THE STUDY OF NATURALLY OCCURRING RADIOACTIVE MATERIALS NORM) IN WATERS OF THE STATE OF MISSISSIPPI

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THE ORIGIN OF NORM

The Naturally Occurring Radioactive Materials originate from the following sources: (1) Cosmic or extraterrestrial; (2) Terrestrial; and (3) Technologically Enhanced Natural Radiation (TENR).

The surface of the earth is bombarded by cosmic-rays continuously. In the U.S., the average annual whole body dose equivalent from cosmic-rays is estimated to be about 45 mrem or 0.45 mSv with a range of 0.30 mSv in Hawaii and Puerto Rico to 1.30 mSv in Wyoming. In Mississippi, this value is estimated to be 0.40 mSv (Kaplan 1965; Kathern 1984).

URANIUM AND OTHER RADIOISOTOPES

Terrestrial radioactive materials are long lived radioisotopes with half-lives of the order of 109 and 1010 years and their daughter products. These time ranges are the estimated life of the planet Earth. These natural radioactive series are Uranium series (U-238), Actinium series (U-235), and Thorium series (Th-232). Figures (1a,b,c) and Tables (Ia,b,c) illustrate and tabulate the decay scheme of these series (Ref 1, 2). Uranium has three natural isotopes with an abundance of 4x10⁻⁴ percent and estimated amount of 1014 tons in the earth's crust. It is more abundant than elements such as Ag, Hg, Bi, and Cd. Thorium is three times more abundant than uranium (Ma 1983; Cochran et al. 1990). There is uranium in seawater with an estimated concentration of 0.003 ppm and about 1 ppm in marine muds (Ma 1983). These series and their daughter products, especially Ra-224, Ra-226 and Ra-228, are main contributors of NORM in terrestrial aquatic. In addition, there are other naturally occurring radioisotopes such as K-40, H-3, C-14, Rb-87, Po-210 and Rn-222 that contribute an annual dose of 0.25 mSv on average to the whole body dose. Other NORM of concern are Th-232, Th-230, and Pb-210, but because of their low solubility and difficulty of measurement, there are not many data available about them

Technologically Enhanced Natural Radiation (TENR)

Major contributors of TENR are uranium and thorium mining and milling and the mining of phosphate-based fertilizer. It is estimated that 150 million tons of phosphate rock is mined for the use of fertilizer in the US. They have relatively high concentration of radium, thorium, and uranium. Typical concentration for Th-232 is 0.5-2 pCi/g, for U-238 and Ra-226 ranges from a few pCi/g to 130 pCi/g. Potassium based fertilizers contain appreciable quantities of K-40 in the order of 10s-100s of pCi/g (Cochran et al. 1990; Kathern 1984). Oil and gas mining is another major contributor of the NORM at areas where it exists.

The South Central and Gulf Coast states are very active in oil and gas production and processing. According to the data from the Oil and Gas Board of the State of Mississippi (Smith 1995; Lewis 1995; MSOGPR 1995), there are over 27,000 oil wells that have been drilled in Mississippi so far. The 1995 data indicates that there were 4,283 active oil and gas wells in Mississippi producing 20,124,303 bbls of oil, 120,502,946 millions cubic feet of gas, and 227,208,929 bbl of water during 1994. Presently there are not offshore oil and gas activities near the coastal areas of Mississippi, but as a member of the Gulf Coast states and because of heavy oil and gas activities at offshore in the neighboring states of Louisiana and Texas, the coastal and estuaries of Mississippi may contain NORM substances via transport and mixing due to those activities. The Radiological Health Division of the Department of Health, State of Mississippi, considers radioactivity up to 2 Pico Curie Per gram (PCi/gm) in sludge and soil as the accepted background and 5-15 PCi/gm as unrestricted area and anything above that is considered restricted areas. The results of NORM studies from neighboring states (Oddo et al. 1995; Fisher 1995) indicate that the salty and fresh underground waters and sludges that are brought to the surface during oil and gas exploration can contain NORM from nil to several thousand PCi/liter or Pci/gm of sludge. These contaminants either are deposited on the site of the wells after evaporation of moisture or find their

ways into the surrounding streams. Therefore, the radiological health department believes there are 27,000 potential health hazard sites with hazardous materials in the State of Mississippi (Smith 1995; PCGB 1995; Kerr et al. 1988).

