, the nitrates can settle out in the beaker when the temperature decreases. Furthermore, researchers adopt a 5.5-cm glass fiber filter in a Büchner funnel to suction filter the samples, and let the solution flow into a 1-L filter flask. The remaining filtrate is abandoned and rinsed out. Afterwards, the funnel is replaced with a new one, and inside the beaker some water is added in to dissolve the precipitate, which the suction filter is utilized again. The new solution is transferred to a 150-mL beaker, and placed aside until the liquid evaporates to dryness. After the evaporation, 23 mL of H_2O is added into the beaker. Meanwhile, the temperature is increased to dissolve the precipitates, and the solution is stirred by a magnetic bar. In the next stage, 77 mL of 90% fuming HNO_3 is added into the beaker, and heated constantly. By cooling the well-mixed solution, some $Sr(NO_3)_2$ settles on the bottom of the beaker. The precipitate is then suction filtered into a 250-mL beaker, through a double 2.8-cm glass fiber filter that is assembled

with a Fisher filtrator and Teflon filter. The filtered solution is transferred to a 40-mL centrifuge tube in 1 250-mL beaker, which is placed inside the filter funnel. The rest of the precipitate has to be dissolved in the beaker by using water that does not exceed 20 mL in total.

After all of the processes described in this section, ^{90}Sr is counted and calculated. A scintillation counter is used. The counting data are obtained from the ^{90}Y precipitate, derived from ^{90}Sr.

2.2.1.5 Graphic Presentation

Because EML's database was established on a nation-wide scale, the ideal way to present the comparison is to show the values of different cities on the same graphs. ArcGIS can plot graphs on the map of the U.S., and illustrate fallout values through years. Therefore, we chose ArcMap subset of ArcGIS (ESRI, Redlands, CA) to do the graphing. The version of ArcGIS used in this thesis is ArcGIS 10.2.2. for Desktop.

2.2.2 Data collected from Radiological Health Data (RHD)

Radiological Health Data (RHD) is a monthly publication of a radiological database in

different fields and criteria. The publication first appeared in 1959 from the Department of Health, Education and Welfare, and then conducted by the Division of Radiological Health, Public Health Service. In the monthly reports, the contents were divided into five sections, including the sources of milk, air, water, other data, and a presentation of the radiation levels. In June 1965 *Radiological Health Data* was renamed *Radiological Health Data and Reports.* The new format presents a more accurate compilation, however the analysis of surface raw water was discontinued.

The database of RHD was obtained from various sources, reports, and literature. ^{90}Sr content retrieved from RHD is the ^{90}Sr values detected in pasteurized milk. Milk was chosen to be an important source because ^{90}Sr can concentrated in milk and will accumulate in human bones. As bones are primarily formed by calcium that is obtained significantly from milk in the U.S., researchers believe milk might be a dominant access for the ^{90}Sr ingestion to human bodies.

Sampling of milk was conducted under a monitoring project by Public Health Service Milk Monitoring Network beginning in 1957. The surveillance selected raw milk and pasteurized milk for the analysis. From the beginning of the program, 12 sampling stations

were adopted, including Chicago, Salt Lake City, New York City to collect milk samples. By the summer of 1960 the numble of sampling was expanded to 60 stations around the U.S. These stations were chosen due to some common criteria, for example, milk was produced beyond a certain level and supplied consistently from metropolitan milkshed. In each month, a gallon of milk was collected and sent through air parcel to the Robert A. Taft Sanitary Engineering Center, Cincinnati. For the newly added stations, the samples were sent to Southwestern or Southeastern Radiological Health Facility for the analysis. The complete analysis normally took 4 months to reach final results. The data of pasteurized milk analyzed by an operating agency, PHS, and Southeastern and Southwestern Radiological Health Laboratories, show the individual concentrations of calcium, ^{90}Sr, ^{89}Sr, ^{137}Cs, ^{140}Ba,and ^{131}I. The unit presented in the results is μμc/L (=pCi/L). ^{90}Sr is determined by continuous procedures of radiochemical separations, and measurements of ^{90}Y (yttrium-90), the decay element of ^{90}Sr, by utilizing a low background anticoincidence beta counter.

Eventually, eight stations were picked for analysis in this paper, including Chicago (IL), New York City (NY), Honolulu (HW), Louisville (KY), Pittsburgh (PA), San Francisco (CA), and Chattanooga (TN). These sites are located in different regions of the U.S., and are also

monitored for ^{90}Sr concentrations in EML's database. In this thesis, if both of the Robert A. Taft Sanitary Engineering Center and Southwestern Radiological Health Laboratories reported concentrations of ^{90}Sr at the same sites, the average value between the two laboratories for the analysis was used.

2.3 Gross Beta

The database of gross beta radiation was assembled from the RHD reports. The resources adopted are raw surface water and air.

2.3.1 Gross beta in raw surface water

Water samples collection is a project under the National Water Quality Network, which was established in October 1957. The network first chose 75 sampling stations in the U.S. to do water analysis, and planned to expand the scale to more than 250 stations. The main analysis focused on the potential chemical, physical and biological impacts caused by the various contaminants to the water. By analyzing the dissolved and suspended solids in the raw surface water, gross beta was detected. The samples of raw surface water were collected

from rivers in the U.S., for example, Colorado River, Illinois River, and Mississippi River. These rivers drained areas over many miles; therefore, the samples were not obtained at a single site, but from several cities and lanuses in the watershed. Their radioactivity reports present the concentrations of ^{90}Sr and beta activities in a unit of μμc/L, which equals to pCi/L.

In this thesis, the following seven stations were chosen to present their gross beta values: Yuma and Page (Colorado River), El Paso (Rio Grande River), Massena (St. Lawrence River), Peoria (Illinois River), Chattanooga (Tennessee River), and New Orleans (Mississippi River).

2.3.2 Gross beta in air

Air samples were surveyed by the Public Health Service Radiation Surveillance Network since 1956, with cooperation from the Atomic Energy Commission. In total, 45 stations were chosen to conduct the monitoring program prior to 1961, and after 1961 the number of stations was increased to 60 stations nation-wide. The gross beta measurements were mainly recorded at ground level due to the high sensitivity and ability to quickly observe changes.

The collected air samples were first filtered using a cellulose carbon-loaded dust filter. Those radioactive materials attaching onto the small particles were retained inside the filter. If fission products were gaseous, they would be absorbed to the surface of carbon and collected. The whole measurements took up to five hours to compare with other sources and natural background radiation. Advanced measurements were further conducted in the central laboratory of the Radiation Surveillance Network, Washington.

Besides Public Health Service Radiation Surveillance Network, U.S. Naval Research Laboratory and Air Sampling Network developed by Public Health Service also program researches to analyze surface air. The former department not only records daily values, but also quarterly and monthly data. The latter project was launched in 1953. The analysis includes the quantity of suspended particulate matter, different metals, and gross beta emission. Results of these projects present maximum, minimum and average radioactivity in a unit of $\mu\mu c/m^3$.For the convenience to compare these data with the data from EML, the unit was transformed to pCi/L in this paper.

3. Analysis

The data sources used in this paper were obtained mainly from the governmental reports and websites. Great care was taken to accurately transcribe the data, but stations were monitoring in different years and sometimes by different agencies, there are occasional missing period, especially in the early years of collection. Table. 1 summarizes the data sets used in the database compilation.

Table 1. Information retrieved from EML and RHD

Sources	Target	Collection locations	Period	Measurements adopted
EML	^{90}Sr from fallout	1. 24 countries 2. 55 cities in the U.S. (only 53 cities were used to analyze in this report)	1954-1976 (1957-1966 investigated in this paper)	Three collectors: Pot collector, ion-exchange collector, wet/dry deposit and collector
RHD	Gross beta	1. More than 50 stations in different states	1957-2005 (1957-1966 investigated in this paper)	Three criteria: Raw surface water, air, and milk

In order to present the data clearly and precisely, the results of EML's fallout sample report were divided into 5 geographical areas: Alaska, east regions, central regions, west regions, and the sites located in the Pacific Ocean or North Atlantic Ocean. Missing data in the graphs are assigned a value of -0.1 mCi/km^2. It is not a real value, but a method to

differential missing data from low values or values below detection limits.

3.1 EML results

Strontium values are plotted in columns on the US map with ArcGIS to create a clear comparison among different locations. There are 53 cities included in EML's database, but some are not presented in specific years if their monthly values cannot be detected or are missing.

3.1.1 Alaska

There are six sites in Alaska: Anchorage, Barrow, Cold Bay, Fairbanks, Juneau and Nome recorded in EML's program. From January of 1957 to March 1959, there were no data for strontium in Alaska. However, in April of 1959, Anchorage, Barrow and Cold Bay began to detect ^{90}Sr. Because Juneau is the closest city tested in Alaska to the bomb test sites in Nevada, it was expected to have the highest average ^{90}Sr content and radiation. The concentration of ^{90}Sr was highest in Juneau in May 1959. Fig. 1 shows the fallout concentrations in Alaska in 1959. The figure shows that in Juneau ^{90}Sr level increased to 2.17 mCi/km^2 in May, and decreased to 0.55 mCi/km^2. In June, Cold Bay showed an obvious

increase of ^{90}Sr, which was reported as 1.37 mCi/km^2, the second highest of the year. By comparing other sites to Juneau and Cold Bay, it can be seen that the more northern the sites had the lowest monthly ^{90}Sr concentrations. No data were collected at Fairbanks and Nome.

In 1960, Juneau had the highest average concentration (0.161 mCi/km^2), which was three times higher than other cities in Alaska, and the peak accumulations occurred in the summer. September and December were the only two months that had no ^{90}Sr detections in Juneau. Stations in southern Alaska, including Juneau and Cold Bay, more routinely detected fallout as compared to Alaska's northern regions. Barrow in the upper north had the highest January ^{90}Sr concentration, but the concentrations declined or were undetected in other months. Fairbank and Nome had low concentrations, and some monthly values approaching 0 mCi/km^2. The concentrations observed in Anchorage were significantly lower in summer.

A significant decrease of ^{90}Sr occurred in 1961. The highest fallout value was 0.56 mCi/km^2 in Juneau. Though average ^{90}Sr recorded in Juneau was higher than other sites, the values were still low compared to other years. Thus, the resumption of bomb testing in the U.S. had no contribution to the increase of ^{90}Sr in 1961 in Alaska.

In 1962, a tremendous increase could be observed in the eastern side, including Juneau,

Anchorage, and Fairbanks in June. The ^{90}Sr concentrations increased from 0.33 to 0.88 mCi/km^2, 0.22 to 0.99 mCi/km^2, and 0.52 to 1.39, in Anchorage, Fairbanks, and Juneau, respectively through May to June. It is observed that Juneau still had a second peak in September, and the value remained small through the rest of the year. Northern cities, such as Barrow and Nome, did not show the same trend as found in the cities analyzed above; however, there was still an obvious peak in September.

Fig. 7 to 9 show that from 1963 to 1965 only small amounts of ^{90}Sr, approaching 0 mCi/km^2 were detected in Barrow and Nome. The rest of the cities in Alaska had high values in the summer of 1963. Compare to the previous years, 1963 has the highest recorded concentrations since 1957. For example, in Juneau, Cold Bay and Fairbanks, concentrations reached 5.07, 2.82, and 2.54 mCi/km^2, respectively in July. Anchorage also showed a 4 mCi/km^2 concentration in August. Fig. 6 shows that radioactive ^{90}Sr decreased compared to 1963. Nevertheless, the values were still noticeable and had increased throughout the year. A significant difference is that an intense increase of ^{90}Sr did not occur in the summer. Obvious increasess were observed in May in the southern Alaska and June in Fairbanks. In 1966 (Fig. 10) ^{90}Sr on each site was difficult to detect and stayed at a very low level.

