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Department of Energy's Historic Nuclear Bomb Manufacturing
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March 2007

Prepared for Miamisburg Environmental Safety & Health
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By Hydro-Log LLC

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and Technical Assessment Fund.*

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Tritium in the Environment

Tritium is a radioactive variety of hydrogen. For a number of years a large amount of radioactive tritium gas was used in the assembly of nuclear bombs at the U.S. Department of Energy's Mound Facility in Miamisburg, Ohio.⁽¹⁾ The Mound Facility manufacturing process that used radioactive tritium allowed substantial amounts of radioactive tritium gas to escape into the atmosphere through a tritium emission stack specifically designed to look like a smoke stack. According to the US EPA, tritium gas released into the atmosphere readily combines with oxygen to form radioactive (tritiated) water.⁽²⁾

Radioactive (tritiated) water in the Environment

Radioactive (tritiated) water is generally indistinguishable from normal water and is transported throughout the environment in the same manner as water. After being emitted from the Mound Facility tritium stack and combining with oxygen, the humidity in the air around the Mound Facility became slightly radioactive. Depending on wind direction at any given time the radioactive (tritiated) humidity blew downwind, away from Mound. Thus people living in the greater Mound region were breathing air containing radioactive humidity. Rain, fog, dew, and snowfall moved radioactive (tritiated) water from the atmosphere to the ground. Evaporation of radioactive (tritiated) water from the ground transported radioactive (tritiated) water back into the atmosphere. In soil, radioactive (tritiated) water was taken up by plants (including food plants). Some radioactive (tritiated) water moved downward through the soil and into underlying ground water. When tritium decays radioactively, it is transformed to helium with a half-life of about 12.3 years. Intake of radioactive (tritiated) water by humans and other animals mostly occurs through breathing and skin contact, although drinking local water and eating locally grown plants would also have been a source of some radioactive tritium contamination for people living in the greater Mound region. Monitoring data indicate that the general population may be exposed to tritium via breathing, eating, drinking, and contact of skin with tritium in rainfall and in the air.⁽³⁾

Radioactive (tritiated) water in humans

A March, 2006 report on Tritium by the California EPA Office of Health Hazard Assessment reports that tritium is readily taken into the body, presenting an internal radiation hazard.⁽⁴⁾ The Kirk-Othmer Encyclopedia of Chemical Technology reports that people absorb 98-99% of radioactive (tritiated) humidity breathed into the lungs. After breathing in air containing humidity with

radioactive (tritiated) water, uniform distribution of the radioactivity throughout body fluids occurs within 90 min. Also, when a person is exposed to air that contains radioactive (tritiated) water vapor, the water that enters the body through the total skin area will approximately equal that entering through the lungs.⁽⁵⁾

A 2001 report by the World Health Organization (WHO) describes radioactive (tritiated) water vapor as rapidly absorbed from the lungs into the blood. Volunteers who drank radioactive (tritiated) water rapidly and virtually completely absorbed the water from their gastrointestinal tract into the rest of their body.⁽⁶⁾

The WHO International Programme on Chemical Safety reports that radioactive (tritiated) water in blood equilibrates with other body fluids in about 12 minutes. However, in tissues that do not contain very many blood vessels, such as bone and fat, equilibrium of radioactive (tritiated) water in the blood with other water may take days to weeks. According to WHO, when radioactive (tritiated) water is ingested in one single dose, about one-half of the radioactive (tritiated) water is excreted from the body in three separate periods: about 10 days, one month, and one year. The first component is believed to reflect the normal turnover of body water from normal water ingestion and consumption, while the second and the third components are thought to represent the turnover of radioactive (tritiated) water incorporated into organic compounds within the body.⁽⁷⁾

The WHO, in another report, states that since water in the body is distributed fairly uniformly, it is generally assumed that all organs and tissues receive the same radiation dose from radioactive (tritiated) water. WHO states that the half-times of retention in the body of radioactive tritium that has become bound within the body's organic molecules have not been well characterized in humans, but the range appears to be 20-80 days for most of the radioactive tritium and 280-550 days for a smaller component. It has been estimated that total incorporation of radioactive tritium into organic molecules within the body adds little (< 10%) to the total radioactive dose from exposure to radioactive (tritiated) water.⁽⁸⁾ The same WHO report states that radioactive (tritiated) water crosses the placenta rapidly and equilibrates between maternal and fetal tissues.⁽⁹⁾ Mettler et. al. state that since the fetus has a higher water concentration than an adult, the radiation dose to the fetus is about 40 to 70% higher than the dose to the mother.⁽¹⁰⁾