MISSISSIPPI AND NATIONAL NORM REGULATIONS

Literature surveys indicate that "NORM issues of drinking water regulations, offshore, and coastal effluent limitation guidelines are being reviewed by Environmental Protection Agency's (EPA) Office of Drinking Water" (Kerr et al; DNORMSM 1993). EPA is also concerned about NORM issues in the oil and gas industry. NORM in water is one of the aquiferic parameter and its status is desired by society and, hence, regulatory agencies. There is no standard for uranium in water supplies as a radioactive element. The NRC (Nuclear Regulatory Agency) limits are based on toxic heavy metal with chemical toxicity of 3x104 pCi/l. The NORM regulatory responsibility is not clearly established as yet (DNORMS 1993). The Department of Energy (DOE), according to some interpretation of the Atomic Energy Act, believes parts of NORM and Naturally or Accelerator produced Radioactive Materials (NARM) come under their jurisdiction. The NRC is also looking into NORM and NARM issues. The oil and gas industry is very interested in NORM issues because of its effects on their operations and economics (PCGB 1995).

The water content of NORM has been used to provide data in geology and geomorphology of the region for selection or omission of all kinds of mineral activities and underground water migration studies. NORM data around the oil and gas fields are also of interest to agriculture and fisheries both inland and at coastal areas. The same issues are of interest to the state of Mississippi whose rules and regulations in this area closely follow the federal agencies and sometimes supplement them.

NORM IN WATERS OF THE STATE OF MISSISSIPPI

Out of eleven geological regions in the continental US, the Gulf Coast and the State of Mississippi come under coastal plain. The nature of ground-water flow systems is "regional flow in sand and limestone aquifers with intervening clay confining layers; predominant flow direction seaward; discharge upward through confining layers and to stream." There is a hypothesis that indicates that certain regions or sub-regions only produce ground waters with a specific radionuclide, or conversely, without a specific nuclide (Hess et al 1985; Reid et al. 1985; Cross et al. 1985). Figure 2 shows the geological provinces of the United States.

The population averaged uranium concentration for domestic surface and ground waters of Mississippi is estimated at 0.075 picocuries per liter (pCi/l) Figures (3a,b,c) (Hess et al 1985; Reid et al. 1985).

Norm in Drinking Waters

Available data indicates that out of about 60,000 public water supplies in the United State, 80% use underground water sources. This percentage is much higher for the state of Mississippi. Only three or four out of about 400 communities draw their drinking waters from surface waters; underground water supplies are the source of the water for the rest of the state population. The EPA's Maximum Contaminant Level (MCL) of combined Ra-226 and Ra-228 sets a limit of 5pCi/l with a gross alphaparticle activity of 15 pCi/l excluding radon and uranium. All states except (IL, CO, MT, and OR) have reported the MCL violations. Figure 3 illustrates approximate locations and general areas where public water supplies exceeded EPA's MCL guideline.

Radon isotopes are the daughter products of isotopes of Radium. Rn-220 is the progeny of Ra-224 and has a halflife of 56 sec, and Rn-222 is the progeny of Ra-226 with a half-life of 3.84 days. Waters containing Rn contributes to indoor radon. There are no standards for Rn in water. In Mississippi, the geometrically averaged Rn concentration for ground-water is 23 pCi/l and 260 pCi/l for combined ground and surface waters. (Hess et al. 1985; Reid et al. 1985; and Cross et al. 1985). Figure 5 illustrates geometrically averaged Rn concentration in pCi/l of ground water that is 23 for the state of Mississippi.