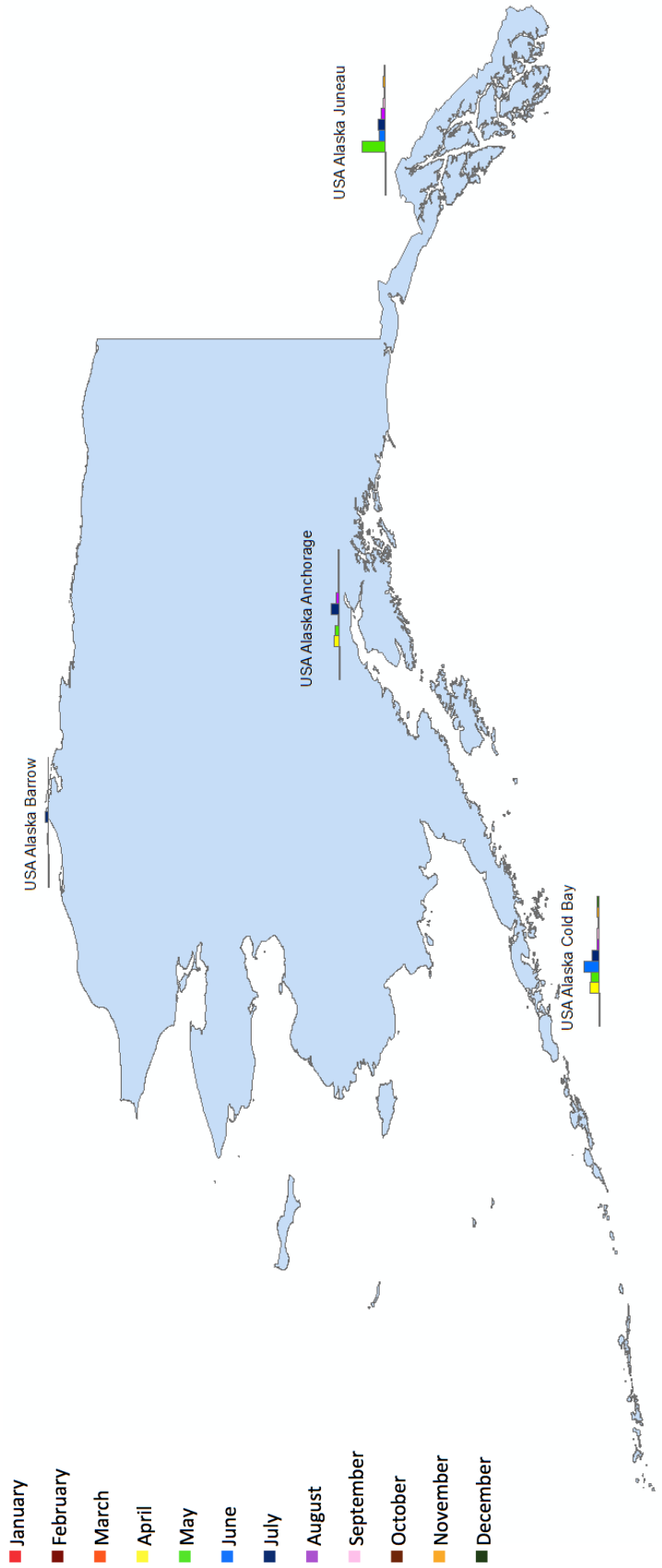


Fig. 4. ^{90}Sr concentrations in Alaska in 1959

Fig. 3 ^{90}Sr concentrations in Alaska in 1959

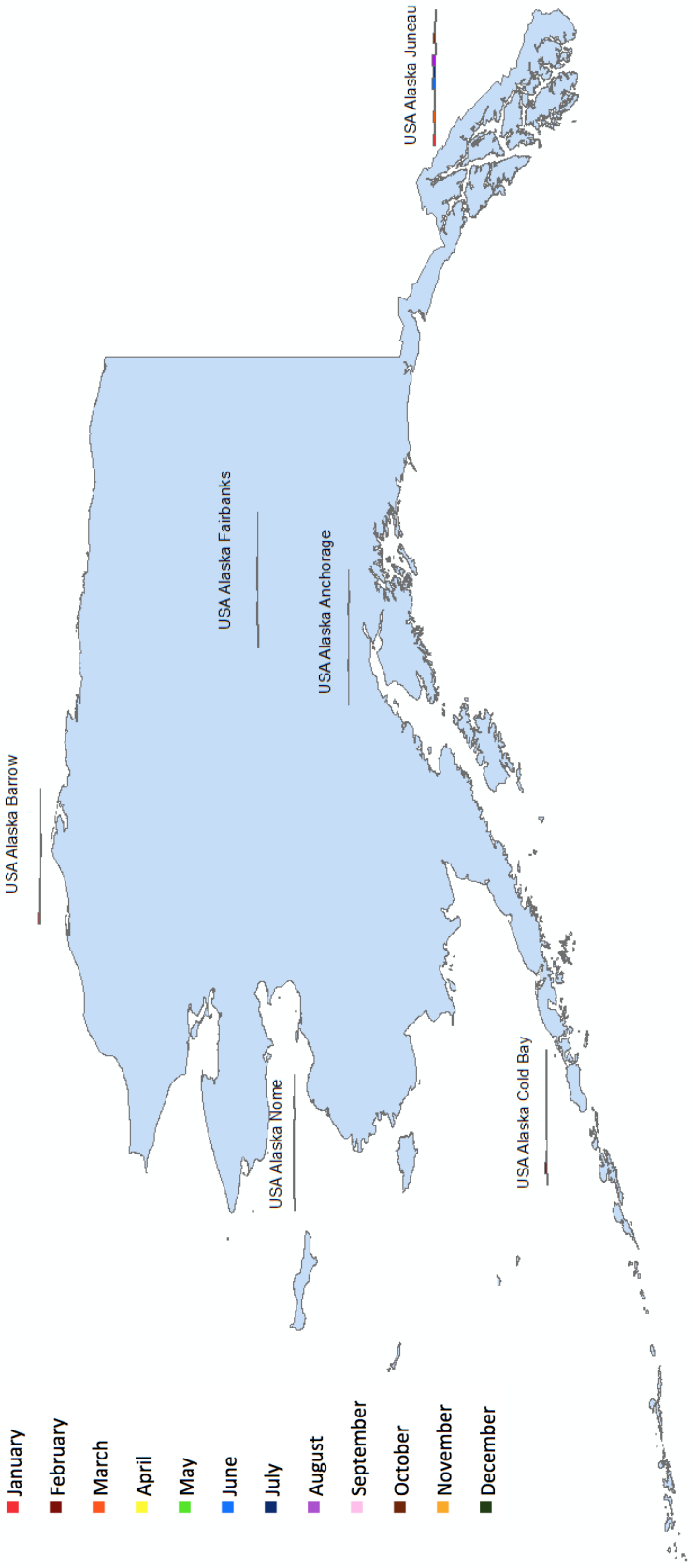


Fig. 5. ^{90}Sr concentrations in Alaska in 1960

Fig. 6. ^{90}Sr concentrations in Alaska in 1961

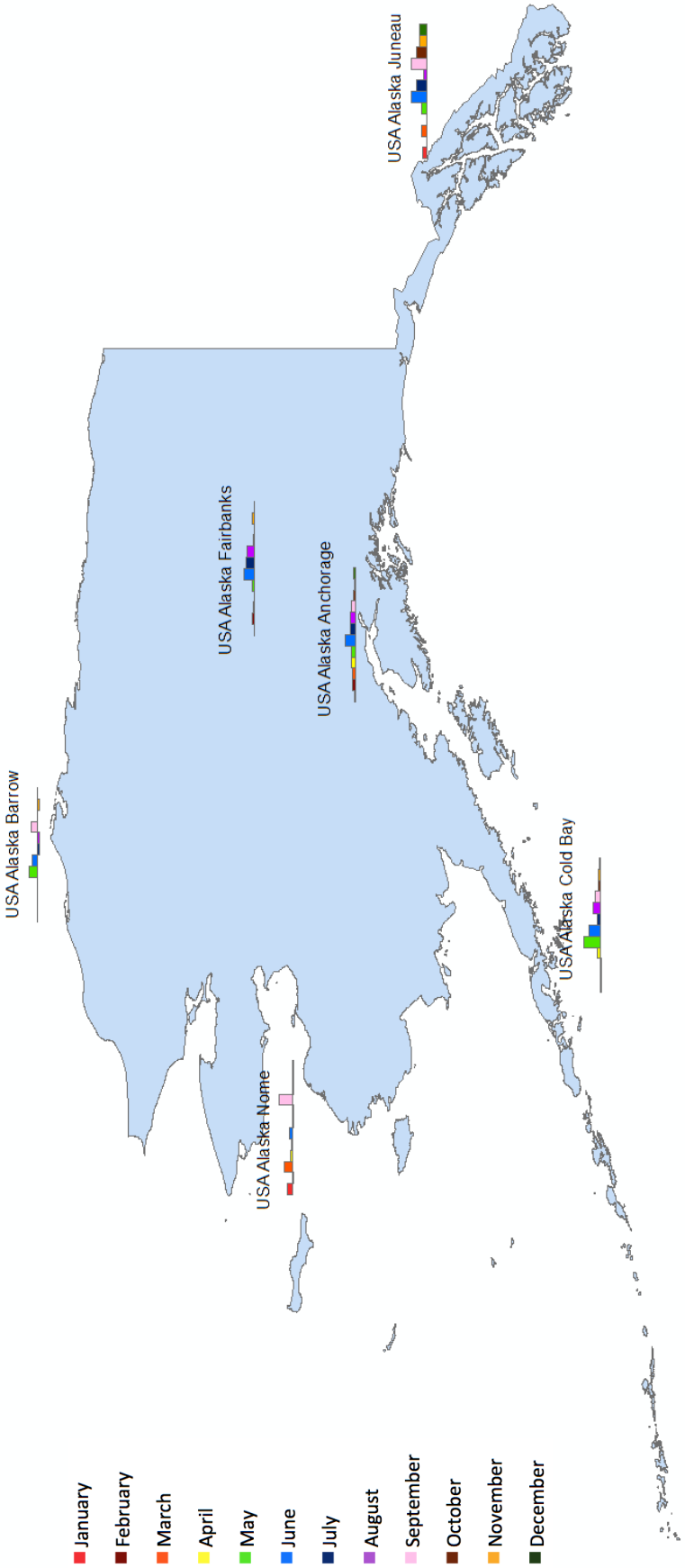


Fig. 7. ^{90}Sr concentrations in Alaska in 1962

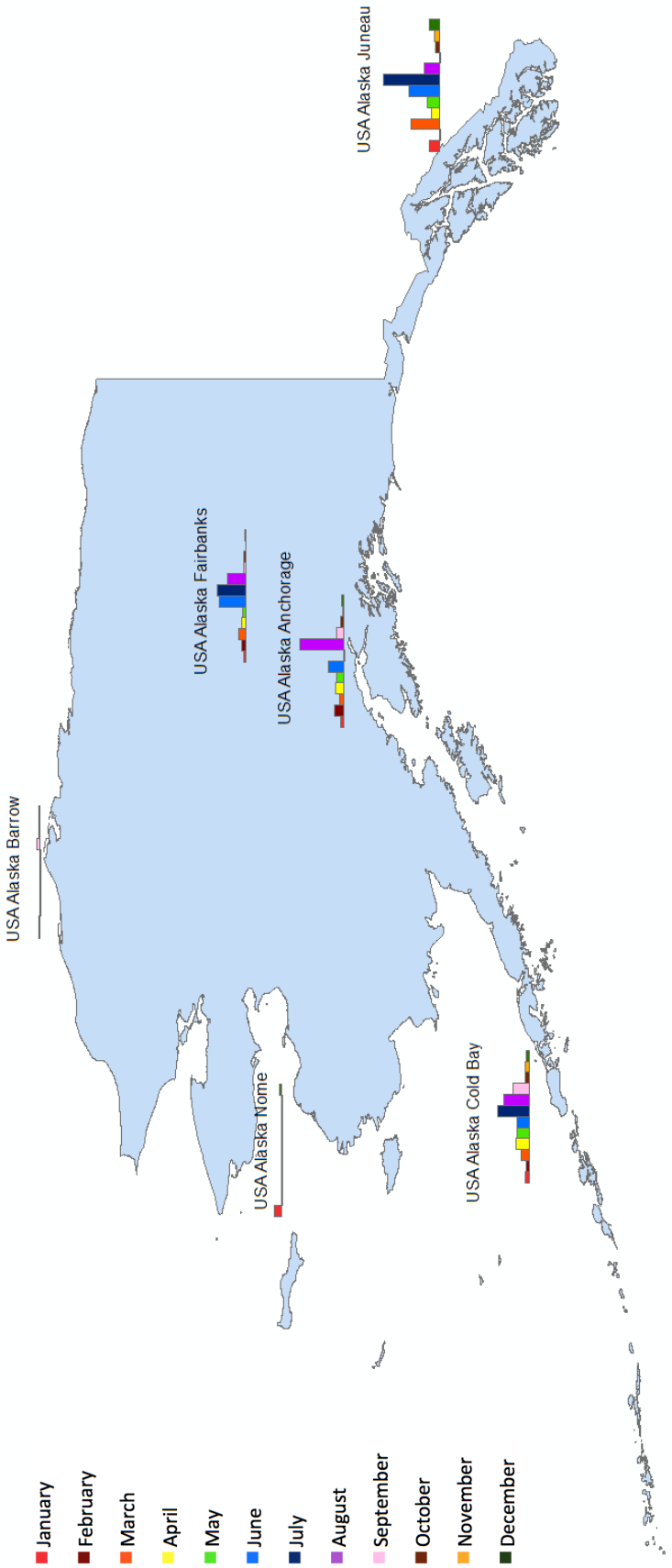


Fig. 8. ^{90}Sr concentrations in Alaska in 1963

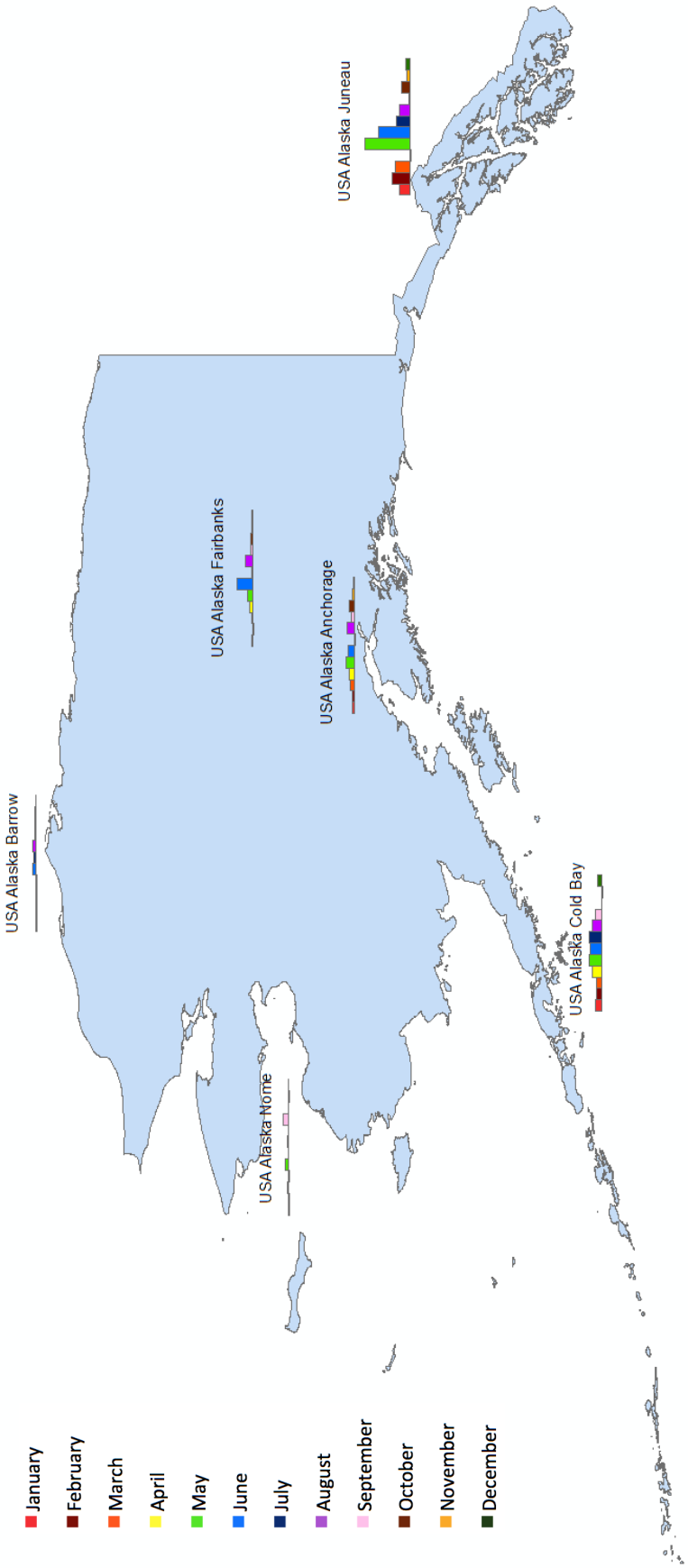


Fig. 9. ^{90}Sr concentrations in Alaska in 1964

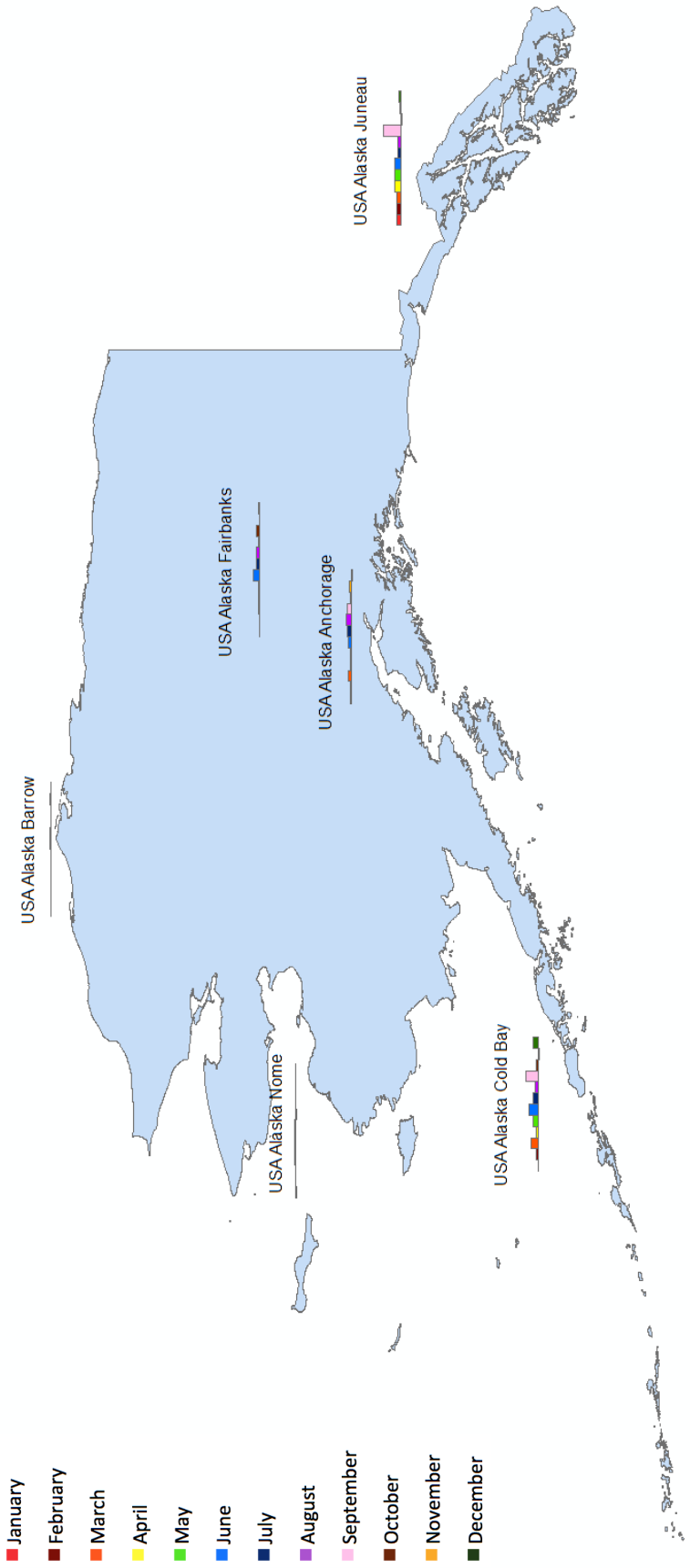


Fig. 10. ^{90}Sr concentrations in Alaska in 1965

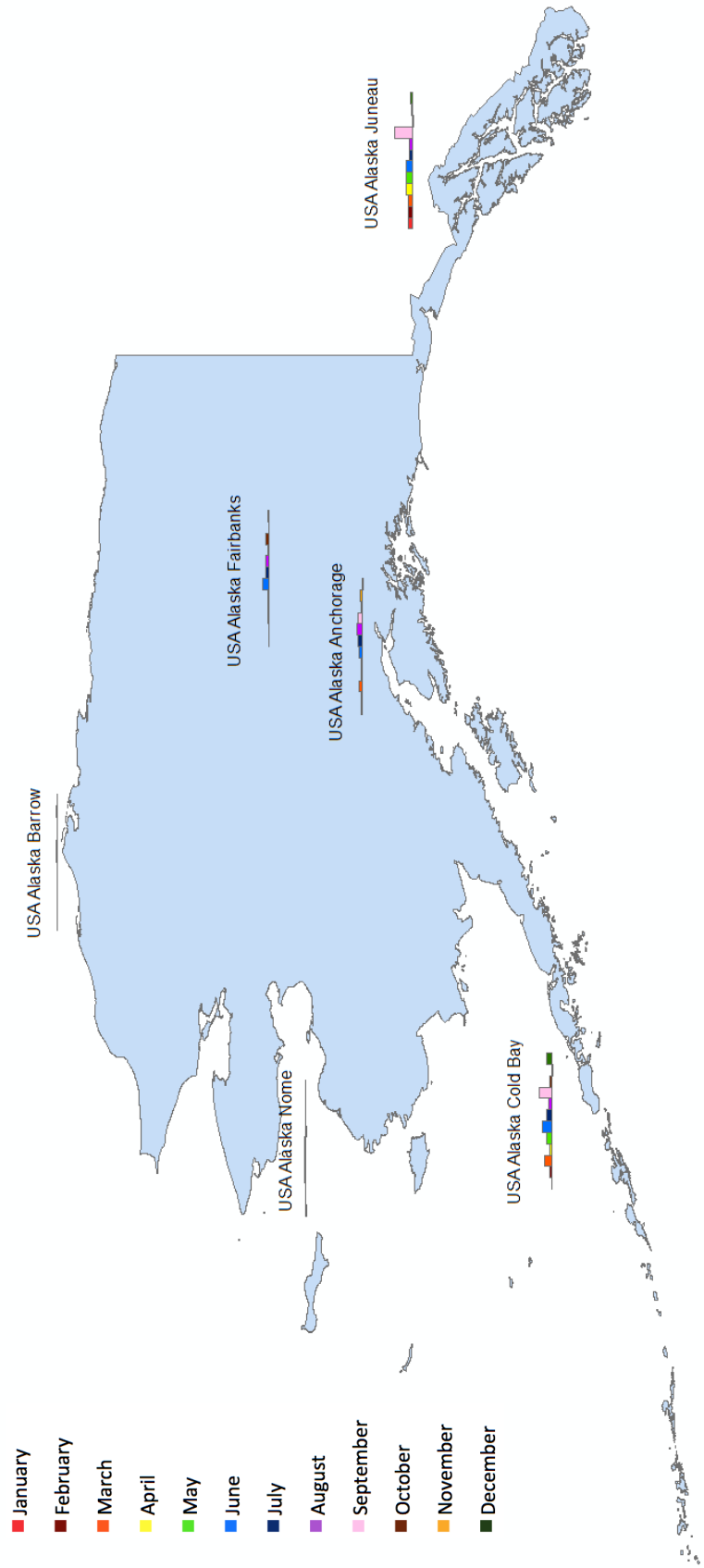


Fig. 11. ^{90}Sr concentrations in Alaska in 1966

3.1.2 West Regions

The west region in this paper includes: Forks (WA), Seattle (WA), Medford (OR), Richmond (CA), Palo Alto (CA), San Francisco (CA), and West Los Angeles (CA), which are located west of the Nevada test sites. Inside the parentheses is the abbreviation of the state where the city is located.

In 1957, only in West Los Angeles were ^{90}Sr concentrations detected throughout the year. Though data of other sites in this year were not presented by EML (Fig. 11), it is not certain that fallout couldn't be detected or were just missing in the database. ^{90}Sr were found between 0.3 mCi/km^2 and 0.4 mCi/km^2 in January, April and July in West Los Angeles. During other months, the values were recorded less than 0.1 mCi/km^2, except in February the value slightly decreased to 0.29 mCi/km^2 from 0.38 mCi/km^2 in January.

In 1958, Richmond and Seattle along with West Los Angeles showed detectable ^{90}Sr data. Fig. 12 indicates that sites located in California, had a large increase of ^{90}Sr in spring. The highest value in Richmond was 1.44 mCi/km^2 detected in March, but concentrations in other months were mostly less than 1 mCi/km^2. West Los Angeles showed a gradual increase in spring, followed by a rapid decrease, and remained steadily low until the end of the year.

Fig. 13 and Fig. 14 show the ^{90}Sr in 1959 and 1960, respectively. Both of the figures show that the concentration was higher in spring than in winter. On the other hand, summer had the lowest concentration over the whole year. Though Richmond, San Francisco, and Palo Alto are close to each other, the data demonstrate a significant difference with Richmond having highest ^{90}Sr content among them. While the maximum value could reach up to 1.03 mCi/km^2 in Richmond, the highest value recorded in San Francisco was only 0.19 mCi/km^2, which is approximately 20% of 1.03 mCi/km^2. Comparing Richmond and West Los Angeles, a similar trend of the change of concentration was found. Palo Alto at the same time did not show any results due to no detection. Fig. 14 presents a wide detection of ^{90}Sr along the west coast. The sites in Oregon and Washington states had higher ^{90}Sr values than those in California. A trend in ^{90}Sr can be observed with northern locations having higher concentrations and summer observations being generally lower than in other seasons. In March 1960, strontium value in Forks is recorded 0.45 mCi/km^2, which was the highest of the year. However, between the summer and spring, it is noted that Seattle and Medford had their highest values of 0.3 mCi/km^2 and 0.27 mCi/km^2, respectively. Fig. 15 shows a definite trend of decreasing ^{90}Sr in the west regions in 1961 with concentrations decreasing to less

than 0.3 mCi/km^2. Also, the northern stations, including Washington and Oregon exhibiting higher values than the southern stations. It indicates that fallout had an intendancy to flow northerly.

Since 1962, the inventory of ^{90}Sr had increased in most of the cities, except in Palo Alto, which is ambiguous because of missing data. Fig. 16 indicates that in Richmond, San Francisco, and West Los Angeles experienced as peak in February, which was more than 4 times as high as in January. Cities in Washington showed a peak in March and April. After these large peaks, concentrations began to decrease dramatically. However, another significant peak was detected at 2.47 mCi/km^2 in Forks in November, and at 2.03 mCi/km^2 in San Francisco in July.

In 1963 ^{90}Sr increases were clearly observed in California (Fig. 17). The concentrations gradually increased from January and reached their peaks in March in West Los Angeles or April in other cities of California. A dramatic decrease from over 2.5 mCi/km^2 to less than 1.5 mCi/km^2 was observed after April. Medford had a peak in May at 1.9 mCi/km^2, followed by a decrease, and then a slight increase in November. Washington State in 1963 had a remarkably high inventory. Forks, which is between the two Washington cities had an

average concentration of 1.73 mCi/km^2, and concentrations over 2 mCi/km^2 for 5 months of the year. Seattle had a similar pattern as Forks, but the values were not as high as those in Forks.

Fig. 18-Fig. 20 show ^{90}Sr concentrations from 1964 to 1966, respectively. All of the figures consistently show higher values in the north than the south. In 1964, none of the values at California sites exceeded 0.6 mCi/km^2. However, in Washington State, it was common to detect values at more than 1 mCi/km^2, and the highest concentration at 2.69 mCi/km^2 in Forks. Fig. 18 shows significant peaks at 1.855 mCi/km^2 and 1.04 mCi/km^2 in the beginning of the summer in Seattle and Medford, respectively. Fig. 19 shows another year of the scarce of ^{90}Sr from fallout samples. In Washington State, an increase was seen in February, while in California State, an increase occurred in April.