According to the California EPA, a two-compartment model describes the whole-body retention of radioactive tritium taken in by adults and children as radioactive (tritiated) water. The first compartment is normal body water, which contains 97% of the total tritium. The second compartment represents radioactive tritium that is incorporated into molecules within the body. This compartment represents 3% of the total tritium.⁽¹¹⁾ Following absorption into the blood, the 3% of the tritium that becomes bound in organic molecules is incorporated into body tissues to an extent that depends on the metabolic activity of the individual tissue examined, and on the specific chemical that is involved.⁽¹²⁾

In the United States it used to be common to report levels of radiation in the environment in Curies. One Curie is defined as 37 billion radioactive decays/second, a huge number. Since a Curie is such a large number, environmental radiation standards are often set in one-trillionth of a Curie, which is known as a picoCurie, and abbreviated as pCi. Radioactive decay of tritium produces what is known as ionizing radiation. According to the National Institutes of Health, Hazardous Substances Data Bank, Ionizing radiation has carcinogenic effects in many tissues. A dose-response relationship exists between exposure to ionizing radiation and the risk for the subsequent development of cancer. This means the more radiation a person is exposed to, the greater their chances of developing cancer.⁽¹³⁾ Ionizing radiation is genotoxic and causes breaks in the structure of DNA, resulting in mutations or chromosomal structural aberrations. Double DNA strand breaks that result in mutagenic and carcinogenic effects have been reported. Incorrectly rejoined DNA after broken DNA rejoins leads to DNA deletions and rearrangements. Large scale changes in DNA structure appear to be typical of most radiation-induced mutations.⁽¹³⁾

In the recent past a large number of radiobiological studies of tritium have become available. Many of the recent studies focus on the relative biological effectiveness of radiation produced during the radioactive decay of tritium in the body. These recent studies and previous studies indicate that tritium in body water and incorporated within body molecules produces the same spectrum of radiogenic effects (e.g., cancer, genetic effects, developmental abnormalities, and reproductive effects) that are observed following whole-body exposure to penetrating radiations such as gamma rays and x rays. However radiation from radioactive decay of tritium within the body is of greater biological effectiveness than gamma rays and x rays. For example, radioactive tritium in water within the body is about 2 to 3 times more dangerous at low doses or low dose rates than gamma rays from several sources commonly used in radioactive treatment of cancer patients. When tritium is bound to molecules within the body, relative biological effectiveness values may be somewhat larger than those for tritium in body water. It is clear from the wealth of tritium data now available that relative biological effectiveness values for tritium radioactive decay are higher than the quality factor of one (unity) generally used in radiation protection calculations.⁽¹⁴⁾

Most recent studies of tritium have been conducted on laboratory animals. It is generally considered that effects frequently seen in laboratory animals exposed to relatively high doses may be seen at a much lower frequency in humans if humans are exposed to the same materials at much lower doses. Diseases identified in animals dosed with radioactive tritiated water include myeloid leukemia,⁽¹⁵⁾ decreased weight of brain and genital tract organs,⁽¹⁶⁾ impaired central nervous system development,⁽¹⁷⁾ difficulties in both learning and memory retention,⁽¹⁸⁾ microcephaly,⁽¹⁹⁾ reductions in female egg counts and reduced testis weight,⁽²⁰⁾ tumor development, especially uncommon tumors including lung

and liver tumors, and leukemias, increased with increasing tritium exposure levels.⁽²¹⁾ A study of chromosomes showed that chromosome breaks at higher dose range increased linearly with dose, while those at lower dose range were significantly higher than would be expected by a straight line downward extrapolation.⁽²²⁾

Relative risk to the public living around Mound Facility as compared to workers inside the Mound Facility