TERRESTRIAL TRANSPORT OF NORM IN WATERS

Transport of NORM in coastal and estuaries can follow Fick's law and diffusion theory. In one dimensional approximation where the maximum concentration along the axis of current at distance x is concerned, the formula is given by

$$C_m = \frac{Q}{2D(2\pi K U x)^{0.5}}$$

Where Q is the released activity per unit time, D is the depth, K is the diffusion coefficient, typically about

 $6 \times 10^{-3} \text{ m}^2/\text{min}$, U is the velocity of the current and C_m maximum concentration along the axis.

For estuaries, assuming semi-closed coastal waters, the time dependent diffusion equation can be written as follows:

$$\frac{\partial C}{\partial t} + u_f \frac{\partial C}{\partial x} = E_L \frac{\partial^2 C}{\partial x^2} - \lambda C$$

that can be solved for instantaneous release of activity and looks like

$$C = \frac{Q}{A\sqrt{4\pi E_{L}t}} \exp \left[\frac{(x - u_{f}t)^{2}}{4E_{L}t} + \lambda t\right]$$

where A is estuary cross-section at distance x, at time t, E_L is the longitudinal diffusion coefficient, Q is the quantity of the released radioactivity, is the decay constant of radionuclide and U_f is the net down-stream freshwater velocity.

For rivers, streams, and lakes the classical hydraulic principles for open channel yields a general solution of the following form

$$C_x = \frac{C_d F_d + Q_x}{yzu + F_d + D - E} e^{-kx}$$

Where C_x is the concentration at distance x at downstream, C_d is the discharge concentration, F_d is discharge of radioactive containing water in m³/sec, Q_x is the discharge rate in Ci/m³, y and z are the channel width and depth, u is the mean velocity in the channel, D is the dilution ratew, E is the evaporative loss and k is the removal coefficient.

For subsurface seepage, the Darcy equation gives the following formula

 $Q = ui\Lambda$

where Q is the rate of seepage, u the permeability of the soil medium, I the hydraulic gradient, A the area, K the coefficient of removal, d depth in the soil, decay constant and t time after administration (Cross et al. 1985). This formula is a generalized somehow ideal model for computing downstream concentration.

CONCLUSION

NORM activities, especially the oil and gas related part, are at their early stages in the state of Mississippi. Lack of public awareness has been the cause of unfortunate incidents in the past. Similar studies in neighboring states and studies in general indicate that the radioactivities of the water and mixed water and minerals could be nil to several thousands of Pico Curie (pCi) per liter of water or gram of scale on the pipes or sludges that resulted from drilling (Greer et al. 1995; Oddo et al.; Fisher 1995; Smith et al. 1995; and Fisher 1994).

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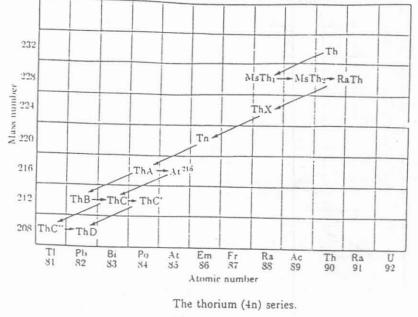


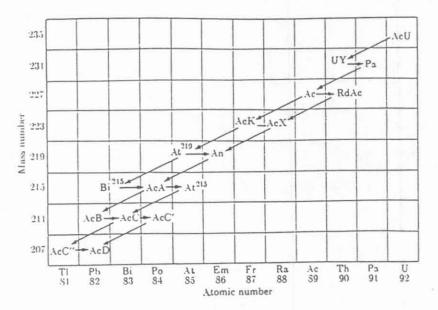
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| HE | THORIUM | DERIES |