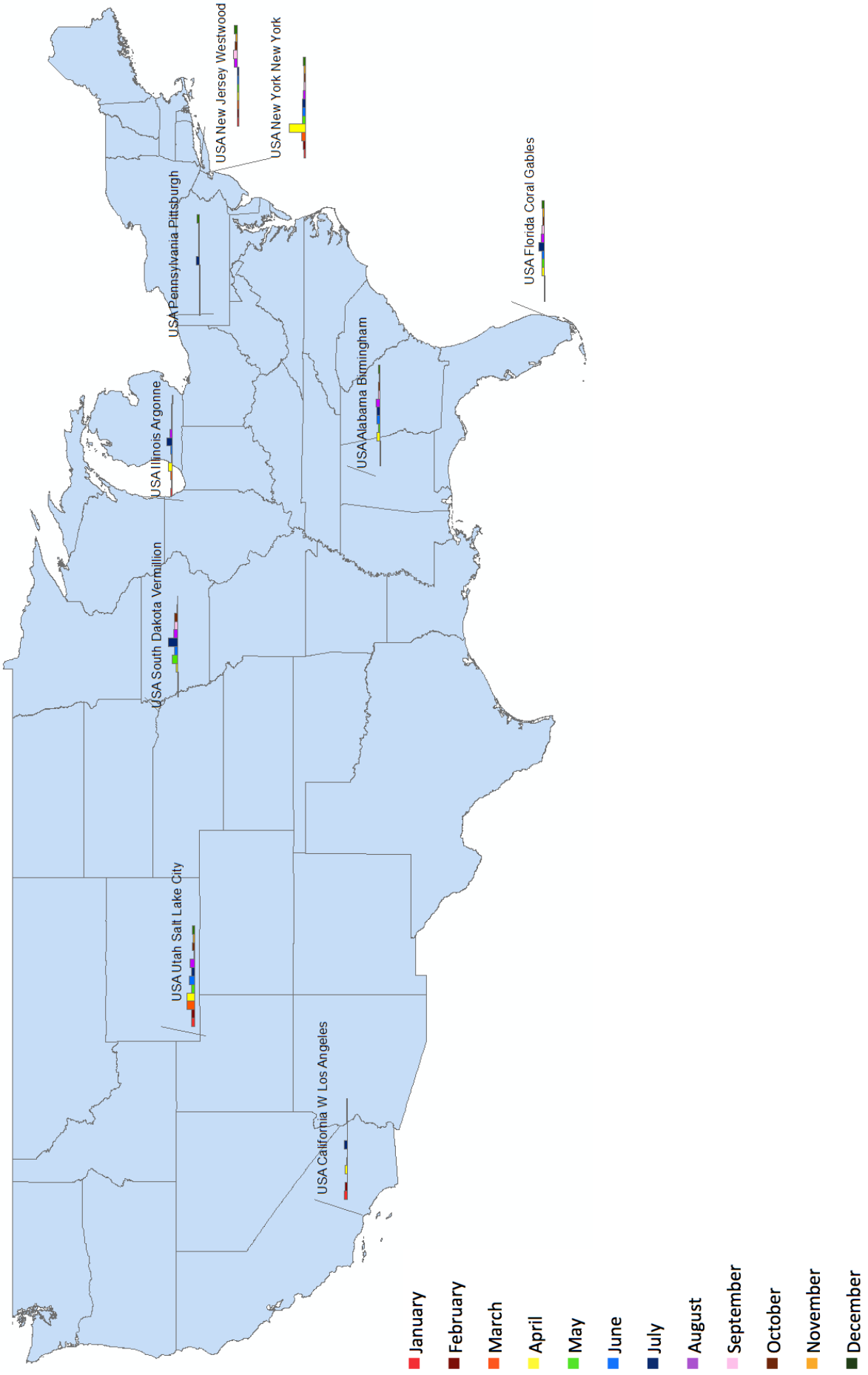


Fig. 12. ^{90}Sr concentrations in the U.S. in 1957

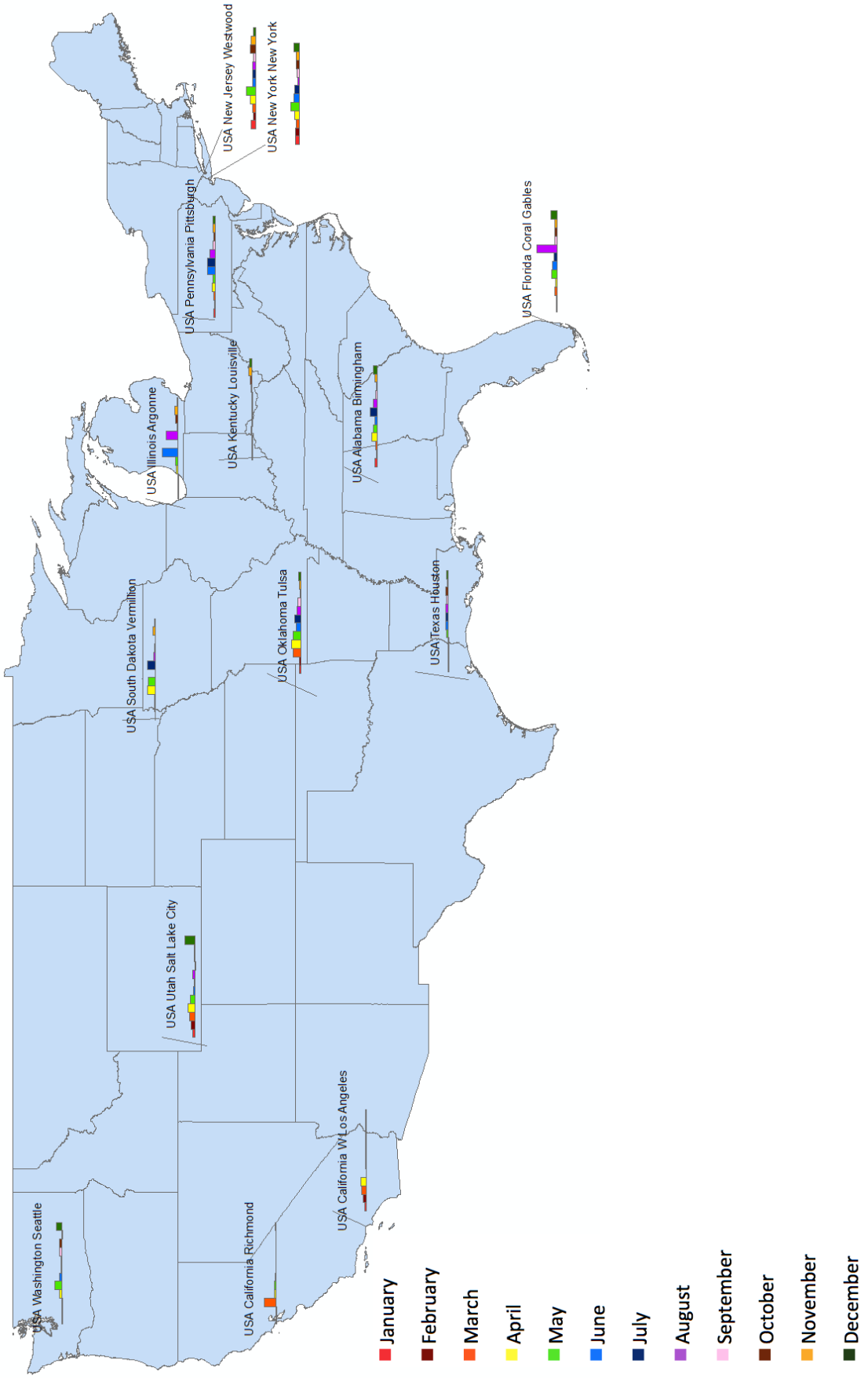


Fig. 13. ^{90}Sr concentrations in the U.S. in 1958

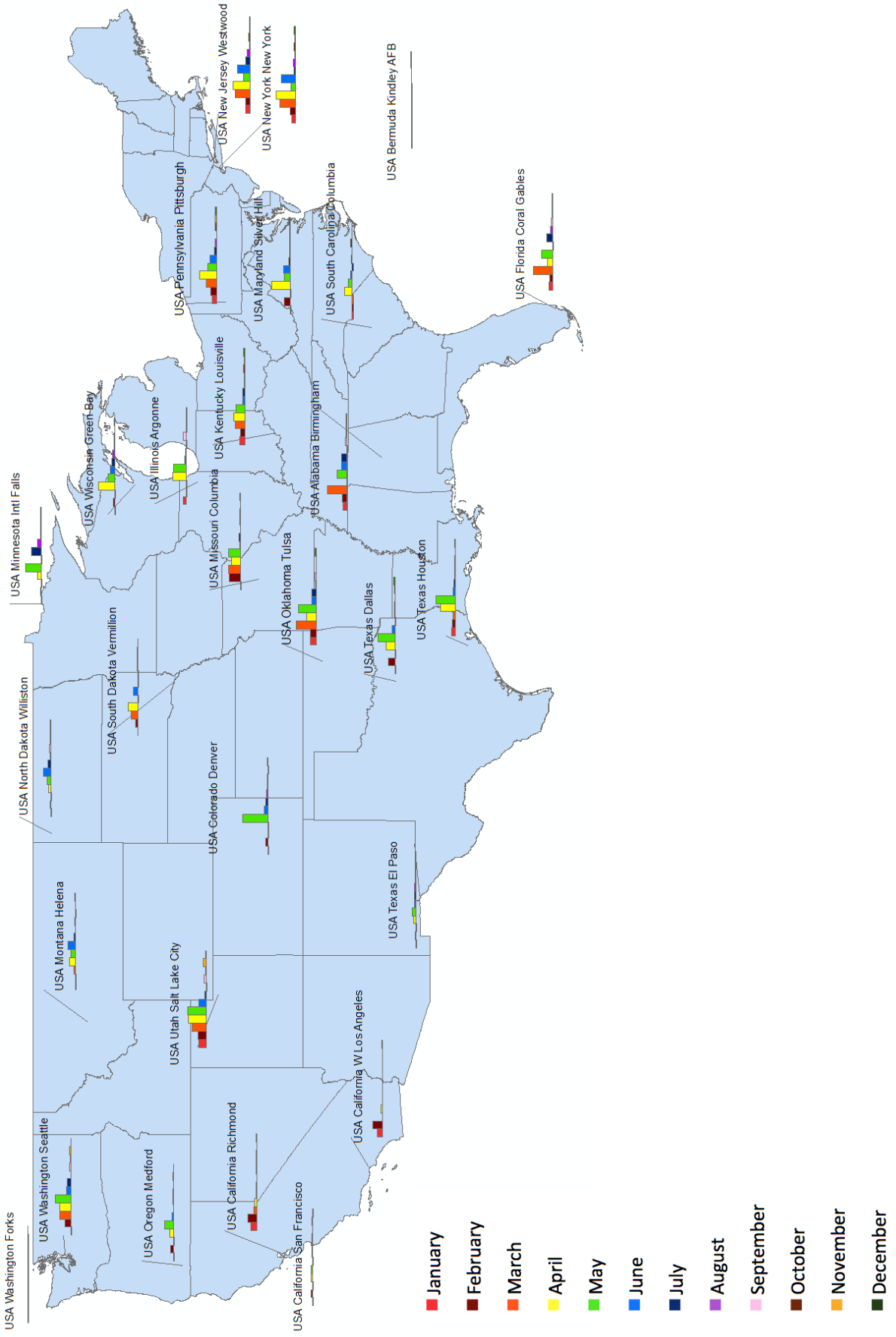


Fig. 14. ^{90}Sr concentrations in the U.S. in 1959

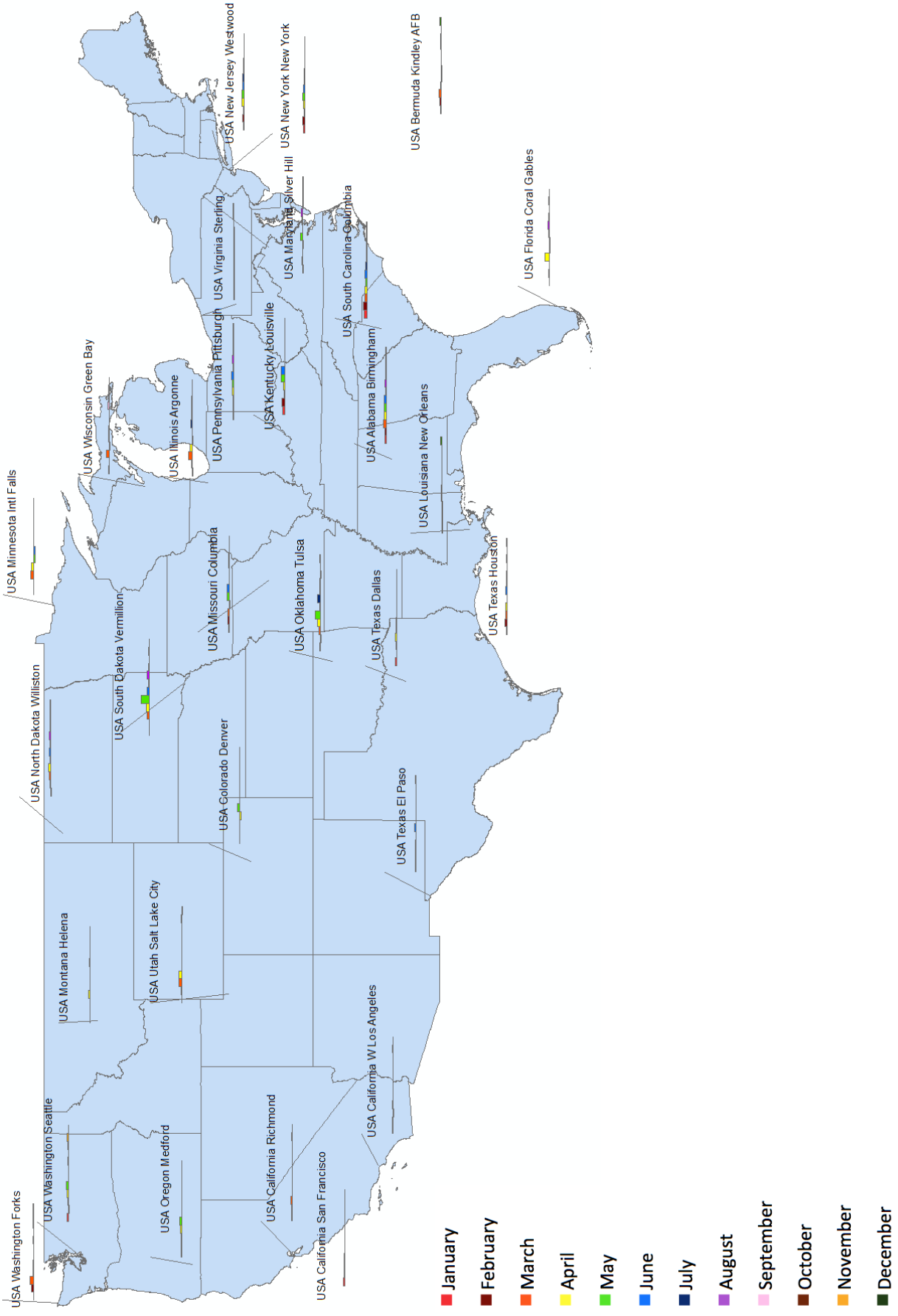


Fig. 15. ^{90}Sr concentrations in the U.S. in 1960

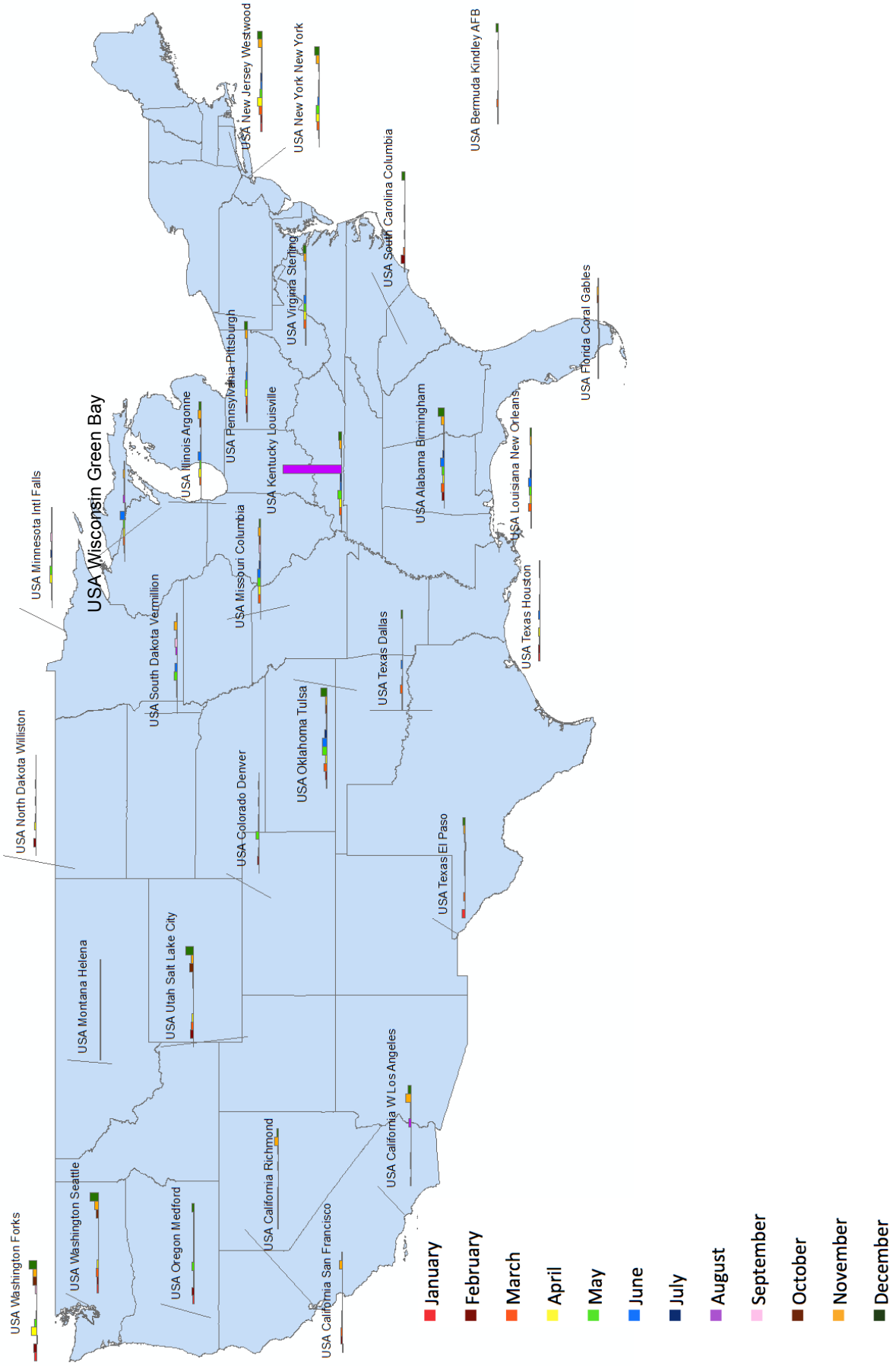


Fig. 16. ^{90}Sr concentrations in the U.S. in 1961

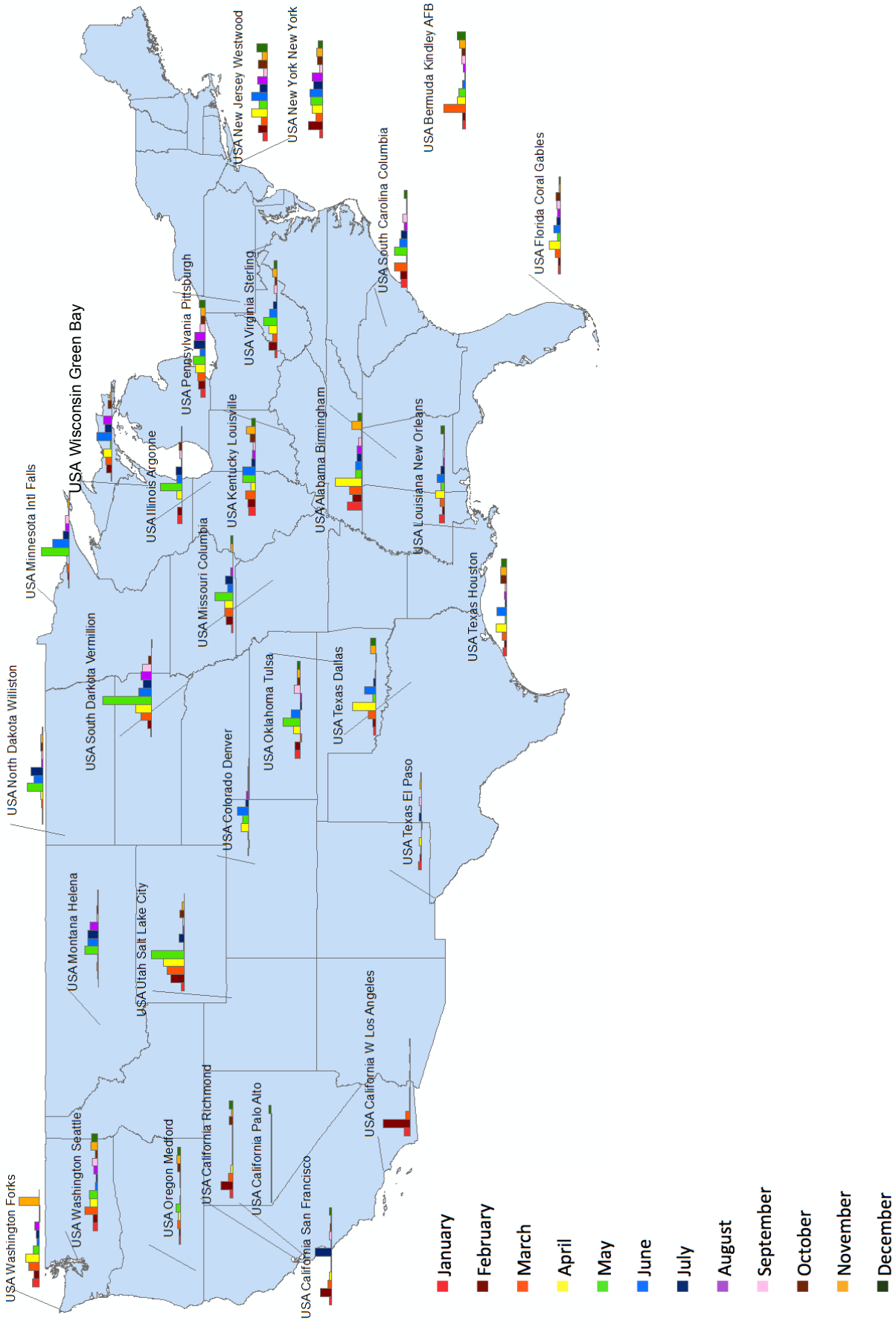


Fig. 17. ^{90}Sr concentrations in the U.S. in 1962

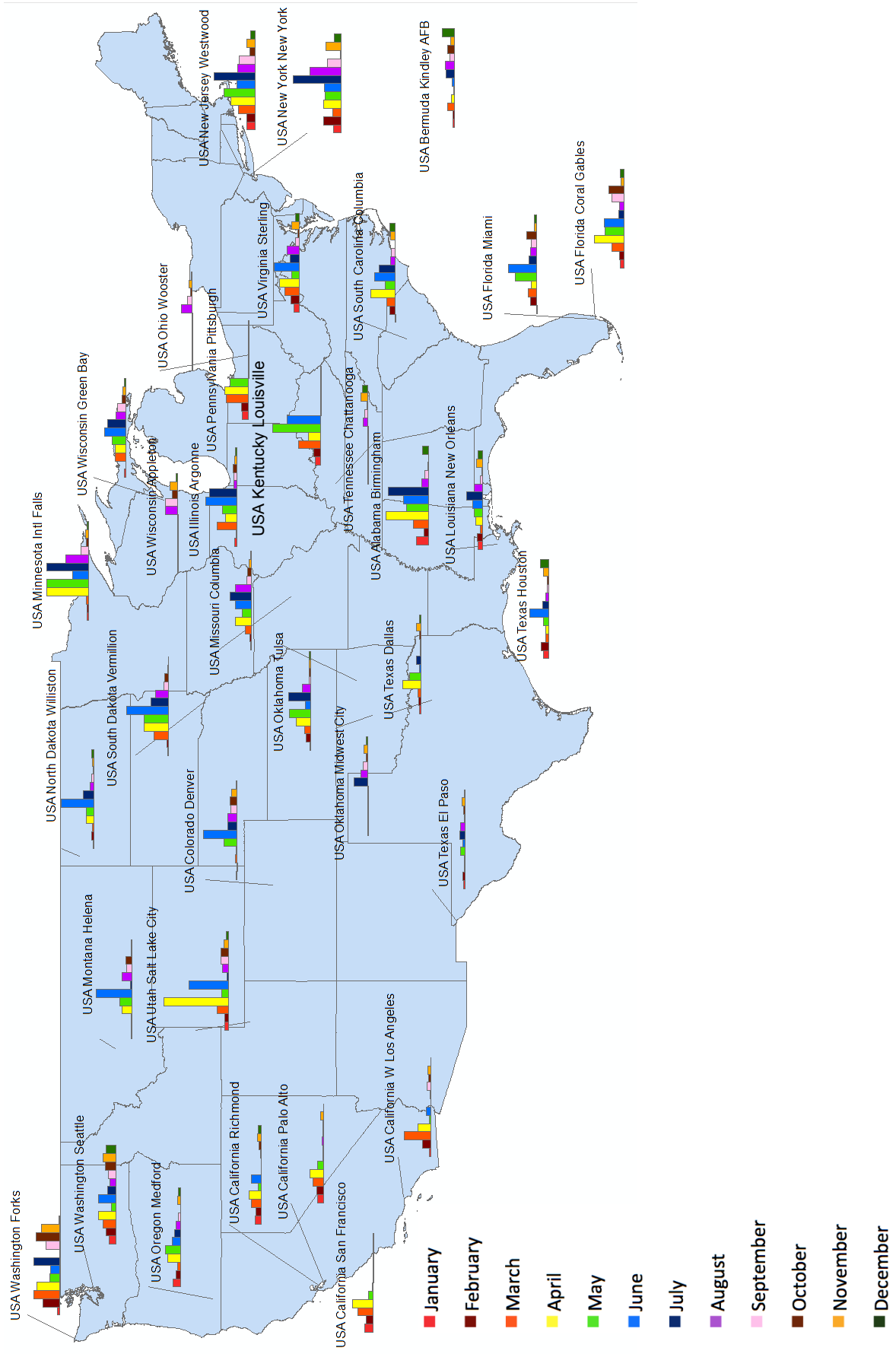


Fig. 18. ^{90}Sr concentrations in the U.S. in 1963

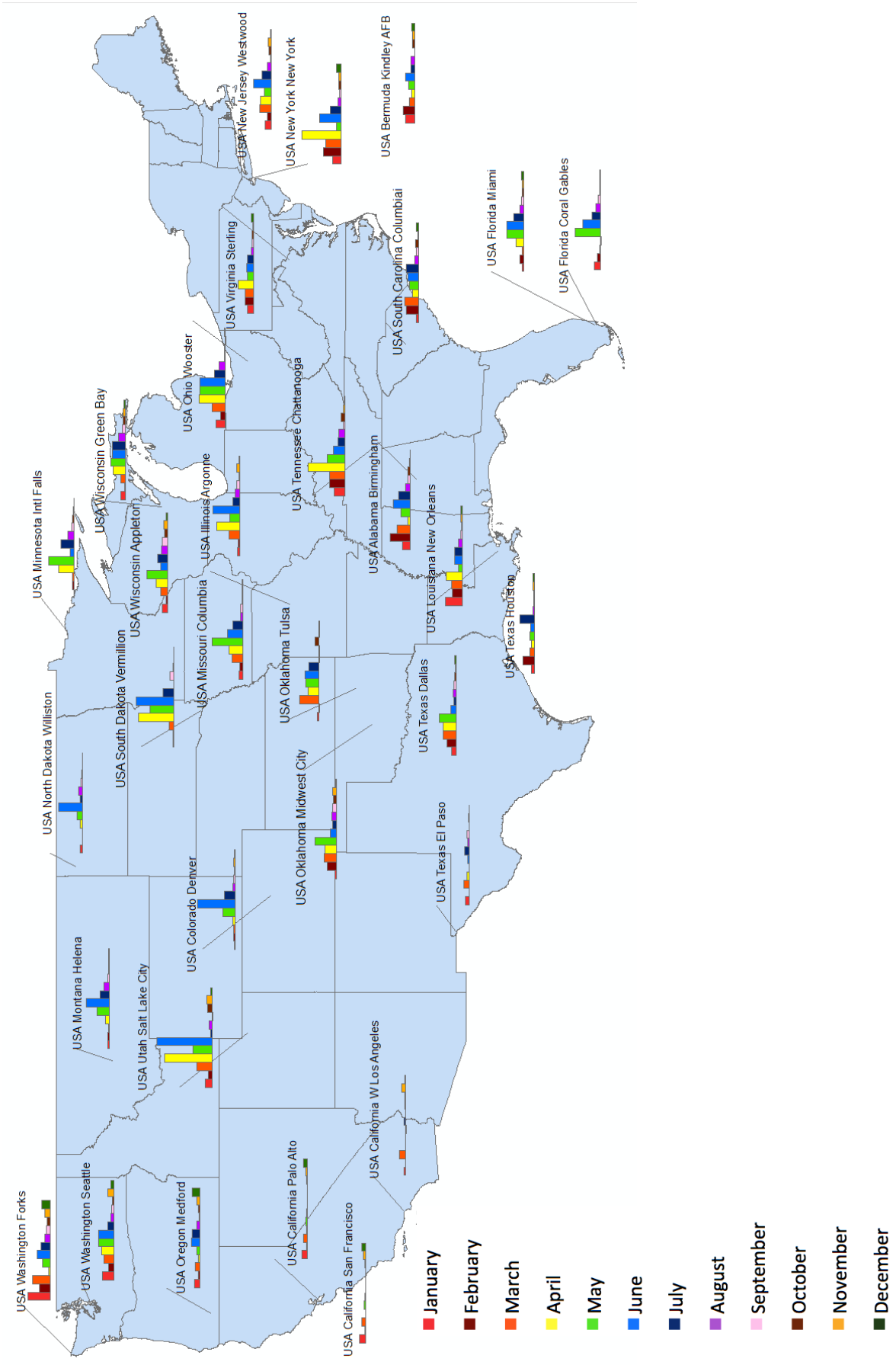


Fig. 19. ^{90}Sr concentrations in the U.S. in 1964

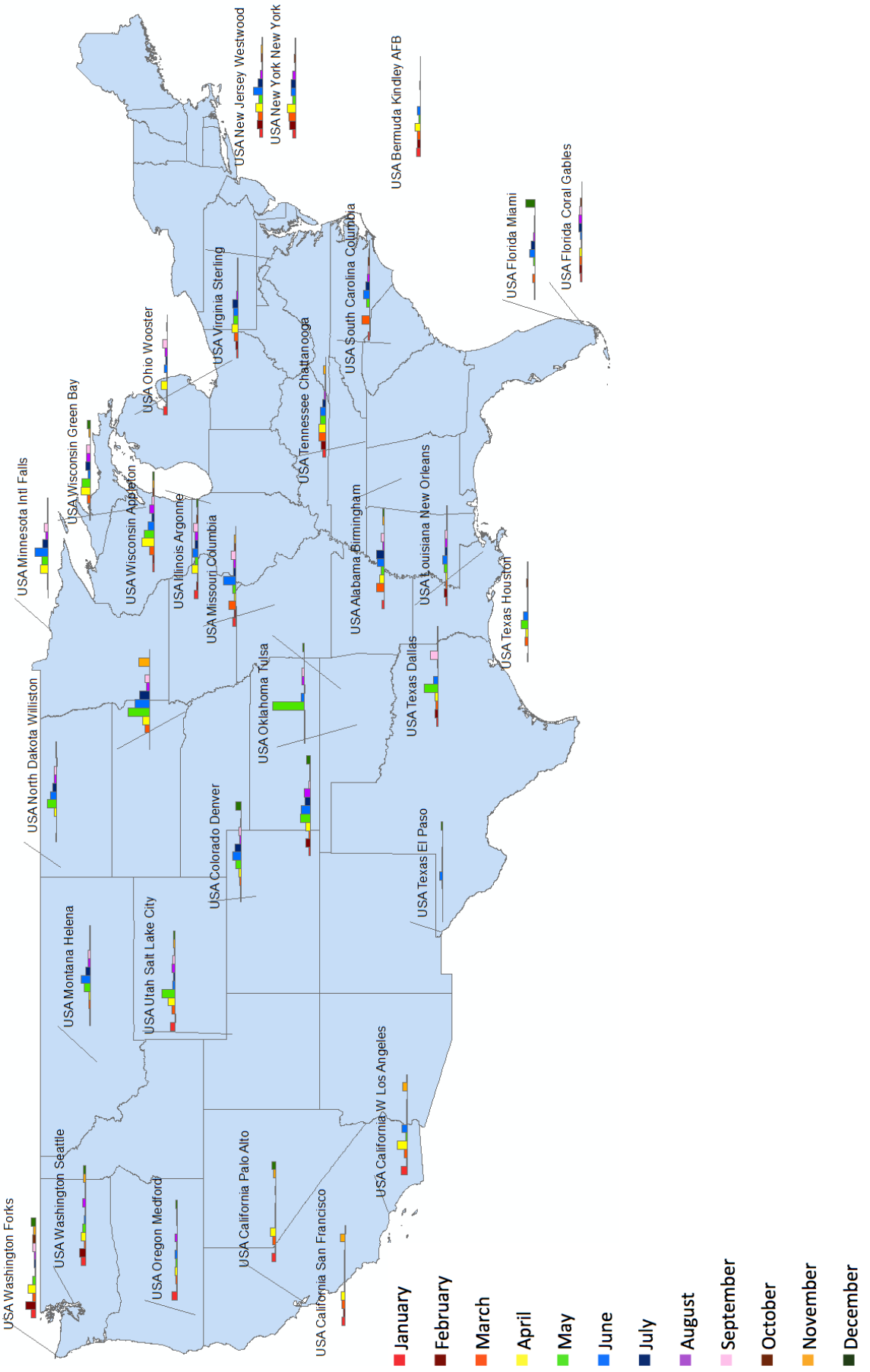


Fig. 20. ^{90}Sr concentrations in the U.S. in 1965

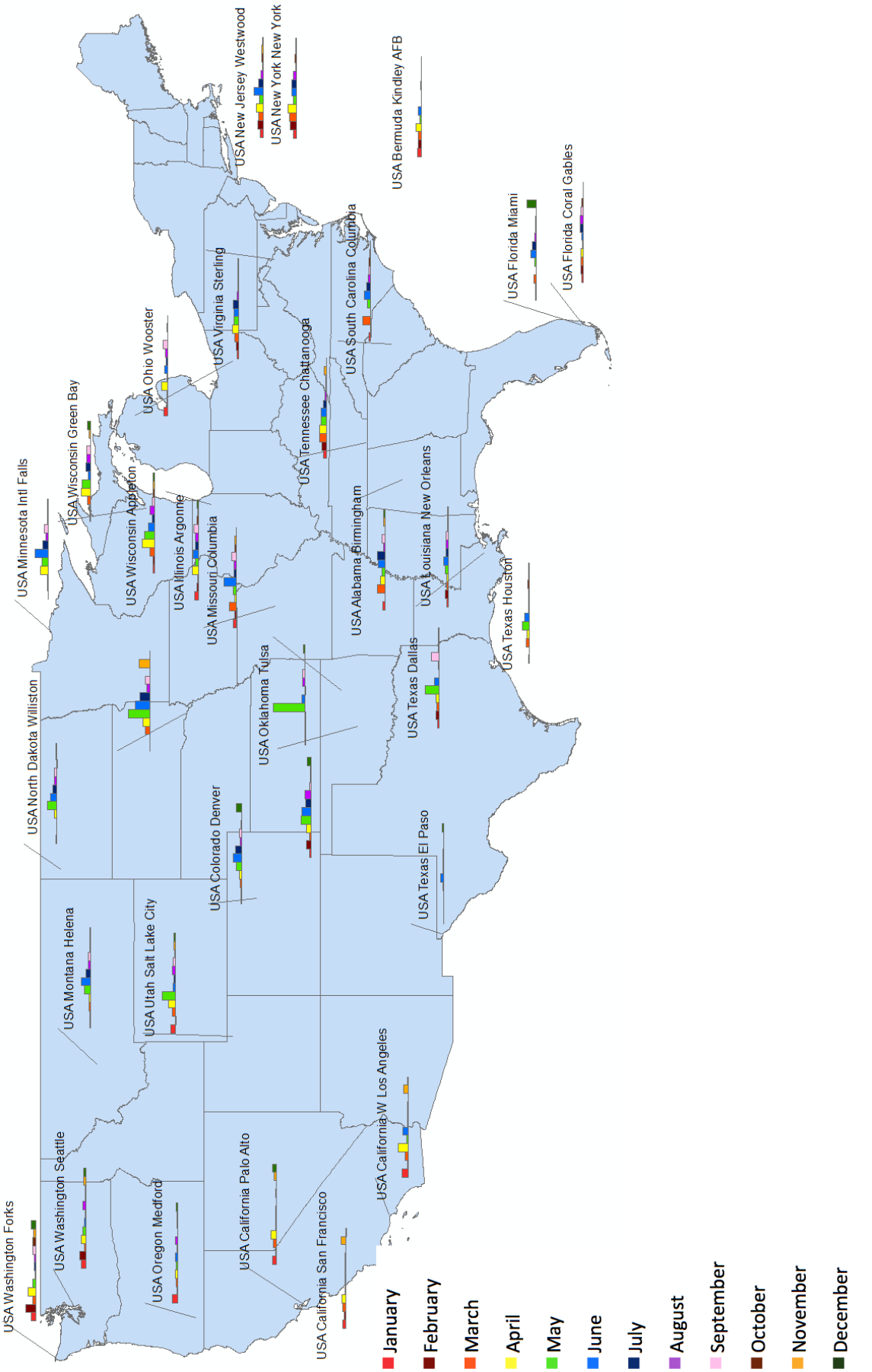


Fig. 21. ^{90}Sr concentrations in the U.S. in 1966

3.1.3 Central regions

The analysis for the central included 13 cities or sites for analysis: Helena (MT), Salt Lake City (UT), Williston (ND), Denver (CO), El Paso (TX), Vermillion (SD), Tulsa (OK), Midwest City (OK), Dallas (TX), Houston (TX), International Falls (MN), and Columbia (MO).

Fig. 11 and Fig. 12 indicate an obvious observation of ^{90}Sr values in Salk Lake City and Vermillion. The former one is the closest city to the Nevada Test Site in the central regions; therefore, the concentrations may be easily affected by the bomb tests. Fig. 11 shows that high values of ^{90}Sr were detected in March and April at 0.92 and 0.89 mCi/km^2, respectively; while in Vermillion a peak was found in July of 1.08 mCi/km^2. In 1958 (Fig. 12), sites such as Tulsa and Houston began to observe consistent radioactivity. Salt Lake City showed a monthly increase of approximately 0.15 mCi/km^2 from January to April, followed by a decrease until the end of November. In December, the greatest value of 1.19 mCi/km^2 within the whole central regions was observed in Salt Lake City. It is noted that in other cities of the central regions, no high values were detected in December.

Fig. 13 indicates a drastic increase of ^{90}Sr in various cities in the central regions in 1959.