All air contains water as humidity. If air humidity contains radioactive (tritiated) water, humans readily adsorb radiation through the skin, and by breathing. Exposure to radiation through tritiated water in air is 25,000 times more hazardous than exposure to gaseous tritium.⁽²³⁾ Unlike radioactive (tritiated) water vapor, tritium gas in the air is not particularly dangerous. The US Department of Energy reports that the body does not readily absorb tritium gas from inhalation or through the skin. If inhaled in elemental gaseous form almost all tritium gas is exhaled. Only a very small fraction of tritium gas is retained in the lungs.⁽²⁴⁾ Thus, strangely enough, workers within the Mound Facility who were sometimes exposed to radioactive tritium gas, may have been exposed to lower risk due to tritium radiation than the public living in the area around the Mound Facility. When the Mound Facility was emitting radioactive tritium gas from their tritium stack, after leaving the stack the tritium gas combined with oxygen to make radioactive (tritiated) water. Because it is so much more easily absorbed, the radioactive (tritiated) humidity in the air within the greater Mound region was 25,000 times more hazardous per unit of radiation to the people living around the Mound Facility than was pure tritium gas inside the facility.

Radioactive monitoring in the Greater Mound Region

Mound Facility measured the amount of tritium released from the tritium stack, and also the amount of tritium in the moisture in the air in the greater Mound region. The air monitoring stations were small wooden structures somewhat like a large birdhouse with louvered sides. Air was drawn into a sampling device that was located inside the sampling "house".⁽²⁵⁾ Since a sampling device located under a roof does not sample radioactive rain or dew, reported values for tritium in the greater Miamisburg area are less, perhaps much less, than actual exposure to tritium for people in the area if they were breathing fog-laden air on a crisp fall morning, if they were out in the open on an evening when a heavy dew was falling, if they were walking across dew laden grass on a sunny morning, or if they were out in the rain.

History of Radioactive Emissions from Mound Facility

Table 1 shows documented releases of radioactive materials from Mound Facility, 1959-1989, into the surrounding air and water. Table 1 shows both radioactive tritium gas (H-3) and Plutonium 238, the variety of Plutonium handled

at Mound Facility. Plutonium 238 is measured in 1/1,000,000 of a Curie, a unit known as a microCurie, and abbreviated μCi .⁽²⁶⁾

Tritium gas emissions from the tritium stack at Mound are shown in the column labeled H-3 Air (Ci). Considering that one Curie represents 37 billion radioactive decays per second, the numbers in the tritium (H-3) air column are impressive indeed. From 1960 through 1970, annual tritium gas releases exceeded 100,000 Curies, with a peak year (1967) of about 365 Curies, or one whole Curie each day for the entire year.

Table II.1. Summary of Mound Plant Effluents 1959-1989

Year	Pu-238 Air (μ Ci)	Pu-238 Water (μ Ci)	H-3 Air (Ci)	H-3 Water (Ci)
1959			31,527	
1960	250,125		102,427	
1961	160		240,644	
1962	140		244,455	
1963	108		313,932	
1964	252		262,638	
1965	5,803		206,750	
1966	30,442		199,561	
1967	54,347	24,900	364,685	169
1968	5,720	243,800	275,856	202
1969	10,544	109,700	315,252	2,332
1970	4,342	7,420	179,468	250
1971	401	15,234	73,503	399
1972	74	60,586	30,483	244
1973	84	16,043	15,331	149
1974	28	19,755	10,031	105
1975	23	17,862	8,859	58
1976	15	2,973	6,206	46
1977	12	3,584	4,896	57
1978	14	4,947	7,346	32
1979	12	3,157	3,831	34
1980	15	773	3,795	26
1981	8	1,110	4,285	22
1982	21	1,207	4,283	14
1983	4	1,003	4,293	8
1984	7	1,342	3,430	8
1985	5	991	4,795	6
1986	6	691	3,555	6
1987	5	472	3,863	6
1988	5	997	3,204	4
1989	4	1,419	41,534	6

Xeroxed copies of the original lab record sheets for this study were obtained from DOE – Office of Legacy Management (OLM)-Mound Site. The copied records then had to be hand entered into a database created by Hydro-Log LLC. DOE-OLM-Mound Site is still searching for the manual describing the details of the laboratory analysis. Without this manual, and its description of the specific equation used to extrapolate from concentration of tritium oxide in sample solution to the concentration of tritium oxide in the air it is not possible to convert the original data into air concentrations.