| Radioactive species | Nuclide | Type of disinte- gration | Half-life | Disinte- gration constant, sec ⁻¹ | Particle energy, Mev |
|-----------------------------------|----------|--------------------------------|---------------------------|---|----------------------------|
| Thorium (Th) | 90Th232 | α | 1.39 × 10 ¹⁰ y | 1.58×10^{-18} | 4.007 |
| Mesothorium1(MsTh1) | 88Ra228 | β | 6.7 y | 3.28×10^{-9} | 0.04 |
| Mesothorium2(MsTh2) | \$2Ac228 | β | 6.13 h | 3.14×10^{-5} | 2.18 |
| Radiothorium (RdTh) | 20Th228 | α | 1.910 y | 1.15×10^{-5} | 5.423 m |
| Thorium X (ThX) | 38Ra224 | α | 3.64 d | 2.20×10^{-5} | 5.681 m |
| Th Emanation (Tn) | 35Em220 | α | 51.5 s | 1.34×10^{-2} | 6.280 |
| Thorium A (ThA) | 34Po216 | α, β | 0.16 s | 4.33 | 6.774 |
| Thorium B (ThB) | 32Pb212 | ß | 10.6 h | 1.82×10^{-5} | 0.58 |
| Astatine-216 (At ²¹⁶) | 35At218 | α | 3×10^{-4} s | 2.3×10^{3} | 7.79 |
| Thorium C (ThC) | 33Bi212 | α, β | 60.5 m | 1.91×10^{-4} | α:6.086 m β:2.25 |
| Thorium C' (ThC') | \$4Po212 | α | $3.0 \times 10^{-7} s$ | 2.31 × 10 ⁶ | 8.780 |
| Thorium C" (ThC") | 81 Tl208 | β | 3.10 m | 3.73×10^{-3} | 1.79 |
| Thorium D (ThD) | 82Pb208 | Stable | | | |

TableIa

THE NATURAL RADIOACTIVE SERIES

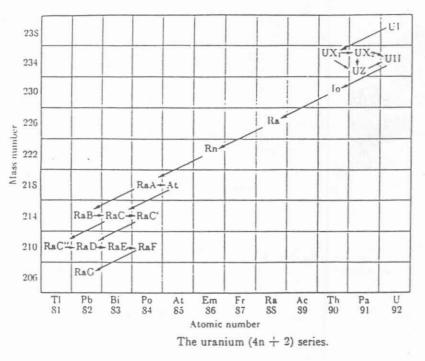


The actinium (4n + 3) series. Fig 1b The Actinium Series

| Radioactive species | Nuclide | Type of disinte- gration | Half-life | Disinte- gration constant, sec ⁻¹ | Particle energy, Mev |
|------------------------|----------------------|--------------------------------|--------------------------------|---|----------------------------|
| Actinouranium (AcU) | 82U235 | α | 7.10 × 10 ⁸ y | 3.09×10^{-17} | 4.559 m |
| Uranium Y (UY) | 20Th231 | B | 25.6 h | 7.51 × 10 ⁻⁶ | 0.30 |
| Protoactinium (Pa) | 91 Pa231 | α | $3.43 \times 10^{4} \text{ y}$ | 6.40×10^{-13} | 5.046 m |
| Actinium (Ac) | \$9Ac ²²⁷ | α, β | 21.6 y | 1.02×10^{-9} | a:4.94 \$:0.046 |
| Radioactinium(RdAc) | 90Th227 | α | 18.17 d | 4.41×10^{-7} | 6.03 m |
| Actinium K (AcK) | 87Fr223 | α, β | 22 m | 5.25 × 10 ⁻⁴ | β:1.2 ·a:5.34 |
| Actinium X (AcX) | 88Ra 2:3 | α | 11.68 d | 6.87×10^{-7} | 5.S64 |
| Astatine-219 | \$5.At219 | α, β | 0.9 m | 1.26×10^{-2} | a:6.27 |
| Ac Emanation (An) | 86Em219 | α | 3.92 s | 0.177 | 6.810 m |
| Bismuth-215 | 83Bi215 | α, β | 8 m | 1.44×10^{-3} | ? |
| Actinium A (AcA) | 5+Po215 | α, β | $1.83 \times 10^{-3} s$ | 3.79×10^{2} | a:7.37 |
| Actinium B (AcB) | 82Pb211 | β | 36.1 m | 3.20×10^{-4} | 1.39 |
| Astatine-215 | 85At215 | α | 10-4 s | 7×10^{3} | 8.00 |
| Actinium C (AcC) | 83Bi211 | α, β | 2.15 m | 5.28×10^{-3} | a:6.617 m |
| Actinium C' (AcC') | #«Po ²¹¹ | α | 0.52 s | 1.33 | 7.442 m |
| Actinium C'' (AcC'') | 81 Tl207 | β | 4.79 m | 2.41×10^{-3} | 1.44 |
| Actinium D (AcD) | 82Pb207 | Stable | | | |