Cities such as Salt Lake City, Houston, Denver, Columbia, International Falls, Tulsa, and Dallas showed a noted peak in May, 1959. Overall, ^{90}Sr values were relatively high in spring, and small after June, except at the International Falls site where a second peak of 1.13 mCi/km^2 was seen in July. In May 1959, Denver had the highest value of 3.1 mCi/km^2 of the year; however, in other months, ^{90}Sr values were undetectable or too low to be presented. Besides Denver, most of the cities observed high concentrations of ^{90}Sr in May. When cities were located more eastern or closer to the Nevada Test Site, monthly ^{90}Sr values could be observed more consistently and well above zero.

Fig. 14 indicates a decrease of average ^{90}Sr concentrations in 1960 compared to 1959. 1960 is the first year that ^{90}Sr could be detected in New Orleans, though its highest value of the year was only 0.13 mCi/km^2 in December. As shown in Fig. 15, in 1961 ^{90}Sr decreased to the lowest level since 1957. The highest value in the central regions was recorded in December in Salt Lake City at 0.89 mCi/km^2. Though the values were small, it could still be seen that an increase occurred in December in Salt Lake City and Tulsa, like the trend noted in Seattle and Forks in Washington State, and Medford in Oregon State.

Fig. 16 shows a drastic increase of ^{90}Sr in the central regions, especially in May when a

peak was found in many cities. The higher concentrations detected in May were mostly observed in the sites more northern, for example Williston, Vermillion, International Falls, etc. Salt Lake City has a stable increase since January and reaches a peak in May at 4.11 pCi/km^2, which was the highest concentration detected at this site since 1957. Vermillion is another city that had an obvious increase in strontium. Fig. 16 shows a similar increasing trend such as the trend observed in Salt Lake City, but the peak value of 5.98 mCi/km^2 was observed in May. For those cities in Texas, for example, Dallas and Houston, the highest values of 2.86 mCi/km^2 and 1.26 mCi/km^2, respectively, were observed in April.

In 1963, peak values were recorded in June at many sites (Fig. 17). Salt Lake City showed two high concentrations in April and June at 7.82 mCi/km^2 and 4.78 mCi/km^2, respectively. Their high ^{90}Sr concentrations were detected at International Falls in April, May, and July, at the concentration of 5.1 mCi/km^2, 5.04 mCi/km^2, and 5.09 mCi/km^2, respectively. In Taxes or Oklahoma, however, the peak values did not always happen in summer.