Appendix I contains Figures 1 through 12. Figure 1 and Figure 7 show the physical locations of air monitoring stations permanently placed in surrounding communities by the Mound Facility. Figures 1 through 6 show air monitoring stations that were placed within approximately 1 mile of the Mound Facility. Figures 7 through 12 show air monitoring stations that were placed greater than one mile from the Mound Facility. Figures 2 through 6 show the average concentration of tritium oxide in sample collection solutions for specific date ranges for samples collected within approximately 1 mile of the Mound Facility. Figures 8 through 12 show the average concentration of tritium oxide in sample collection solutions for specific date ranges for samples collected from more than 1 mile from the Mound Facility.

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Appendix I

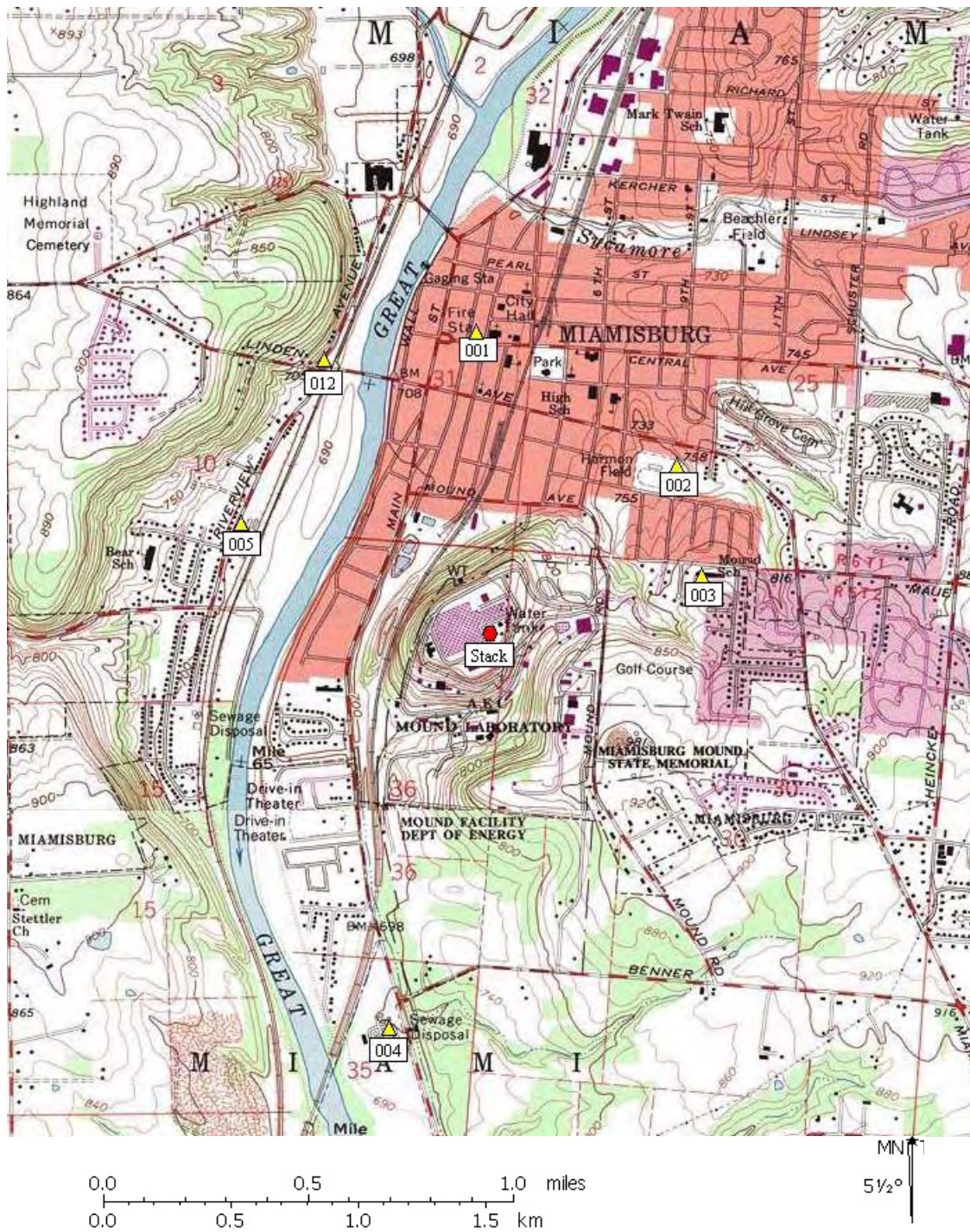
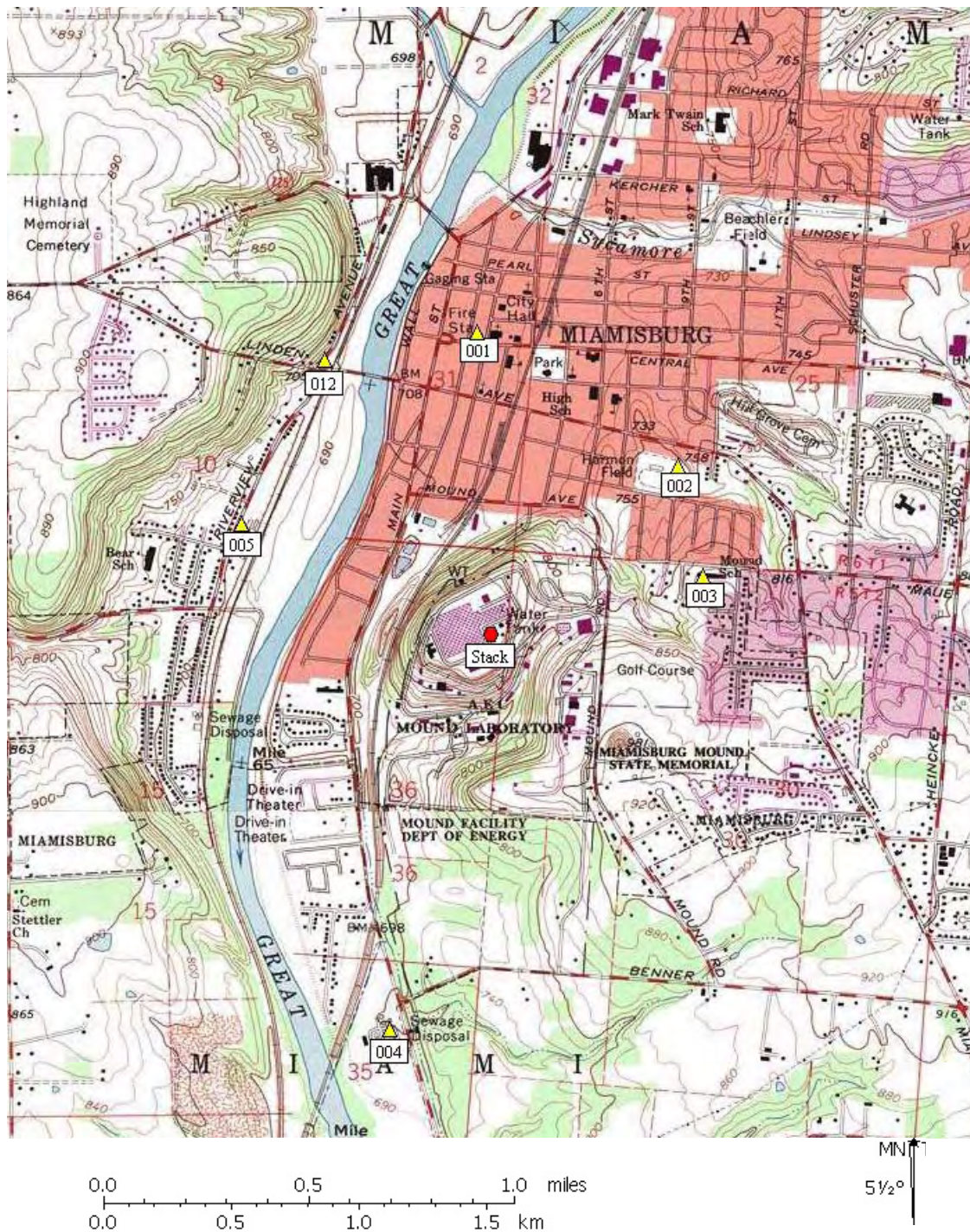
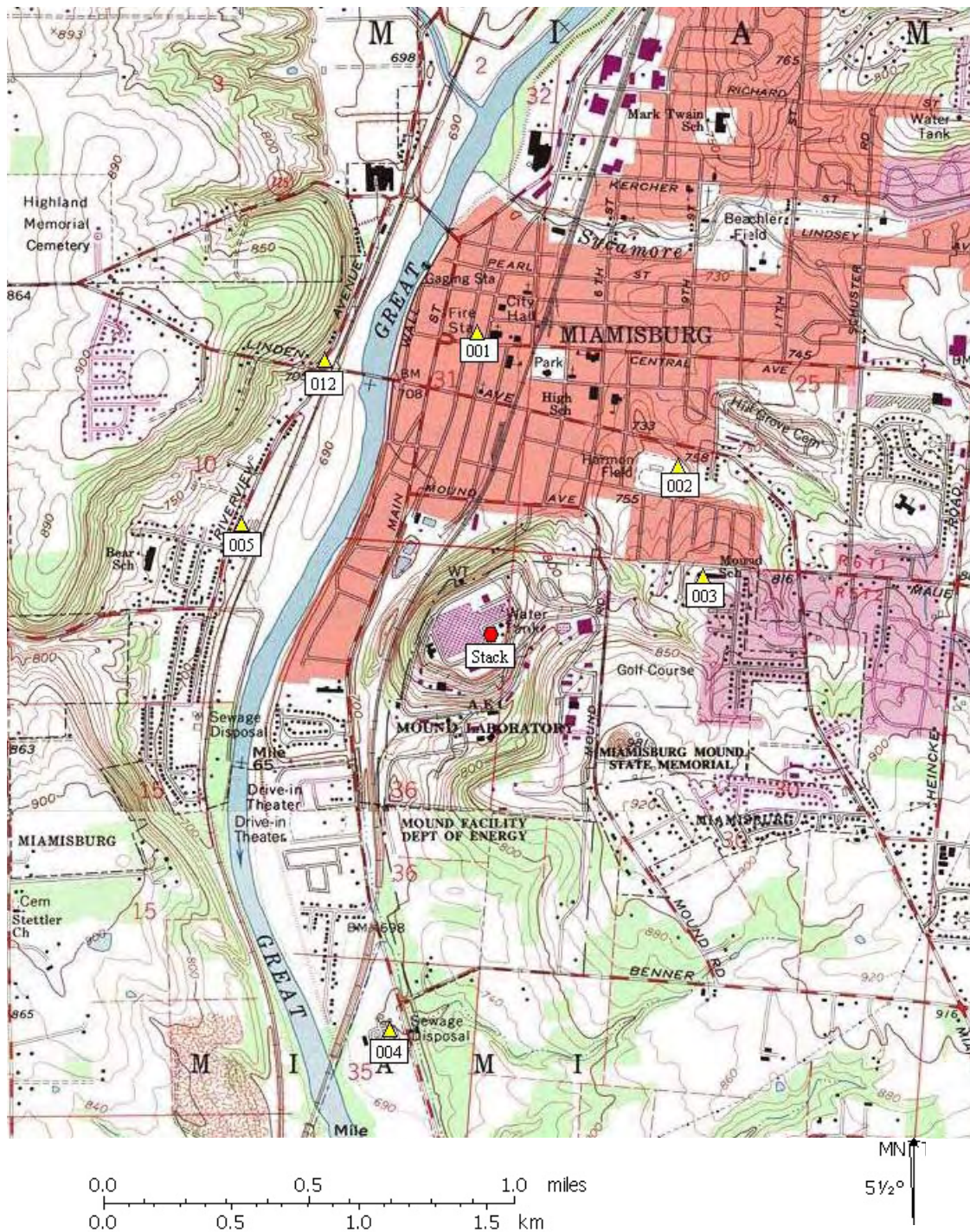