Table Ib





| Fig | 10 |
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| rig | IC. |

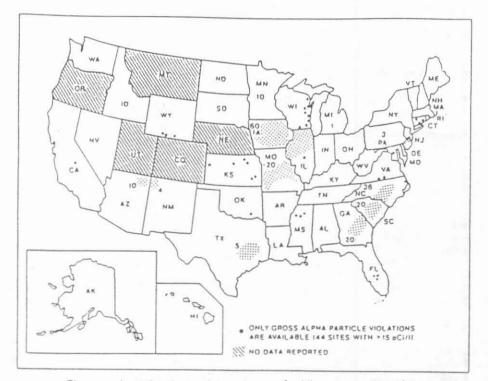
| THE | URANIUM | Series |
|-----|---------|--------|
|-----|---------|--------|

| Radioactive species | Nuclide | Type of disinte- gration | Half-life | Disinte- gration constant, sec ⁻¹ | Particle energy, Mev |
|---|--------------------|--------------------------------|--------------------------------|---|----------------------------|
| Granium I (UI) | 02U=38 | α | $4.50 \times 10^{9} \text{ y}$ | 4.88 × 10 ⁻¹⁸ | 4.20 |
| Uranium X1 (UX1) | 90Th234 | β | 24.1 d | 3.33 × 10-: | 0.19 |
| Uranium X2 (UX2) | 91 P8.234 | β | 1.18 m | 9.77 × 10-3 | 2.32 |
| Uranium Z (UZ) | 91 Pa 234 | β | 6.7 h | 2.88×10^{-5} | 1.13 |
| Uranium II (UII) | 02U234 | α | $2.50 \times 10^{s} y$ | 8.80×10^{-14} | 4.768 |
| Ionium (Io) | 00Th230 | α | $8.0 \times 10^4 \text{ y}$ | 2.75×10^{-13} | 4.68 m |
| Radium (Ra) | 88Ra 226 | α | 1620 y | 1.36×10^{-11} | 4.777 m |
| Ra Emanation (Rn) | 86Em222 | α | 3.82 d | 2.10 × 10-6 | 5.486 |
| Radium A (RaA) | \$4Po218 | α, β | 3.05 m | 3.78×10^{-3} | a:5.998 8:? |
| Radium B (RaB) | 82Pb214 | β | 26.8 m | 4.31×10^{-4} | 0.7 |
| Astatine-218 (At ²¹⁸) | 85At218 | α | 1.5-2.0 s | 0.4 | 6.63 |
| Radium C (RaC) | 83Bi214 | α, β | 19.7 m | 5.86×10^{-4} | α:5.51 m β:3.17 |
| Radium C' (RaC') | 84Po214 | α | $1.64 \times 10^{-4} s$ | 4.23×10^{3} | 7.683 |
| Radium C'' (RaC'') | 81T]210 | β | 1.32 m | 8.75×10^{-4} | 1.9 |
| Radium D (RaD) | 82Pb210 | β | 19.4 y | 1.13×10^{-9} | 0.017 |
| Radium E (RaE) | *3Bi210 | β | 5.0 d | 1.60×10^{-6} | 1.155 |
| Radium F (RaF) | 8+Po210 | α | 138.3 d | 5.80×10^{-8} | 5.300 |
| Thallium-206 (Tl ²⁰⁶) Radium G (RaG) | 81Tl206 82Pb206 | β Stable | 4.2 m | 2.75 × 10 ⁻³ | 1.51 |

Table Ic.