Fig. 18 shows the result of the 1964 monitoring. In Salt Lake City, the concentrations in April and June were at least two times higher than of other months. Concentrations in April and June were 5.7 mCi/km^2 and 6.58 mCi/km^2, respectively. Other sites north to Oklahoma

showed a peak in June, except International Falls, whose highest value was observed in May. Midwest City, Tulsa, Houston and Dallas had most of the ^{90}Sr detected in spring. After July, the concentrations remained lower than 0.6 mCi/km^2.

1965 is another changing point that demonstrates the decrease of ^{90}Sr in the U.S. Though Fig. 19 shows that values reported in May are still significant, especially in Tulsa, the overall concentrations descended compared to the previous years. In 1966, none of the sites in the central regions detected ^{90}Sr concentrations higher than 0.6 mCi/km^2 (Fig. 20).

3.1.4 East regions

The east regions included: New Orleans (LA), Green Bay (WI), Appleton (WI), Argonne (IL), Louisville (KY), Chattanooga (TN), Birmingham (AL), Wooster (OH), Columbia (SC), Miami (FL), Coral Gables (FL), Pittsburgh (PA), Sterling (VA), Silver Hill (MD), New York (NY), Westwood (NY), and Kindley AFB (Bermuda). We included Kindley AFB in the east regions because it is easier to show on the same maps.

Though the east regions are located farther from the Nevada Test Site or Johnston Island, where most of the bomb tests occurred, ^{90}Sr concentrations were more obvious and consistently detectable compared to the central and the west sides.

Since 1957 (Fig. 11), the Argonne, Pittsburgh, Birmingham, Coral Gables, New York and Westwood sites routinely detected ^{90}Sr. Since these sites were evenly distributed in the east regions, it was an evidence to prove that ^{90}Sr from the fallout could be transported to various states in different directions. Because Westwood and New York were close to each other, they showed a similar pattern of ^{90}Sr concentrations, increasing gradually from January to May, and decreasing until the end of the year. In 1958 (Fig. 12), a similar increase detected in Salt Lake City was also observed in Tulsa. The Argonne site showed two high peaks in June and August, without subsequent large changes as observed at other sites following large peaks.

In 1959 (Fig. 13), sites in the east regions showed a trend of relatively high ^{90}Sr concentrations before the summer, as observed in the west and the central regions. Peak values were commonly observed in March or April. Take Birmingham for example. The city showed a high concentration of 2.38 mCi/km^2 in March. In 1960 (Fig. 14) and 1961 (Fig. 15), low concentrations of ^{90}Sr were recorded all over the U.S., except in Louisville where a significant peak value of 7 mCi/km^2 was observed in August 1961.

Fig. 16 indicates an obvious increase of ^{90}Sr in 1962. Cities in Pennsylvania and New

York observed concentrations were mostly detectable and above 0.5 mCi/km^2 over the entire year. The result of 1963 (Fig. 17) shows that ^{90}Sr concentrations increased more rapidly compared to 1962. Birmingham in the same year had high concentrations from April to July, while two peaks were observed in April and July at 5.19 mCi/km^2 and 4.94 mCi/km^2, respectively. The sites in cities around Birmingham, such as New Orleans and Chattanooga did not report constant detection, especially Chattanooga that did not show any data until August. The cities located north of Chattanooga, including Louisville and Pittsburgh had no detections after July, while the peaks were found in May at 5.8 mCi/km^2 and in April at 2.885 mCi/km^2, respectively. New York City and Westwood showed a similar trend as in the past years, having their highest concentrations in July at 5.73 mCi/km^2 and 4.9 mCi/km^2, respectively.

In 1964 (Fig. 18), ^{90}Sr concentrations slightly decreased; however there were still some remarkable peaks observed in Chattanooga and New York City in April at 4.4 mCi/km^2 and 4.74 mCi/km^2, respectively. The overall trend indicated that values dropped to lower than 1 mCi/km^2 after August. Between January and March, although the sites in Wisconsin State did not observe high concentrations, sites in other states mostly detected ^{90}Sr in this period. Sites

in Miami and Coral Gables had increasing concentrations beginning in April, with the concentrations rapidly increasing from 0.96 mCi/km^2 to 2.04 mCi/km^2, and from an undetectable level to 3.04 mCi/km^2. No strong linkin ^{90}Sr concentrations among different states were found in 1964.

^{90}Sr concentrations in 1965 (Fig. 19) and 1966 (Fig. 20) decreased in that the observations were rarely greater than 1 mCi/km^2 at all sites. In 1965, a slight increase was observed from January until April or June, followed by a decrease throughout the year.

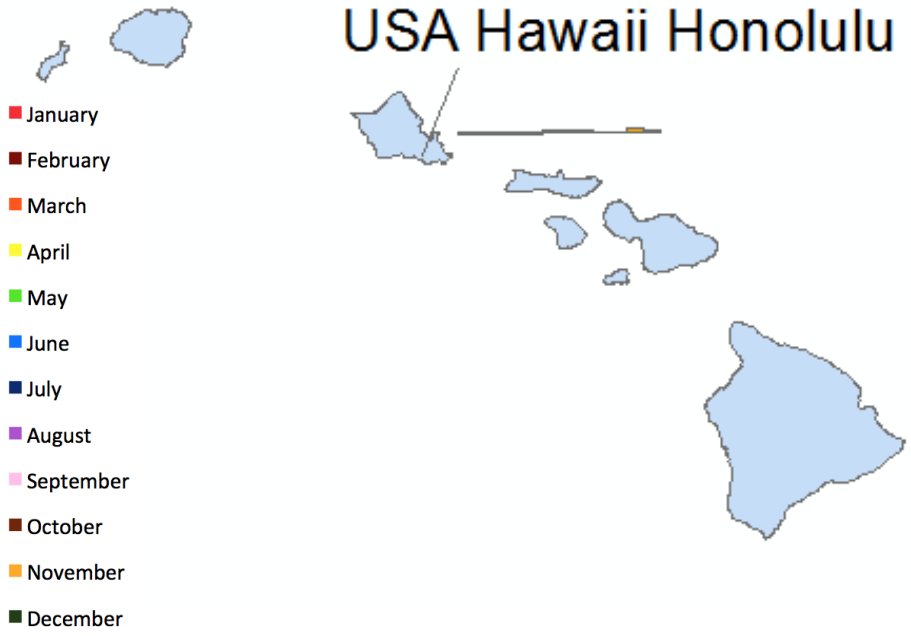
3.1.5 The Pacific Ocean and the North Atlantic Ocean

Pacific Ocean and the North Atlantic Ocean comprise the islands out the U.S., including: San Juan (PR), Hilo (HI), Mauna Loa (HI), Honolulu (HI), Lihue (HI), Johnston Island, Canton Island, Tutuila (AS), Wake Island, Enewetak Atoll, and Anderson AFB (GU).

Fig. 21 shows that in 1957 and 1958, the detected values of ^{90}Sr were very low, especially in 1957 where concentrations were only as high as 0.25 mCi/km^2 in November in Honolulu. No detections were observed at other sites on the oceans, so the maps of Guam, Canton Island, etc. are not included. In 1958 only a slight increase in ^{90}Sr occurred in Honolulu; the lowest concentration was 0.03 mCi/km^2 in July, and the highest concentration

was 0.51 mCi/km^2 in March.

(a)



(b)

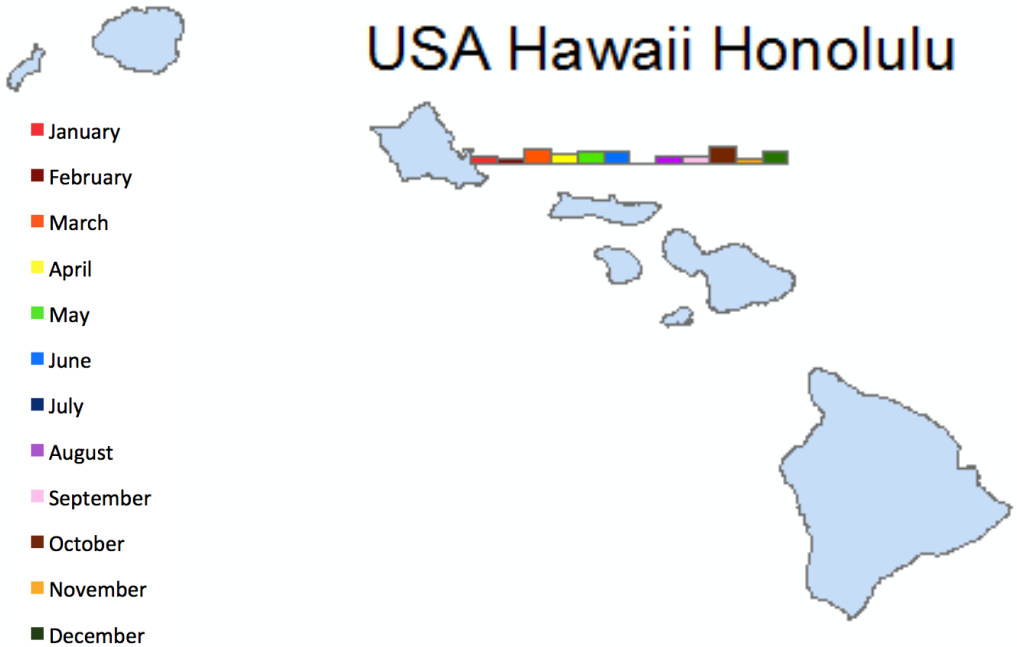
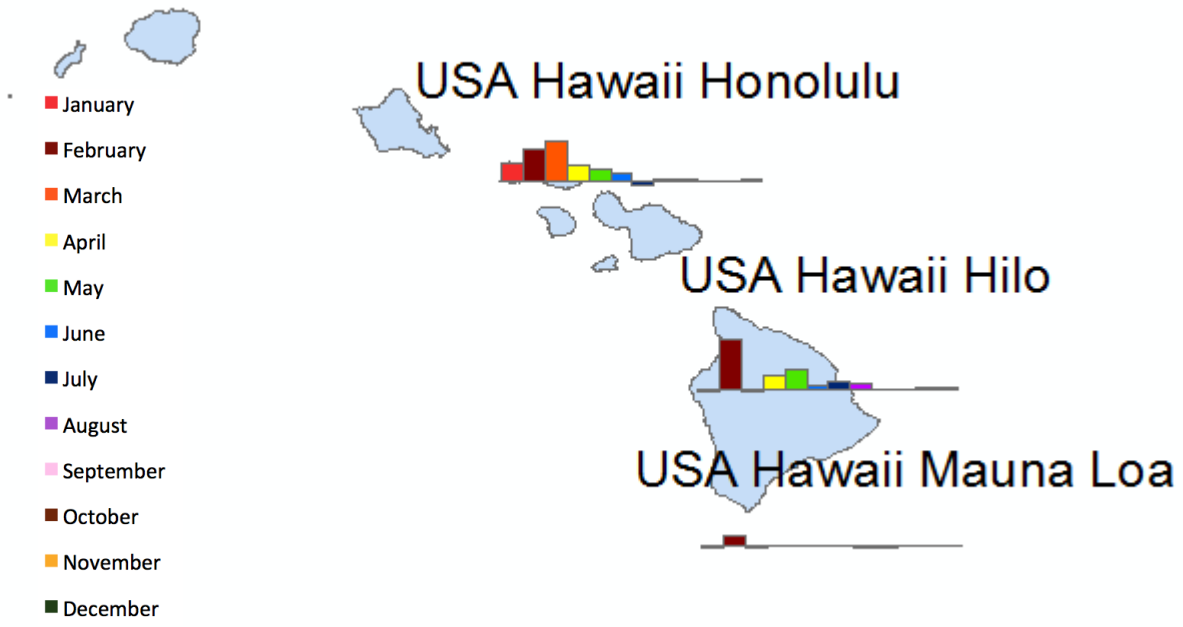


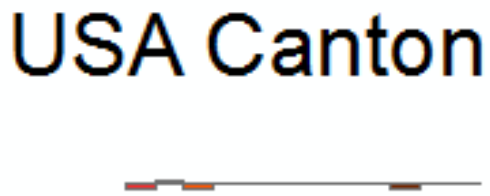
Fig. 22. ^{90}Sr concentrations in Hawaii in (a) 1957 and (b) 1958

The first detections of ^{90}Sr at sites such as Hilo, Mauna Loa, and Canton Island occurreded their first detection in February 1959 (Fig. 22), at 2.32 mCi/km^2, 0.49 mCi/km^2, and 0.16 mCi/km^2, respectively. Western of Hawaii are sites on Wake Island and Guam, where the first values were found later in April at 0.75 mCi/km^2, and in September at 0.02 mCi/km^2, respectively. Following the first detection, the next month usually presented no detection or decrease. Honolulu on the contrary had a stable increase since January, and reached a peak at 1.88 mCi/km^2 in March, followed by a rapid decrease through the end of the year.

(a)



(b)



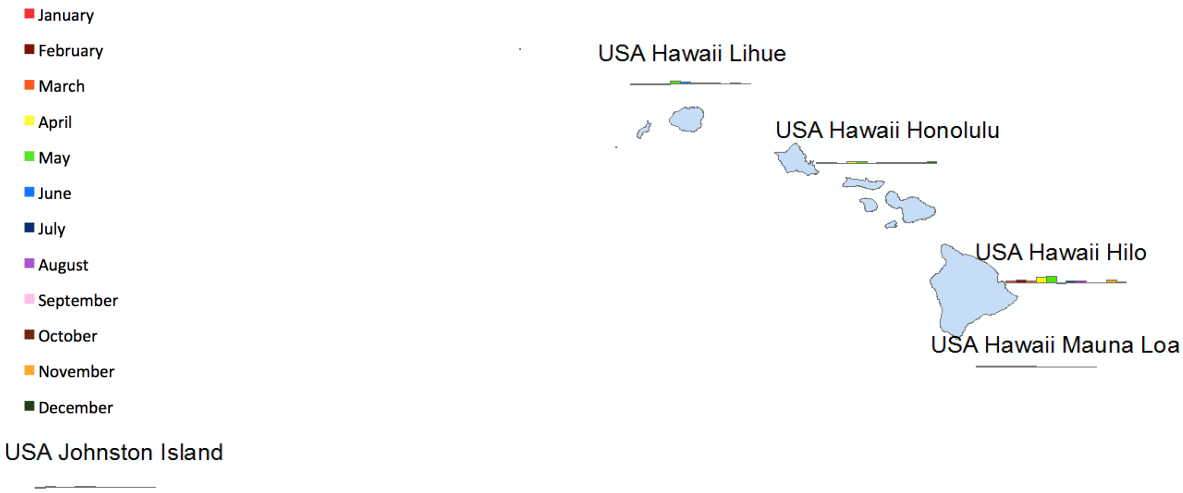
(c)

Fig. 23. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, and (c) Guam and Wake Island, in 1959

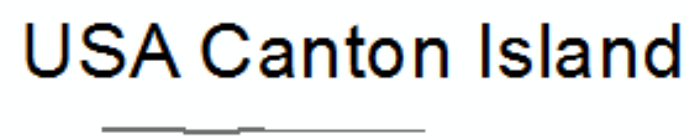
San Juan and Johnston Island were first monitored in 1960, when all sites had very low ^{90}Sr concentrations over the entire year (Fig. 23). The highest value of ^{90}Sr,among all the islands, was 0.61 mCi/km^2 which was detected at Hilo in May. There was no significant increase observed in 1961 as well (Fig. 24); however, since January 1962 many sites showed a large increase of ^{90}Sr and reached the highest ^{90}Sr concentration since the beginning of monitoring (Fig. 25). For Hawaii, sites on Hilo, Lihue, Honolulu, and Mauna Loa had decreasing concentrations. The first three sites had increasing concentrations between

January and May; in contrast, Mauna Loa did not show high concentrations until November, at 2.31 mCi/km^2. Though there were many bomb tests during July to October with high yield (12176.3kt) above Johnston Island, the site itself did not show high ^{90}Sr concentrations during these months. Two increasing trends were observed between January and March, and between May and August. A similar trend was observed in Guam and San Juan, which are located in two different oceans. A peak in concentration was noted in January followed by a decrease and a second peak in April. Other islands, such as Canton Island and Enewetak Atoll had no significant concentrations, except in August on Canton Island, which showed a small peak concentration of 0.44 mCi/km^2.

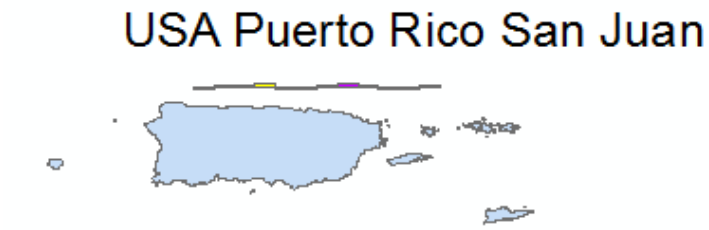
(a)



(b)



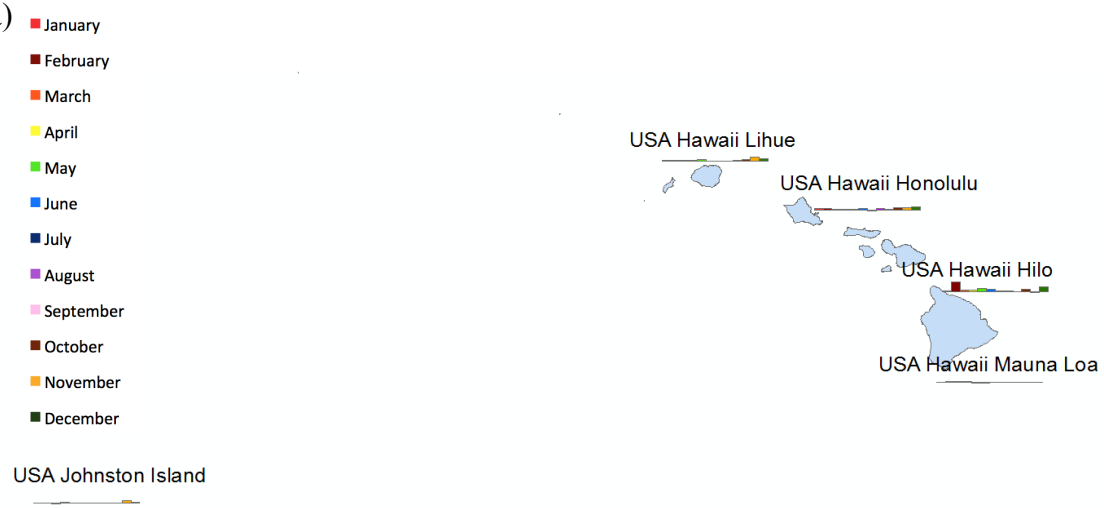
(c)



(d)

Fig. 24. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1960

(a)


January
February
March
April
May
June
July
August
September
October
November
December
USA Johnston Island
USA Hawaii Lihue
USA Hawaii Honolulu
USA Hawaii Hilo
USA Hawaii Mauna Loa

(b)


USA Canton Island

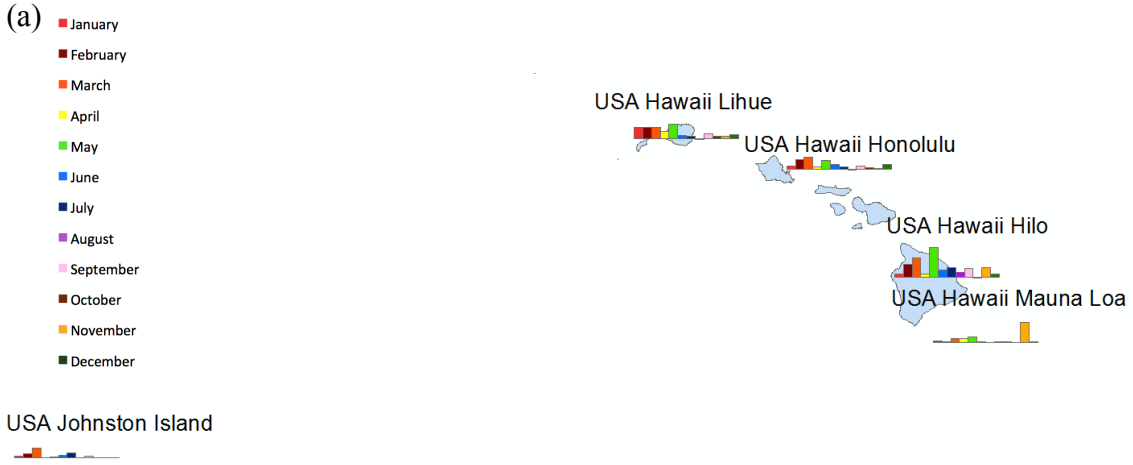
(c)