Figure 1: Air Monitoring Stations within ~1 mile of Mound Laboratory



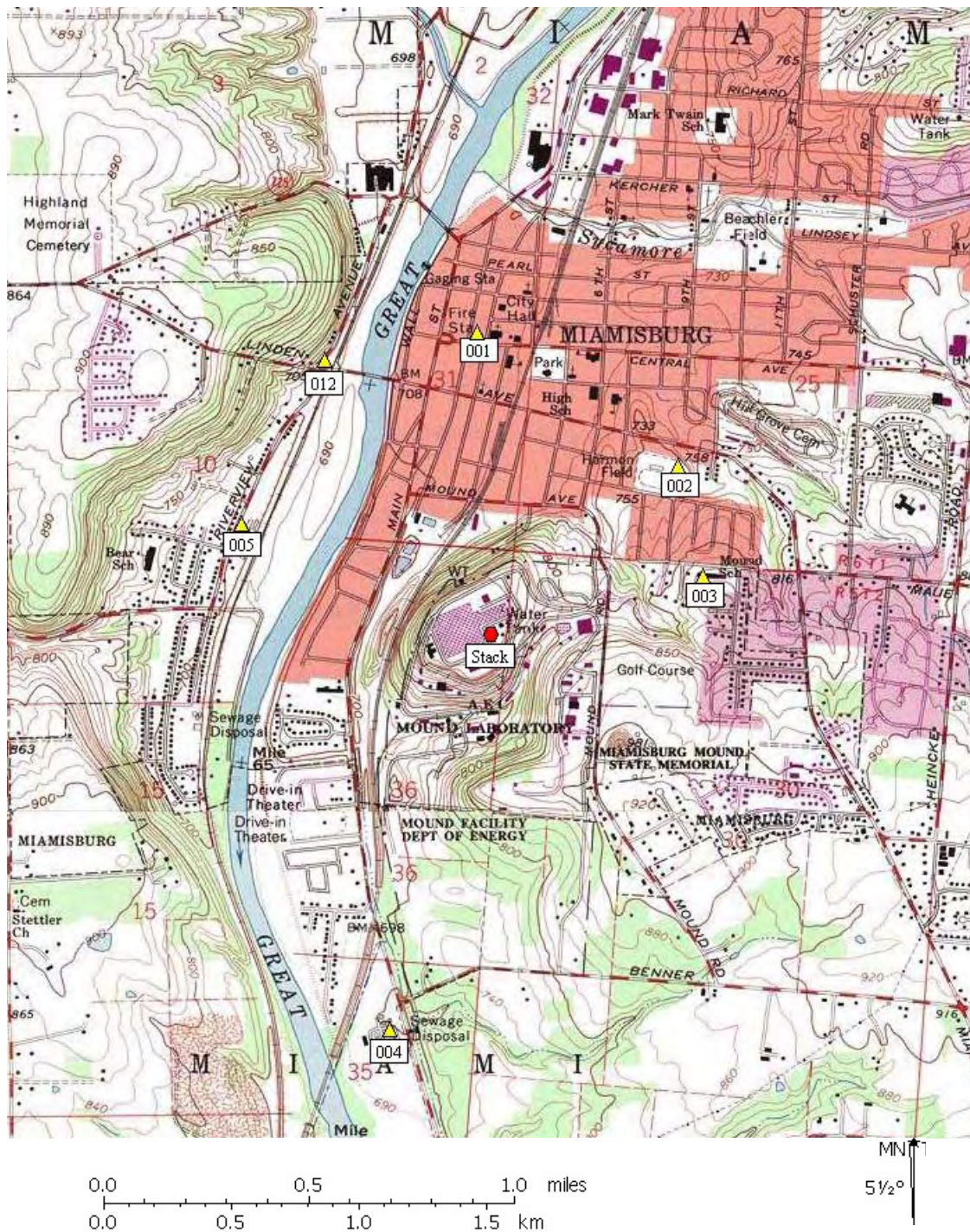
Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
001	132.5
002	84.8
003	148.2
004	27.9
005	29.4
012	143.7