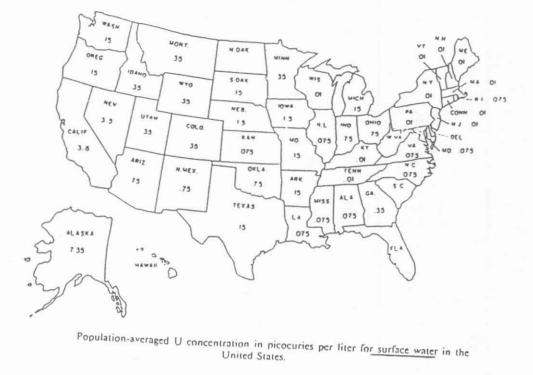


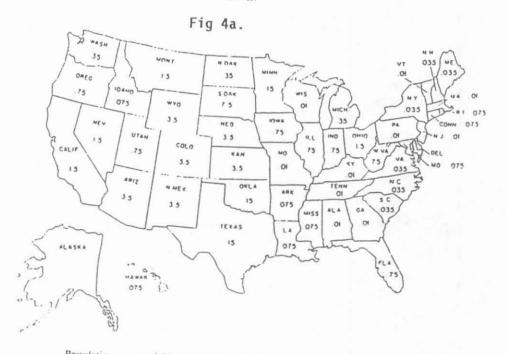


The approximate locations and general areas of public water supplies which exceed 5 pCi/l of total Ra (²²⁸Ra was reported or combined with ²²⁸Ra for about one-half of the sites). Large dots represent individual violations. The dot pattern represents the general area of a group of violations, with the adjacent number indicating the number of violations in that group. When the locations were unknown, just the number of violations was indicated (modified after Cothern and Lappenbusch

Fig 3







Population-averaged U concentration in picocuries per liter for ground water in the United States.

Fig 4b





Population-averaged U concentration in picocuries per liter for domestic water in the United States.

Fig 4c:



Geometric average Rn concentration in picocuries per liter for public ground-water supplies in the United States.

Fiq 5.



Table II.

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1994 ANNUAL PRODUCTION BY COUNTY

| COUNTY NAME | NUNBER OF PRODUCING HELLS | OIL - BBLS PRODUCTION | HATER - BELS PRODUCTION | GAS - MCF PRODUCTION |
|----------------------|------------------------------|---------------------------------------|--|-----------------------------------|
| ADANS | 452 | 1,149,932 491.750 1,620 | 31.376,142 3,143,407 1,217 0 37,360,582 1,751 | 203,727 |
| AMITE | 117 | 491.750 | 3,183,607 | 3,155,945 |
| CHICKASAM | 28 | 1,620 | 1,217 | 921.743 |
| CLAINBORNE CLARKE | | 0 | | 266.620 727.923 768.666 |
| CLAY | 283 | 2,242,759 | 37,390,382 | 121,723 |
| COPIAH | 1 | 398 | 441 | 27 241 |
| COVINGTON | 24 | 331,022 | 791,682 | 2,021,659 |
| FORREST | 15 | 331,022 4,703 501,809 | 451 791,602 39,279 19,001,455 2,196,765 | 27,261 2.021.659 132,191 |
| FRANKLIN | 170 | 501,809 | 19,001,853 | 88,153 1,453,180 |
| GREENE | 38 | 208,482 21,906 | 2,196,765 50,519 | 1,653,180 |
| HINDS | 52 | 282 102 | 1,284,434 | 1,791,933 2,174,698 |
| HOLNES | 12 | 282,102 | 1,412 | 18,657 |
| HUMPHREYS | -1 | 0 | 0 | 0 |
| ISSAQUENA | ī | ō | 0 | ō |
| I TAHAMBA | 6 | 0 | 9 | 47,122 2,227,158 |
| JASPER | 452 | 3,646,334 130,787 211,019 | 35,251,026 2,776,450 124,417 | 2,227,158 |
| JEFFERSH DAVIS | 54 98 | 130,787 | 2,776,490 | 283,496 |
| JONES | 292 | 1 044 945 | 14 176 179 | 12,651,470 933,355 |
| LAMAR | 298 | 1.009.508 | 14,176,179 20,527,748 | 2,160,184 |
| LAHRENCE | 18 | 3,044.965 1,009,508 12,978 | 13,921 | 1,698,975 |
| LEE | 18 | 0 | 0 | 2,160,184 1,698,975 697,249 |
| LEFLORE | | 0 | 0 | 0 |
| LOHNDES | 219 | 664.169 | 1,466,670 3,370 | 73,139 |
| WADISON | 21 | 12.(13 | 2 748 595 | 73,139 #13,265 18,308 |
| HADISON MARION | 67 55 102 174 | 15,745 85,110 207,655 37,822 | 2,368,595 1,705,413 | 9,532,490 |
| MONROE | 174 | 37.622 | 46.644 | 5.474.791 |
| OKTIBBEHA | 1 | 0 | 0 | 9,532,490 5,474,791 47,532 |
| PEARL RIVER PERRY | 20 25 96 12 65 | 13,292 163,904 339,774 | 126,192 505,053 1,381,736 383 | 3,422,494 |
| PERRY | 35 | 163.994 | 505.053 | 103,601 |
| PONTOTOC | 12 | 337.774 | 1,301,730 | 103,601 |
| RANKIN | 65 | 168,512 | 1,403,569 | 508.467 45.387.719 |
| SCOTT | 12 | 168,512 105,744 353,322 | 1,403,569 438,523 | 66.019 |
| SIMPSON | | 363, 322 | 1.397.621 | 3.423.495 3.318.231 73.274 |
| SHITH | 126 | 490, 479 | 2,249,592 | 3.318.231 |
| STONE | 1 | FD 457 | (01 26) | 73.274 |
| HALTHALL HARREN | 48 | 50.457 21.861 | 14 064 | 3.566.118 2.162,958 |
| MAYNE | 339 | 2.651.914 | 604,261 15,064 23,156,893 | \$55.987 |
| HILKINSON | 200 | 613,051 847,004 | 6,584.373 | 6.617.951 |
| YAZOO | 188 | 847,004 | 15,595,312 | 354.009 |
| TOTAL PRODUCTION | 4,283 | 20,124,303 | 227, 208, 929 | 120,502,946 |