USA Puerto Rico San Juan

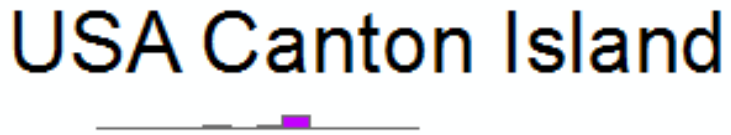
(d)

Fig. 25. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1961

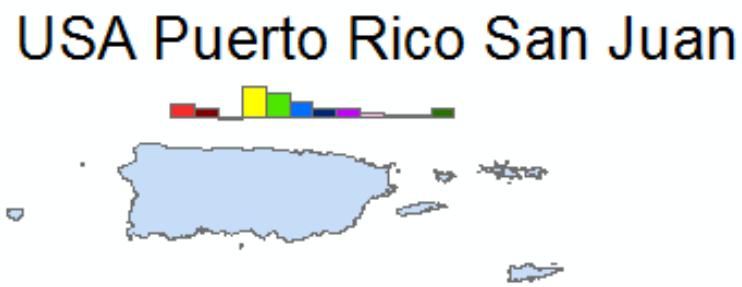
(a)



(b)



(c)



(d)

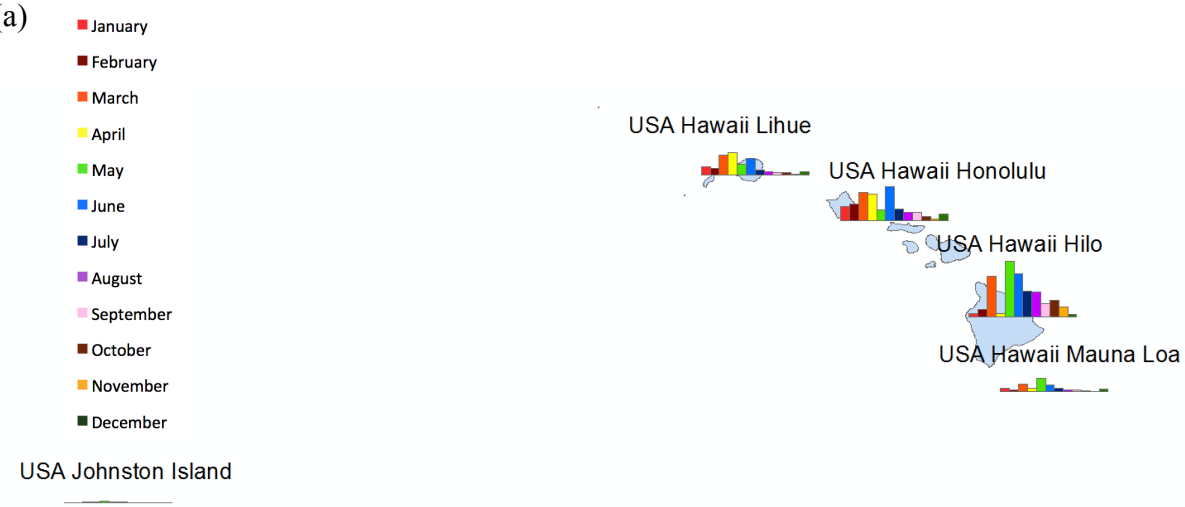
Fig. 26. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1962

In years of 1963 (Fig. 26) and 1964 (Fig. 27), high concentrations of ^{90}Sr were observed at the sites in Hawaii, San Juan, Wake Island and Guam but not in Johnston Island, Canton Island and Enewetak Atoll. In 1963, Hawaii, Hilo showed the most ^{90}Sr inventory, especially in March, May and June, when the concentrations increased to over 4.5 mCi/km^2, and reached as high as 6.17 mCi/km^2. The two sites north of Hilo had a similar change, while Honolulu had a higher average value; however, high ^{90}Sr values were not observed on Mauna

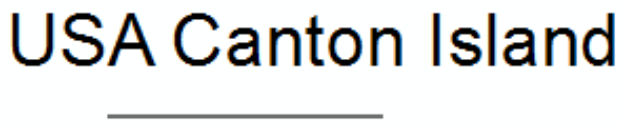
Loa, which is south of Hilo. In the Atlantic Oceans, San Juan had a peak value of 1.79 mCi/km^2 in April, and some medium concentrations were observed between May and August. Though Guam and Wake Island did not indicate a strong increase from the beginning of the year, a rapid increase occurred in May, which reached 4.51 mCi/km^2 and 1.89 mCi/km^2, respectively. Additional peak values occurred a month later in June, but were lowerthan the other sites in Pacific Oceans. In 1964, a similar result was observed, but the concentrations in June decreased significantly from 6.84 mCi/km^2 to 0.25 mCi/km^2 in Hilo, but greatly increased from 0.84 mCi/km^2 to 4.4 mCi/km^2 in Lihue. Overall, the concentrations of ^{90}Sr were lower in 1964, and then decreased more greatly toward 1965 and 1966.

In 1965, monitoring in American Samoa Tutuila began. Detected concentrations except in October of 1965 and September of 1966, were low, approximately 0.1 mCi/km^2 or even less. Though the values were continuing to decline, there were still some high peaks in Hilo and San Juan, especially in June of 1965. The results are presented in Fig. 28 and Fig. 29.

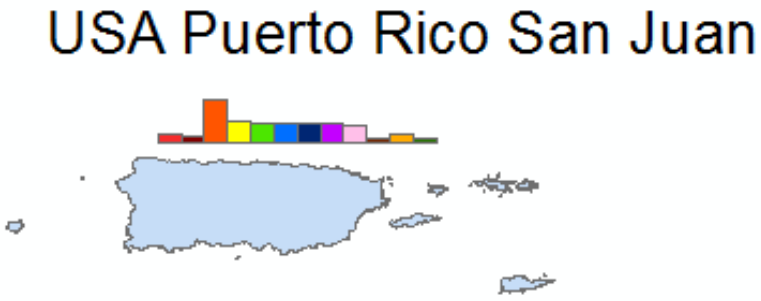
(a)


January
February
March
April
May
June
July
August
September
October
November
December
USA Johnston Island
USA Hawaii Lihue
USA Hawaii Honolulu
USA Hawaii Hilo
USA Hawaii Mauna Loa

(b)


USA Canton Island

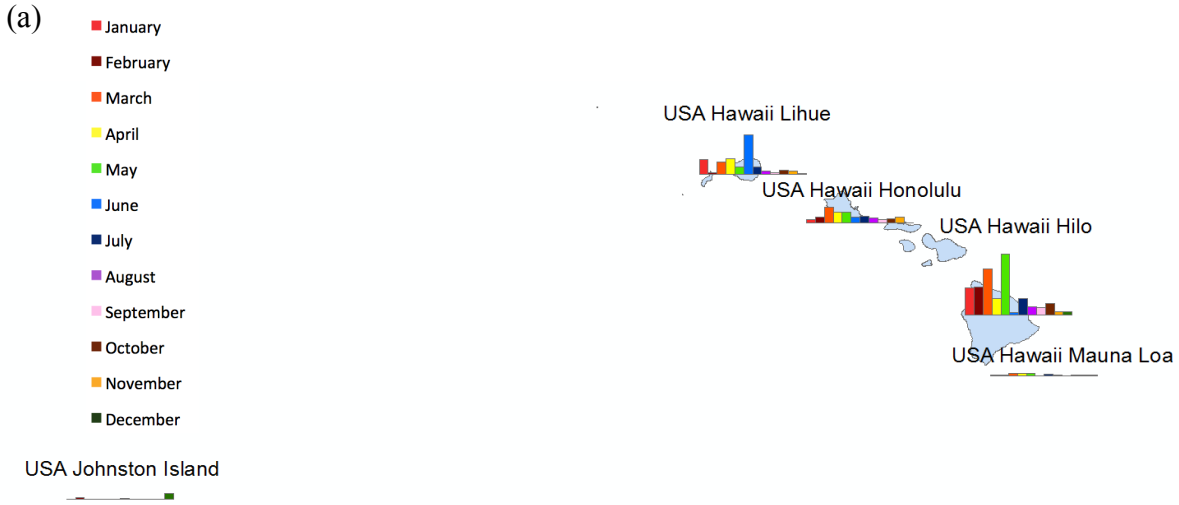
(c)


USA Puerto Rico San Juan

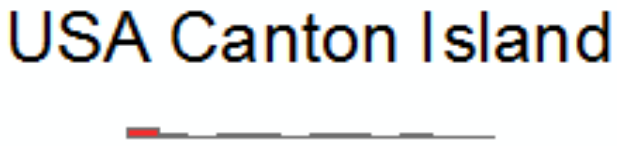
(d)

Fig. 27. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1963

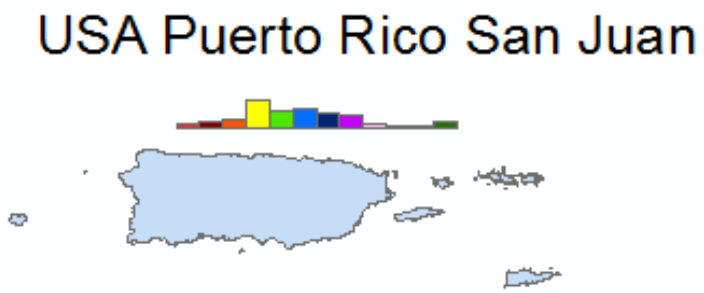
(a)



(b)



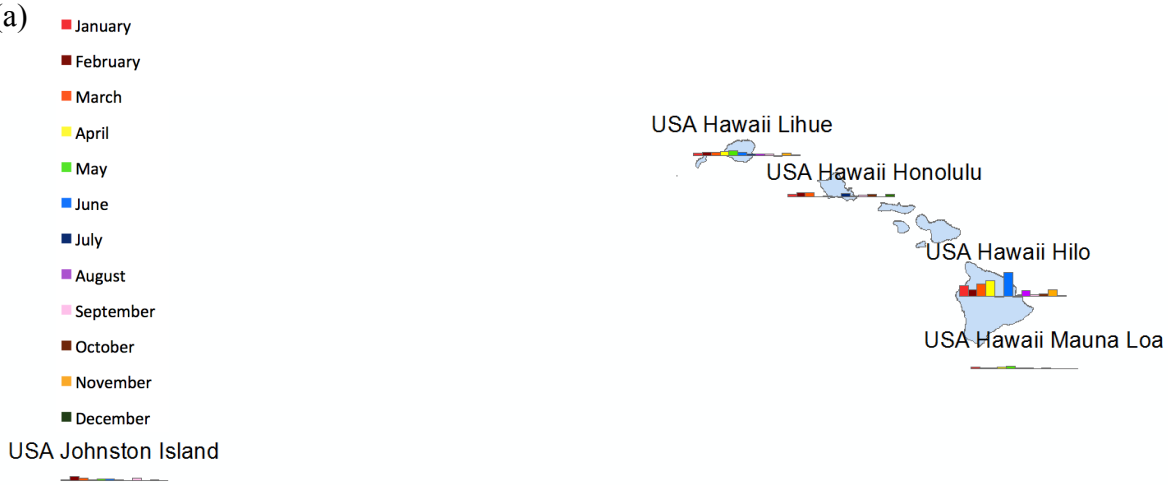
(c)



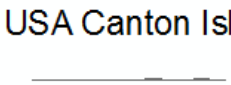
(d)

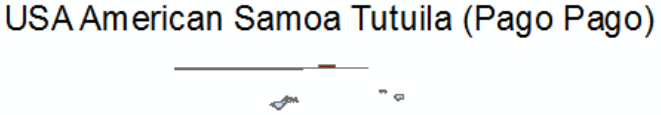
Fig. 28. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1964

(a)

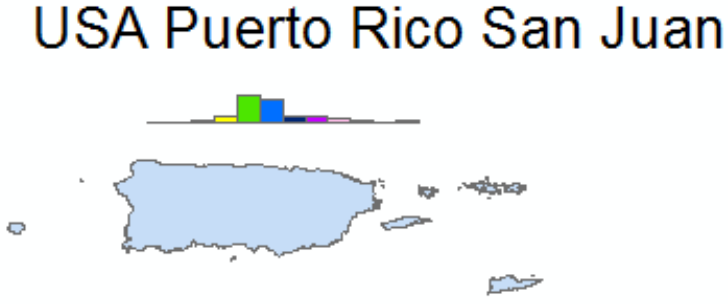


(b)


USA Canton Island


USA American Samoa Tutuila (Pago Pago)

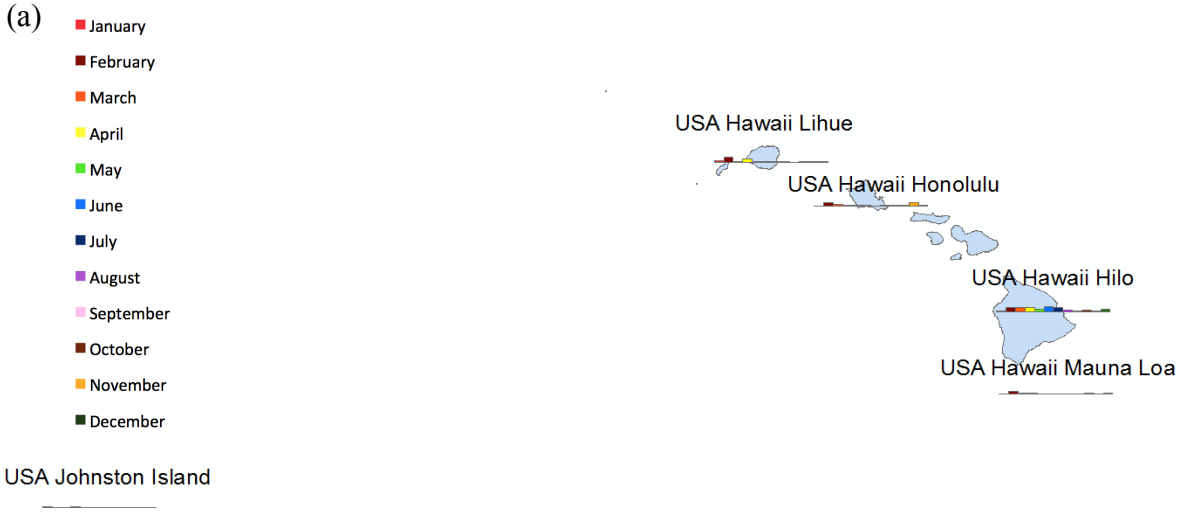
(c)


USA Puerto Rico San Juan

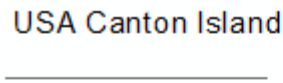
(d)

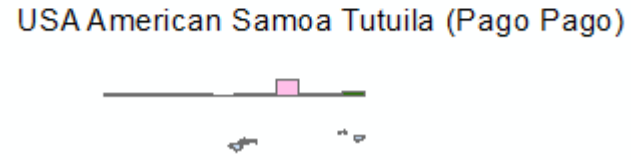
Fig. 29. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1965

(a)

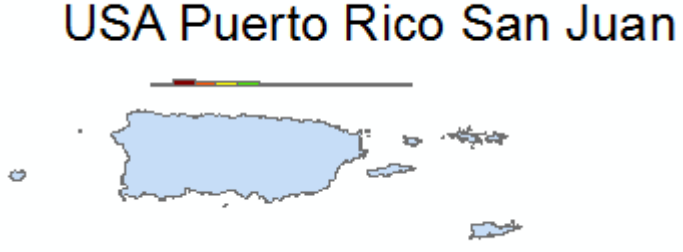


(b)


USA Canton Island


USA American Samoa Tutuila (Pago Pago)

(c)


USA Puerto Rico San Juan

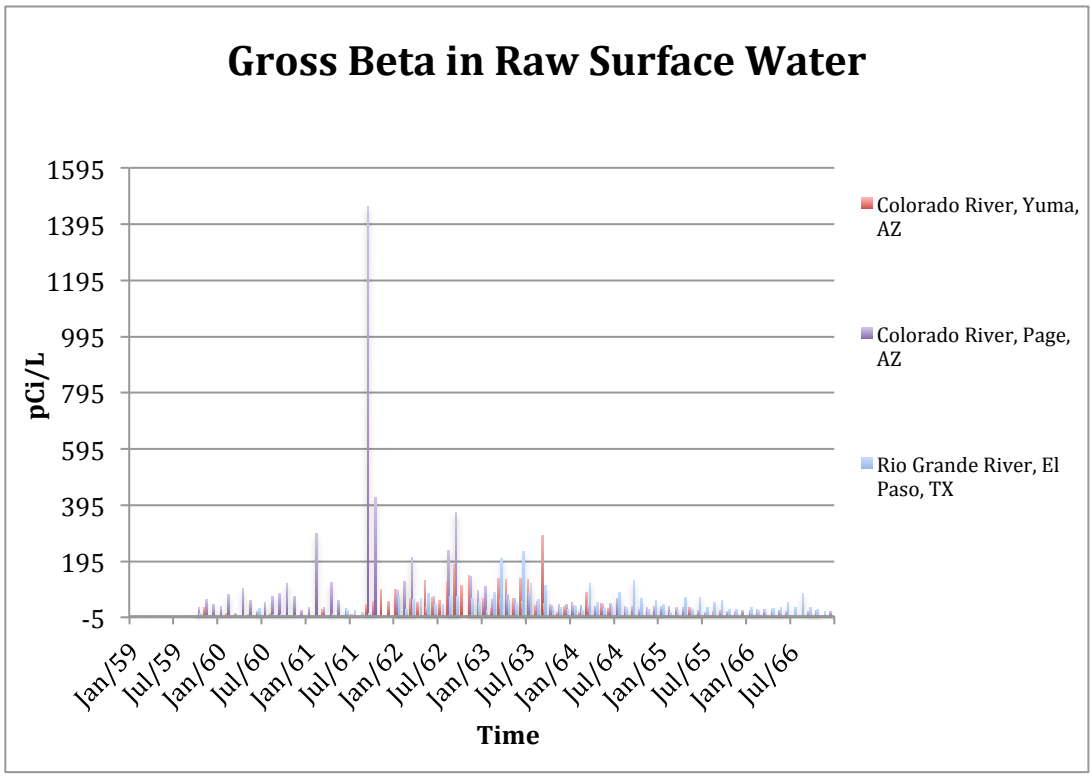
(d)

Fig. 30. ^{90}Sr concentrations in (a) Hawaii, (b) Canton Island, (c) San Juan, and (d) Guam and Wake Island, in 1966

3.2 RHD results

3.2.1 Gross beta in raw surface water

In the RHD reports, two types of values, representing data obtained from suspended particles and dissolved solids were recorded. We sumed up both of the values in each month to generate a monthly total beta gross in the water body. Gross beta detected in raw surface water is presented in Fig.29. To present the figure clearly, it is divided to two parts: the west (including Colorado River and Rio Grande River), and the east (including Mississippi River, Tennessee River, and St. Lawrence River). Because RHD reports before October 1959 are not available, the database from the previous years are not included in our section.



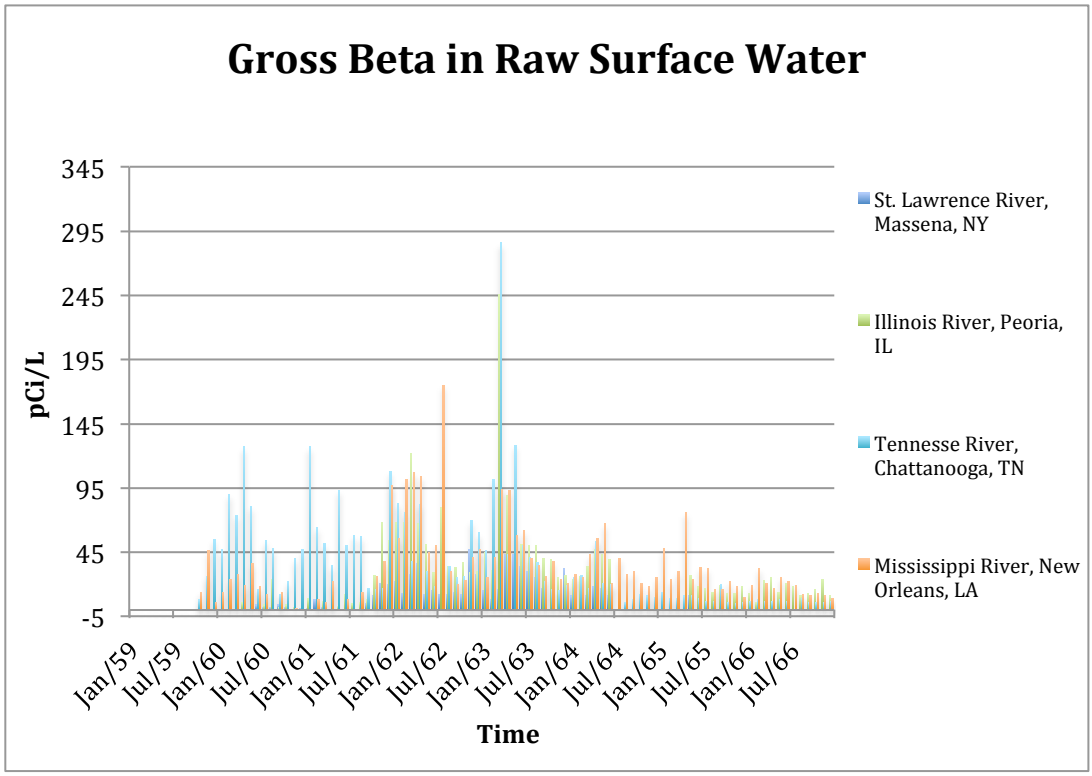


Fig. 30. Gross beta detected in 7 cities by the rivers: Yuma and page (Colorado River), El Paso (Rio Grande River), Massena (St. Lawrence River), Peoria (Illinois River), Chattanooga (Tennessee River), and New Orleans (Mississippi River)

A broad picture of the sampling sites shows that high gross beta was generally detected

during 1961 and 1963. While New Orleans, Chattanooga and Page had high concentrations since 1960, the other cities had no obvious increases or peaks. Because values of ^{90}Sr at Chattanooga were missing in 1960, there was no clear correlation to indicate ^{90}Sr content and gross beta as expected, Page on the other hand was expected to have high gross beta concentrations due to its proximity to the Nevada Test Sites.

In Yuma, except a low value of 1 pCi/L recorded in October 1959, most of the months the values were undetected or recorded 0 pCi/L until September 1961. Gross beta radiation gradually increased in 1962, and was constantly detected. In September 1962 in Yuma, the highest value was 120 pCi/L, and the second highest value of 97 pCi/L occurred in October. After July 1964, RHD reports did not present gross beta at Yuma except May 1965, which was only 1 pCi/L. Other detections of radium-226 and uranium were detected over this period, showing that the site was still actively sampling. Because the reports did not explain why they gross beta concentrations were not shown at Yuma after July 1964, we set these monthly concentrations at -0.1 pCi/L to indicate these were missing data.

Page, Colorado is located on the upstream of Colorado River, north of Yuma and high gross beta concentrations were routinely detected. In most months, gross beta collected at this

site was highest among the seven stations. An extremely high value of 1457 pCi/L was detected in September 1961. Rapid increases and high concentrations were mostly detected in the period of 1961 and 1963, nevertheless, some values were not measured in some months, causing missing values in this period. From February 1963 through December 1966, the values decreased slowly and kept under 100 pCi/L. Though the radiation was normally lower in Yuma, some values were found higher during January and September of 1963. These might be attributed to the transportation from upstream to downstream, and regarded as the residual of beta emitters detected during the peak period in Page.

El Paso had relatively low values at the start of 1960 compared to all of the stations, but a large increase was observed in January 1962. A peak of 230 pCi/L was measured in June 1963, followed by upward and downward swings in concentrations. Many months with values higher than 100 pCI/L were observed, and were among the highest observations compared to other sites.

In contrast, Chattanooga remained high at the beginning of 1960, but fell low after the peak at 286 pCi/L occurring in March 1963. Gross beta no higher than 20 pCi/L was detected after July 1964. Generally the concentrations decreased after July and then increased again in

September.

Gross beta was detected in New Orleans at the beginning of 1960, and declined in 1961 until November, when gross beta concentrations increased. In July 1962, a peak of 175 pCi/L was measured, followed by some ups and downs. At this site, the dataset indicates highest concentrations were observed in April, and lowest concentrations observed in September.

The Illinois River and St. Lawrence Rivers were first included in the monitoring program in March 1960 and May 1960, respectively. Before October 1961, the values recorded at the Illinois River barely exceeded 10 pCi/L, but afterwards gross beta radiation was frequently observed over 30 pCi/L. The greatest value of 246 pCi/L was recorded in March 1963, followed by a reduction to 89 pCi/L in April, and a slow decrease until April 1964, when some light increases happen again until June 1964.

Gross beta radiation was detected in the St. Lawrence River during the whole monitoring program. The highest concentration was recorded in November 1962, at 47 pCi/L, which was the only detected concentration higher than 30 pCi/L. The average gross beta in water from 1959 to 1966 for Massena, Yuma, Page, Peoria, El Paso, Chattanooga and New Orleans were 7.042 pCi/L, 28.125 pCi/L, 66.063 pCi/L, and 22.188 pCi/L, 33.281 pCi/L, 33.229 pCi/L and

28.854 pCi/L, respectively. Gross beta was more likely to be detected in the water system of Colorado River.

3.2.2 Gross beta in air

Gross beta values in air are relatively lower than the ones detected in raw surface water. Fig. 31 shows that the highest concentration recorded at the four sites was approximately 40 pCi/m^3. In this criterion, we notice that between July 1959 and August 1961, data were always lower than 0.3 pCi/m^3. Before this period, there were some outstanding values observed in Los Angeles, for example, October and November of 1958 the values were 19 pCi/m^3 and 38 pCi/m^3, respectively. While the high values were presented in these two months, other sites did not perform the same significant values, and did not change a lot compared to the previous months. However, some similar changes had been detected among these sites as well, which could present a common trend of value change. In the two sites on the east coast, values dropped rapidly from October to November of 1957, and then slowly grew back to reach a peak in April 1958. An obvious decrease was found between April 1958 and October 1958 in all of the sites. For those places located in the west coast of the U.S., the decrease happens one month earlier in March. After September 1961, these four sites

performed a similar change again, where a first decrease continued until September 1962, followed by an increase for approximately three month. The rest of the months before June 1963 might have some bumps and up and down, but overall after this time, the values declined, and then remained lower than 1 pCi/m^3 after July 1964.

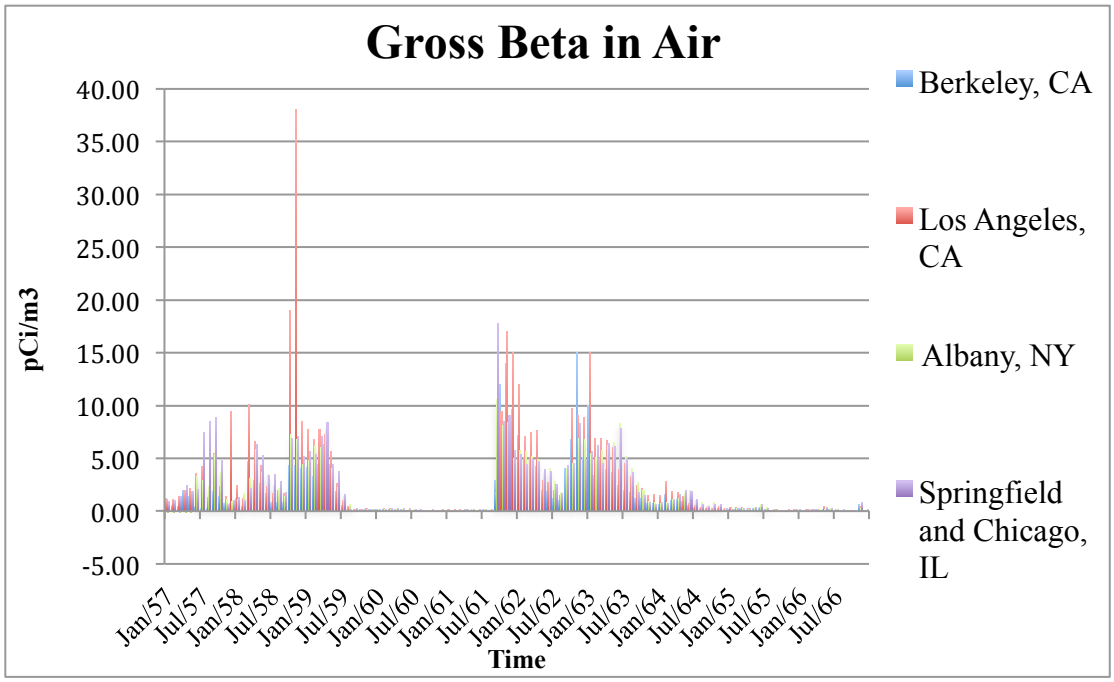


Fig. 31. Gross beta recorded in air from the four sites: Berkeley (CA), Los Angeles (CA), Albany (NY), and Springfield and Chicago (IL)

3.2.3 Gross beta in milk

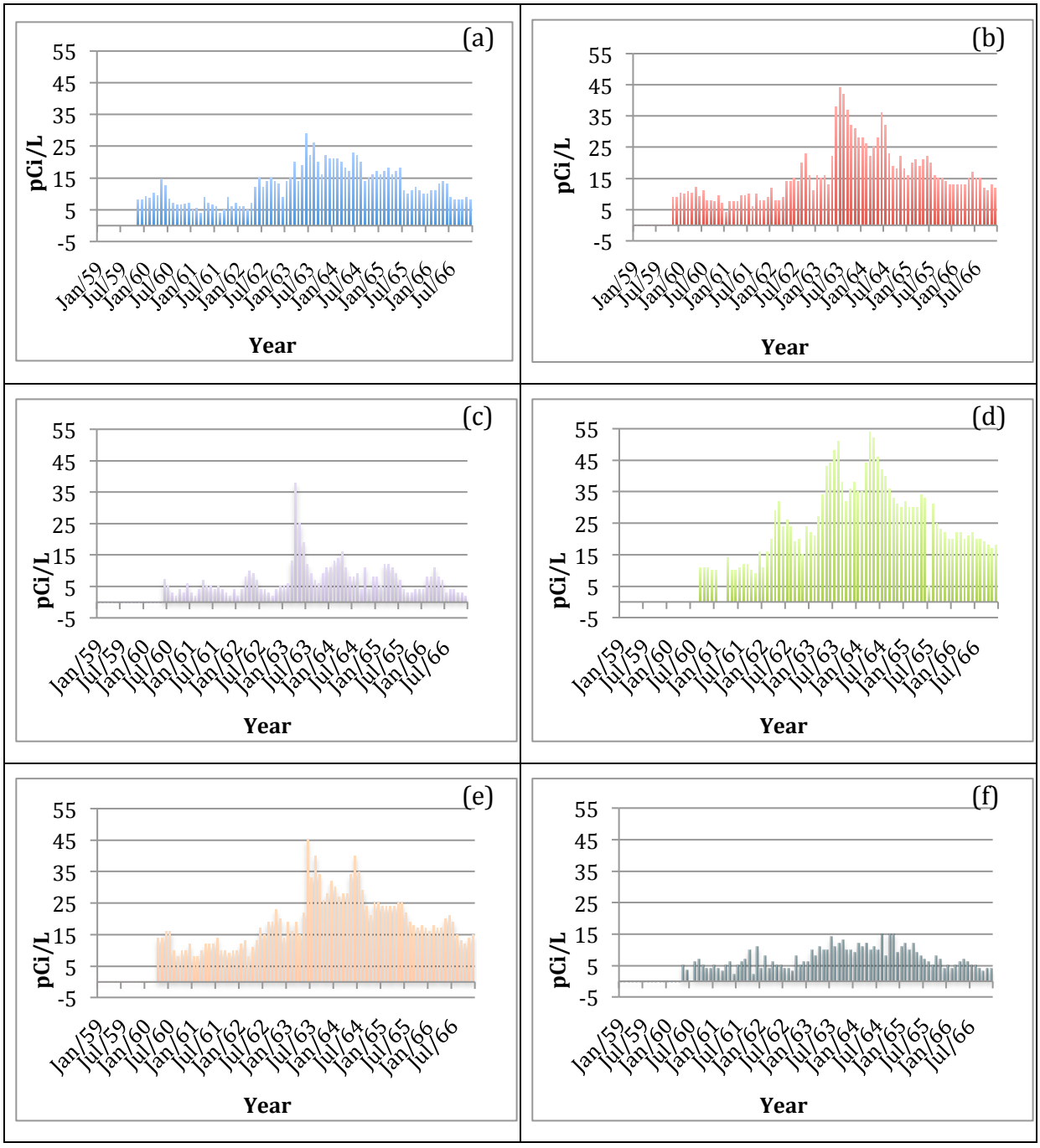
Fig. 32 presents monthly gross beta in milk from January 1959 to December 1966 in the eight sites chosen for this paper. Some stations were included after the summer of 1960; therefore, the first detected concentration of each site does not appear at the same time.

Because of the unavailability of data before 1959, this thesis only shows the gross beta concentrations after 1959. Generally, each site showed a slight decrease of ^{90}Sr concentration between 1961 and 1962, followed by a significant growth after June 1963. The average value of the 8 years (1959-1966) can be ranked from the highest to the lowest in the order of Chattanooga (24.776 pCi/L), Louisville (19.617 pCi/L), Pittsburgh (19.111 pCi/L), New York City (16.248 pCi/L), Salt Lake City (13.716 pCi/L), Chicago (12.509 pCi/L), Honolulu (7.269 pCi/L), and San Francisco (7.143 pCi/L). It can be seen that between October 1959 and March 1960, values in Chicago, New York City, and Salt Lake City performed values between 5 pCi/L and approximately 10 pCi/L. After March, the values vibrated and had some slight ups and downs. Pittsburgh and Louisville started to perform ^{90}Sr in April 1960, at 14 pCi/L and 5.1 pCi/L, respectively, and increased their values gradually for a few months afterwards. Honolulu, San Francisco and Chattanooga were put into the monitoring program in May 1960, June 1960 and September 1960, respectively. While Chattanooga presents a stable value of around 10 pCi/L since the beginning, the other two sites performed relatively low values among the stations.

A changing point is noticed in April 1962, when the ^{90}Sr had risen up gradually,

following a little decrease between October 1961 and March 1962. Louisville and Chattanooga had both increased the values to a range between 20 pCi/L and 27 pCi/L, which are about 5 pCi/L higher than the ones of Pittsburgh, Chicago and Salt Lake City. In contrast, Honolulu and San Francisco have remained steady at the same concentration level. A second changing point presenting a high peak was firstly observed in April 1963, which caused the value detected in San Francisco reach as high as 38 pCi/L. During the period of June and August 1963, most of the sites presented their highest concentration. The values for Chicago, New York City, Salt Lake City, Louisville, Pittsburgh, Chattanooga and Honolulu, were 29, 44, 39, 46, 45, 51 and 14 pCi/L, respectively. However, while other sites began increasing their value since May 1963, the values of San Francisco dropped rapidly in three month, and returned to a low lever under 15 pCi/L, and mostly under 10 pCi/L. Following the first great peak in 1963 was a second peak observed around June 1964. Except Chicago, Honolulu and San Francisco, peaks at other sites could still be higher than 35 pCi/L. After some peaks, ^{90}Sr had slowly decreased. Fig. 32 shows that the concentrations of these sites, except San Francisco remained stable for few months, and then dropped to a lower level but kept remaining stable throughout December 1966.

A clear observation shows that though in Honolulu the values did not vary greatly, a more stable level of the concentrations without big drops made the average concentration still considerable and higher than the one of San Francisco, where some extremely low values had been recorded.



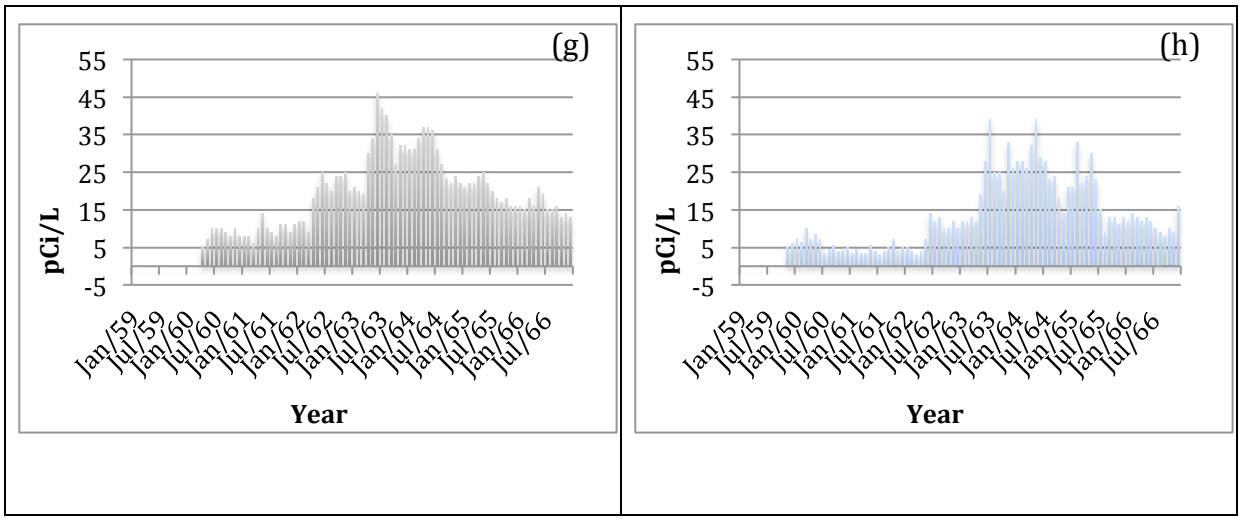


Fig. 32. ^{90}Sr recoreded in pasteurized milk in the following stations: (a) Chicago (IL), (b) New York City (NY), (c) San Francisco (CA), (d) Chattanooga (TN), (e) Pittsburgh (PA), (f) Honolulu (HI), (g) Louisville (KY), and (h) Salt Lake City (UT)

4. Discussion

a. Produced radioactivity has exceeded the international regulations:

A big concern of these radioactive products is that it may affect human health through the food chain, and remain active for a long time to enhance its threat to the environment. A regulation, known as the Guidelines for Drinking-water Quality published by WHO (World Health Organization), which was initially published in 1993 has set a threshold of 1 Bq/L of gross beta as a safety standard in its fourth edition announced in 2011 (Damla, 2006; WHO, 2011). A unit of 1 Bq/L can be transformed to 27.027 pCi/L; therefore by referring this standard number to the database presented in Fig. 30, it is noted that Page, Peoria and Chattanooga have 77, 66, and 84 months out of 96 months, respectively, exposed to a high radiation situation. The water then strictly required treatments before applied as drinking water or any kinds of available water usage.

b. A delay of detection occurs for approximately one to two years after the bomb tests:

For the results shown in Fig. 1, Fig. 13, and Fig. 22, we can see that ^{90}Sr is greatly detected in 1959 in these areas. However, the catalog of U.S. Weapons Tests indicates no event of nuclear bomb testing occurred in the U.S. in the same year. Furthermore, as 1959 is the year for global suspension of bomb tests, the detected data can be assumed that be

contributed from the previous tests before 1959. A similar result was observed in 1960 and 1961 when the bomb testing restarted. In 1962, all of the bomb tests had been conducted fully underground; therefore, the radioactive compounds were more difficult to be released into the atmosphere. However, high inventory of ^{90}Sr between 1962 and 1963 was still observed in the same years. A report published by Dasher et al. (2002) indicated that the distribution of radioactive compounds from the underground tests might leak through the freshwater systems but not directly to the atmosphere. Radioactive gases released to the atmosphere through the drill pipes of the base is the primary route for transporting radionuclides to the atmosphere, but compared to the surface bomb tests the amount of the leaking radionuclides is smaller. This implies that atmospheric deposition of ^{90}Sr fallout becomes more difficult to detect. Eventually, correspond to the discovery of high concentrations of ^{90}Sr in 1962 and 1963, we can assume that the detected ^{90}Sr originated from the previous years. This assumption has been proved by the literature conducted by Sotobayashi and Koyama (1966). The two authors highlighted that a major fraction of ^{90}Sr detected between 18 October 1964 and 31 March 1965 were derived from the 1961-1962 test series. Also, the result was enhanced by the prolonged storage in the stratosphere. Furthermore, another research undergone by Fabian et

al. (1968) indicates that the stratospheric residence time of fallout can last approximately 1.6 years through the precipitation, which can be referred to wet deposition. An expected inter-hemispheric mixing on the other hand normally takes 3.3 years.

c. Precipitation may primarily affect the transport of ^{90}Sr:

Evrard et at. (2012) indicated that radionuclides could travel across oceans within a short period. For example, the ^{131}I produced due to the Fukushima accident was detected in France after 11 days of the detonation due to the transportation through the radioactive cloud. It can be applied to the assumption that fission products produced in the Pacific Oceans or Nevada Test Sites could also reach the east coast of the U.S. However, a comparison of ^{90}Sr concentrations between the east coast and the west coast of the U.S. pointed out that an unexpected result of the distribution was not dominantly related to the distance between monitoring stations and the test sites. Russia was known to develop nuclear weapons in the same decade, so the testing conducted in Europe is thus presumed to be a factor to affect the inventory of radionuclides in the U.S.