Radim-in-water results by state and population. All results are geometric means in units of pCisl. Purcurberes values are in numbers of samples

| State < 100 100-1000 At 83 (10)* At 22 (6) At 240 (2) | | | | |
|--|-----------|-------------|----------|----------|
| 83 (10) 22 (6) 230 (2) | 1000-5000 | 5000-10,000 | > 10,000 | Unknown |
| 240 (2 | 59 (76 | 82 (46) | 8 (4 | - |
| 240 (2 | 6 (3 | 1 | 2 | 8 (13 |
| | 200 (68) | 350 (22) | 340 (30) | 160 (2) |
| | 1 | ł | 1 | 70 (15 |
| 130 (6 | 20 (5 | 0 | 0 | |
| 1 | 9 (4 | 1 | 23 (12 | ; |
| 320 (2 |) 06 | 49 (7 | 4(243 | 1 |
| 57 (4) 42 (12 | 30 (5 | 0 (2 | 2 (45 | 39 (81) |
| 1120 (13 | 30 (3 | 50 (2 | 66 (10 | 5 (2 |
| 6 (3) 210 (14 | 30 (8 | 6 (2 | 0 (30 | ; |
| 71 (1 | 81 (3 | 6) 0 | 10(185 | * |
| : | 5 (7 | 7 (5 | 0 (56 | 1 |
| 260 (2) 230 (1 | 3 (1 | 20 (4 | 8 (8 | 210(10) |
| 20 (10 | 2 (7 | 36 (8 | 5 (10 | 1 |
| 3300 (2) | 80 (4 | 540 (67 | 10 (88 | 8)08 |
| 670 (3) 1600 (23 | 90 (3 | 00 (7 | 4) 0 | 1)00 |
| : | 0 (7 | 68 (43 | AO (90 | 330(2) |
| ND (2) 58 (54 | 26 (5 | 00 (4 | 8 (6 | ND(18 |
| : | 15 (4 | 1 (26 | 3 (33 | |
| 740 (4) 280 (6 | •) 0 | 60 (8 | 8 (4 | ; |
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table III.