No researches had found the relation of fission products detected in the U.S. and the testing activities in Russia, but some latter reports investigating the transport of fission

products from the Chernobyl accident provided some information to analyze this relation. The report published by Feely et al. (1988) highlighted that radioactive cloud of debris transported across the globe, could actually bring radionuclides from Ukraine to New York; thus, the contribution of the radionuclides by the tests in Russia is considerable. The increasing fission products and gross beta radiation were also noticed in New York after half a month of the Chernobyl incident (Larsen et al., 1989). Therefore, as Russia released high yield during its frequent bomb tests in 1960s, it is more possibly to observe its effects onto the U.S. Feely et al. (1988) also emphasized that the transport of radionuclides was significantly affected depending on whether it was a wet or dry deposition. By analyzing the fraction of ^{131}I and ^{137}Cs, it was confirmed that only 20%-40% of the deposition of these radionuclides comes from dry deposition, and the rest of it was due to the wet deposition, which referred to precipitation. Because ^{90}Sr is less volatile than ^{131}I and ^{137}Cs, it is expected predominantly influenced by gravity settling through precipitation. On the other hand, many factors can affect the composition of radioactive cloud, for example, temperature and time. Therefore, they should also be taken into the analysis to examine their effects. It is suggested that the distribution of ^{90}Sr should be analyzed by adopting more factors, especially the

amount and the type of the deposition.

d. Bomb testing sites with high yield does not guarantee higher ^{90}Sr content on site:

By comparing ^{90}Sr from fallout with the yield generated from the bomb testing, the result shows that higher yield does not always determine a higher inventory of radionuclides on site, for example, Johnston Island. A research completed by Simon et al. (2004) pointed out that regions located east of Nevada Test Site could receive high ^{90}Sr content immediately after the tests conducted in Nevada, and many figures in the paper also showed a high intensity of radionuclides detected close to Nevada. Therefore, it may explain why Salt Lake City shows high concentrations because it is the closest station to the bomb testing site, and located between the latitude of 10°N and 60°N which commonly received the most fission products based on several researches (Libby 1957).

However, cities located thousands miles away from Nevada Test Sites, such as New York City and Coral Gables still showed high values. The result may be explained by the fact that yield of bombs on the Nevada Test Sites were less than 500 kt. Based on a research by Warneke et al. (2002), radionuclides from low-yield tests (<500kt) can only retain in the troposphere and create instant effect on the environment. As Johnston Island always had test

yield higher than 500 kt, radionuclides thus entered the stratosphere, and distributed globally.

Fabian et al. (1968) used the database of fallout in rain, which was compiled by Health and Safety Laboratory, the predecessor of EML, to calculate the stratospheric residence time on specific sites. The time was calculated by the following equation:

$$(Concentration\ of\ ^{90}Sr) \cdot \varphi = C(\varphi) \cdot \cancel{X}_0 \cdot e^{-\alpha t}$$

where factors as transfer constant (C(φ)), latitude (φ), ^{90}Sr content at t=0 (X_0), and coefficients of the exponential law (α) were involved. This result showed that the residence time of Johnston Island was 1.23 years, which was higher than sites with higher ^{90}Sr. Thus we can deduce that the longer the stratospheric residence time, the lower the ^{90}Sr content because these radionuclides is less easily to settle down to the land surface. It is noted at some stations that the result presented by Fabian et al. does not positively correspond to EML's database. The difference can be assumed to be affected by other variables or these researchers only adopted fallout sample in the rain, but not through dry deposition. Further experiments need to be implemented for a complete analysis.

e. Summer has lower ^{90}Sr than other seasons:

Though bomb testing was usually conducted in summer, except in Nevada, ^{90}Sr was

mostly observed higher in spring or winter. According to a paper from Krey (1967), it emphasized the vertical transport at the mid-latitude of the northern hemisphere in summer is lifting, and in spring is subsiding. Therefore, it is assumed that the vertical diffusion hinders ^{90}Sr from falling to the ground in summer, and result in a lower detection in the fallout deposition at the same time. The time or the bomb testing therefore is not a dominant reason to affect the time of the discovery of high ^{90}Sr content.

f. Southern California has higher ^{90}Sr than northern California:

There is no strong evidence to prove the reason of higher ^{90}Sr content in West Los Angeles, but not in North California cities, such as in San Francisco, Richmond and Palo Alto. In EML's database, it shows the higher precipitation occurred in northern California, which can barely conclude that high ^{90}Sr content is determined by wet deposition. A further research is recommended for providing a clearer pattern of how radionuclides can distribute horizontally in California.

g. ^{90}Sr shows poor relation to the gross beta radiation in water and milk.

A significant portion and a long half-life of ^{90}Sr make researchers believe ^{90}Sr has a relatively high contribution to the gross beta in the environment. In order to examine the

accuracy of this hypothesis, stations that present gross beta in milk or in water, and ^{90}Sr content are chosen for an advanced analysis. Though the stations used on each of the research are not exactly located on the same location; however, if the stations are located in the same city, they are considered to be able to present the same station.

In the analysis of the relevance of ^{90}Sr and gross beta in water, the following sites are chosen: Chattanooga, El Paso and New Orleans.

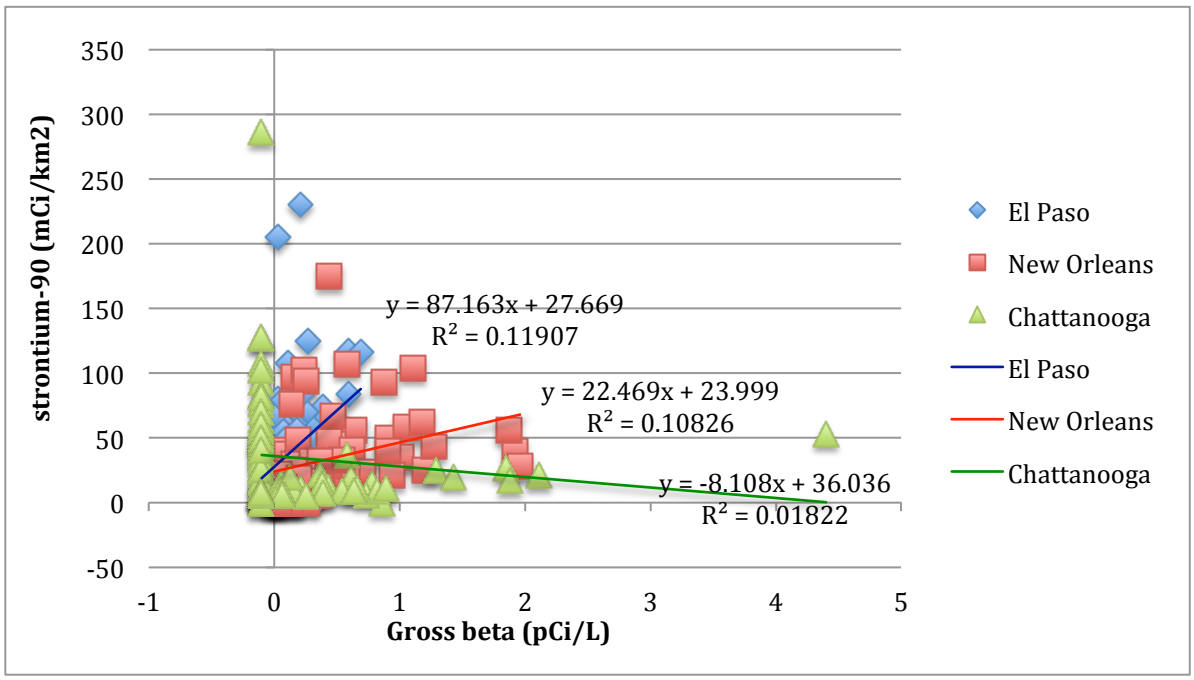


Fig. 33. Relation between gross beta in raw surface water and ^{90}Sr from fallout, in El Paso (TX), New Orleans (LA), and Chattanooga (TN)

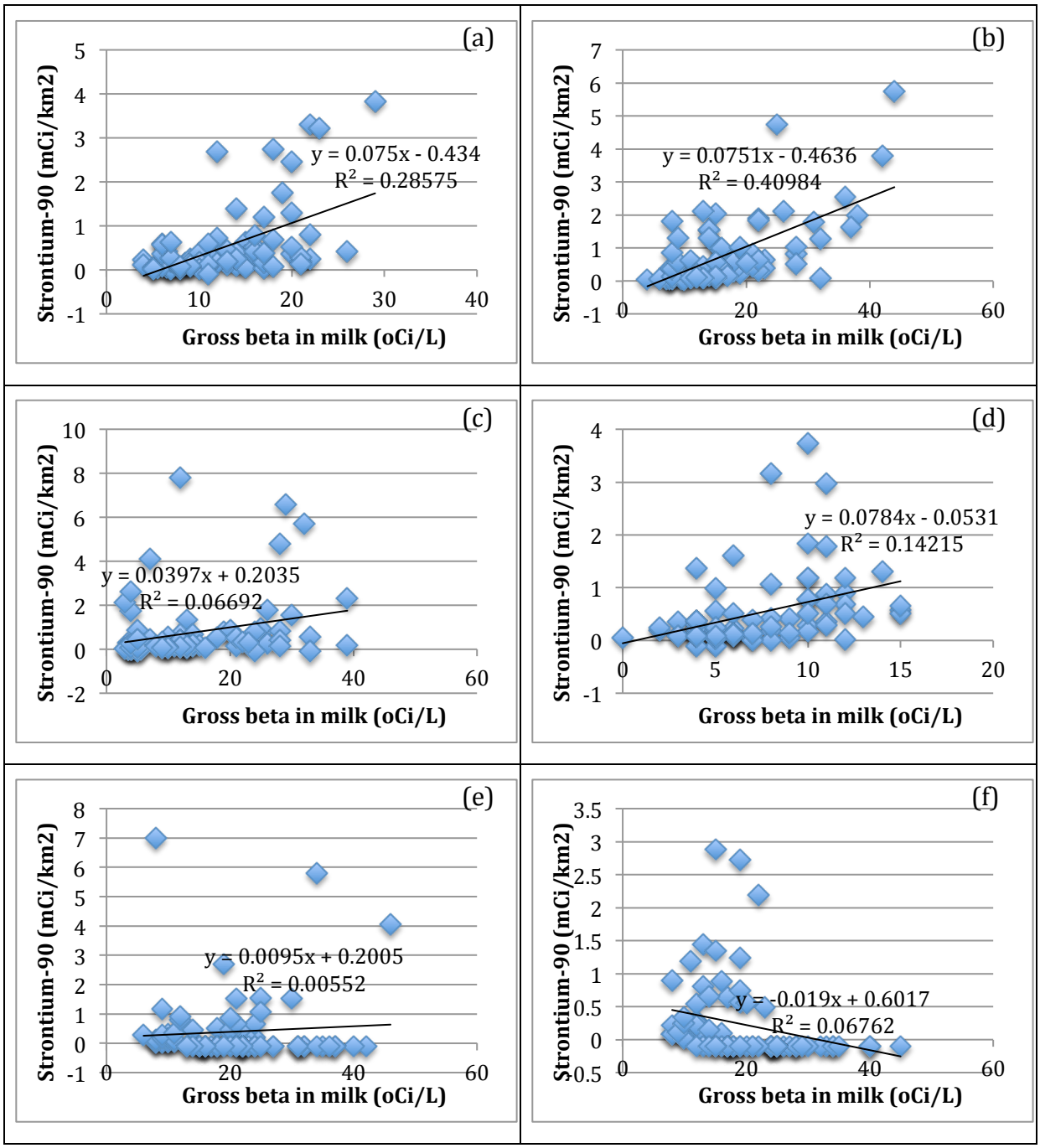
Because there are many months, which were not included under the investigation in the early years, or not able to present values, the period for this analysis was set from July 1960 to December 1961, to prevent the bias caused by missing data. First of all, the points are

plotted on the figure, using the x-axis and y-axis to represent gross beta and ^{90}Sr, respectively. Then, the trend lines are added to show the relationship of the two variables. In statistics, Pearson's correlation coefficients (R) were used to examine the correlation between the variables. When $0< R^2< 0.09$, it is regarded a small association; when $0.09< R^2 < 0.49$, it is regarded a medium correlation; and when $0.49 < R^2 < 1$, it is regarded a large association. The R value shown in Fig. 33 indicates that El Paso and New Orleans have a medium relation between their ^{90}Sr and the gross beta detected in the rivers near the monitoring sites. On the contrary, a small R value of Chattanooga indicates a poor relation between the two variables. Therefore, it is deduced the high ^{90}Sr content may play a considerable role to emit gross beta in water.

For the process to relate ^{90}Sr content and gross beta in milk, Chicago (IL), New York City (NY), Salt Lake City (UT), Honolulu (HI), Louisville (KY), Pittsburgh (PA), San Francisco (CA) and Chattanooga (TN) were adopted for a further analysis. The time period chosen is from July 1960 to December 1966.

Fig. 34 shows the relation between ^{90}Sr content from fallout and its corresponding gross beta in milk at the same location. Though Pittsburgh, Louisville and Salt Lake City showed

low or negative R value, the rest of the sites had their R values larger than 0.3, indicating that a medium correlation exists between the two variables.



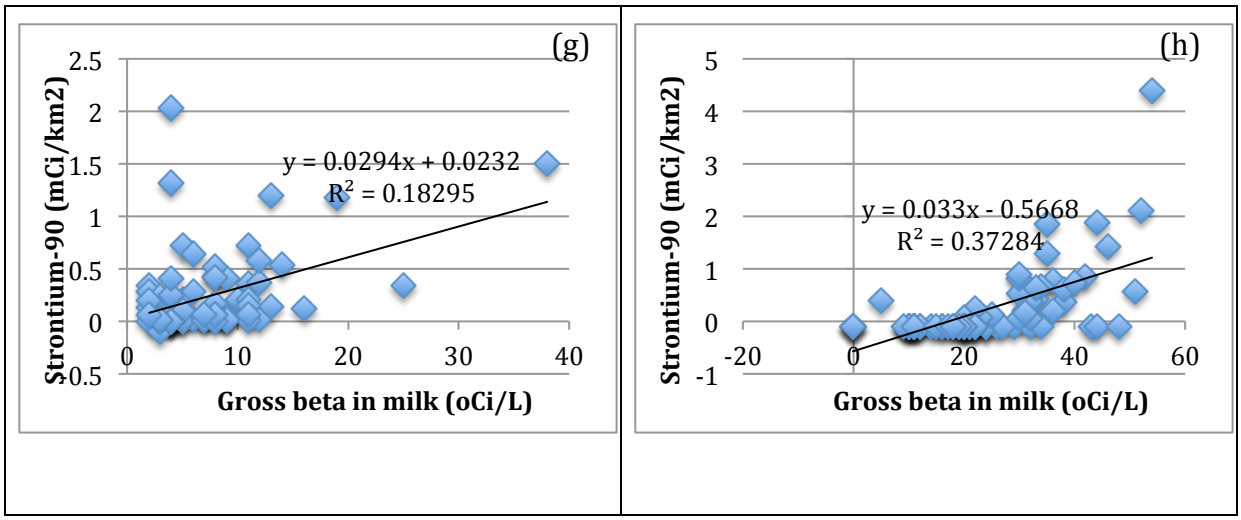


Fig. 34. Relation of gross beta in milk and ^{90}Sr from fallout in the following sites: (a) Chicago (IL), (b) New York City (NY), (c) Salt Lake City (UT), (d) Honolulu (HI), (e) Louisville (KY), (f) Pittsburgh (PA), (g) San Francisco (CA), and (h) Chattanooga (TN)

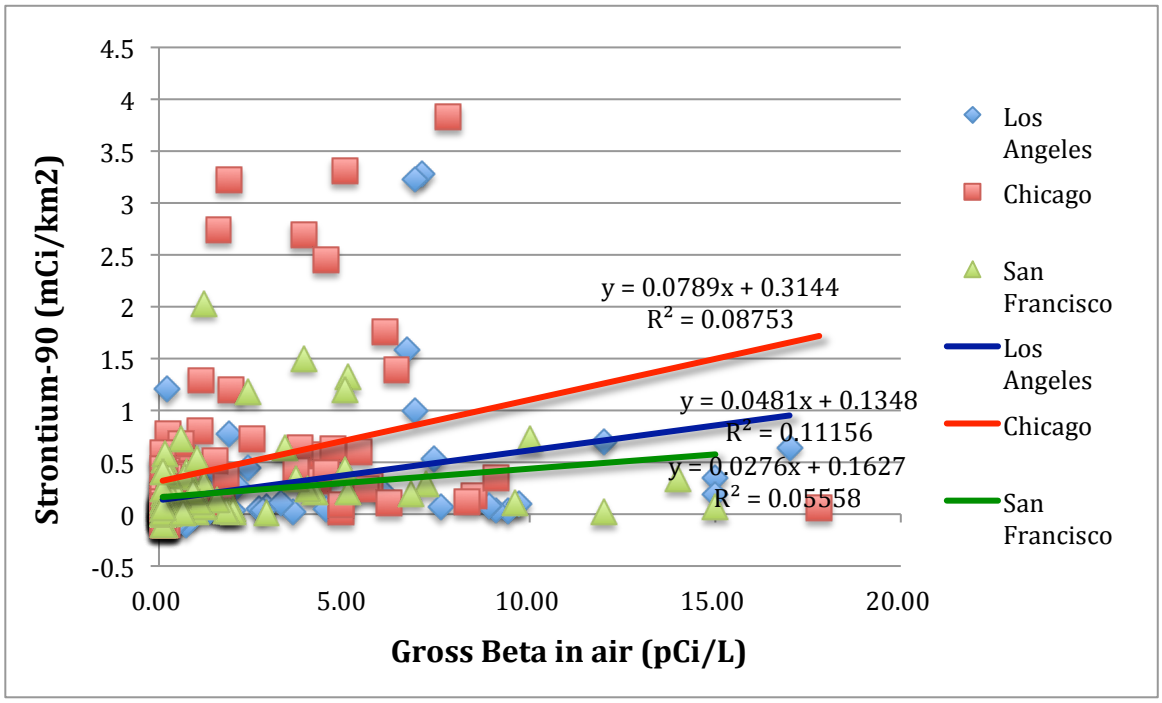


Fig. 35. Relation between gross beta in air and ^{90}Sr

The last criterion is gross beta in air. Though EML's database lacked the station of Berkeley, the data from San Francisco were used to correspond to this site due to the short distance between the both sites. Therefore, the final three sites chosen were Los angles

(referred to West Los Angles), San Francisco (referred to Berkeley) and Chicago (referred to Chicago and Springfield). From Fig. 35, the result shows a weak linkage of the beta values and ^{90}Sr content. Because of the low R value observed, it can be assumed ^{90}Sr does not contribute to gross beta in air. In this case, other radionuclides that tend not to deposit in the air may take place of ^{90}Sr.

Based on these results, it can be said that ^{90}Sr is a remarkable gross beta emitter; however, the portion of the contribution is not ascertained and should require more specialized analyses for it.

5. Conclusion

The databases provided by EML (Environmental Measurements Laboratory) and RHD (Radiological Health Data) contain observations generally from January 1957 to December 1966, and show concentrations of ^{90}Sr (strontium-90), gross beta in air, milk, and raw surface water at various monitoring stations in the U.S. There are approximately 42 sites across the US and 11 sites in US territories outside the US in EML's database. These 53 stations were extracted and analyzed for ^{90}Sr concentrations over the period of 1957 to 1966. Gross was detected over the 1959 to 1966 period and many stations. There was no clear pattern of how these radionuclides were distributed.

A comparison of bomb yield at the various test sites, and gross beta and ^{90}Sr concentrations suggest a delay between detonation and detection of approximately 1.6 years. Stratospheric residence time caused many observations of high concentrations of ^{90}Sr in 1959-1960 and 1962-1963, even though testing was not intense during these periods. Atmospheric lifting force reduces vertical downward transport in summer, leading to lower concentrations compared to other seasons.

Atmospheric deposition, an important transport mechanism, is affected by many factors as predicted by other observers (Hirose et at., 2008; McMahon, 1979). Wet deposition is the

dominant mechanism, but dry deposition should still be considered as anther common path for radionuclides to reside. The magnitude of bomb yield will determine if the fallout can reach the stratosphere to generate a global effect, or only to the troposphere, impacting the surrounding area near the test sites.

Medium correlation between ^{90}Sr and gross beta in milk and raw surface water was observed, proving that ^{90}Sr is a frequent gross beta emitter but not a primary one in the environment.

It is suggested that future studies may be able to estimate how each radionuclide contributes gross beta to the environment. Also, more quantitative ways of determining the impact of missing data on overall conclusions should be developed. A more consistent way of handling missing data is desirable.

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