Figure 2: Average Concentration of tritium oxide in sample collection solution - January through June 1973



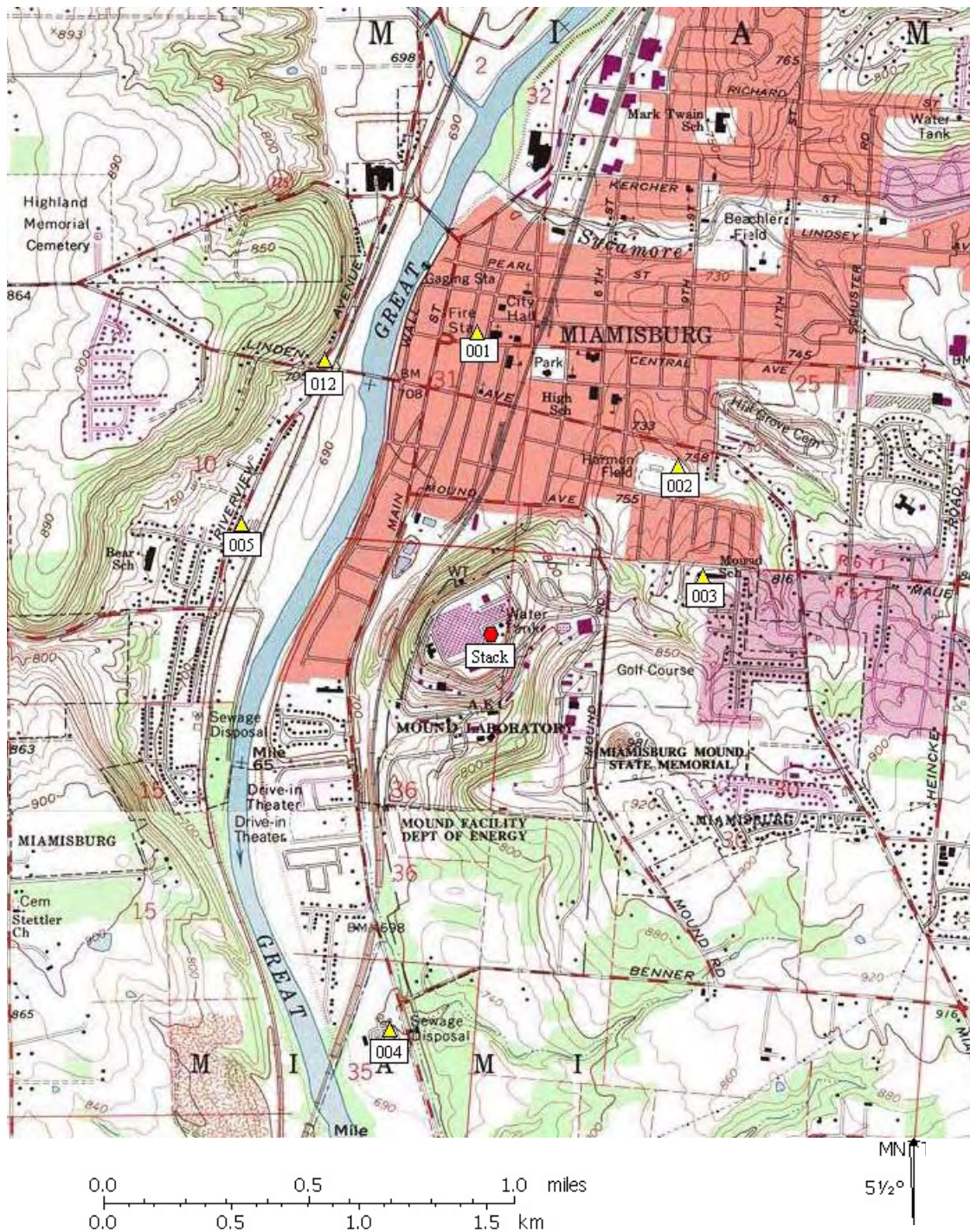
Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
001	43.0
002	53.1
003	35.0
004	29.2
005	20.1
012	19.2

Figure 3: Average Concentration of tritium oxide in sample collection solution - July through December 1973



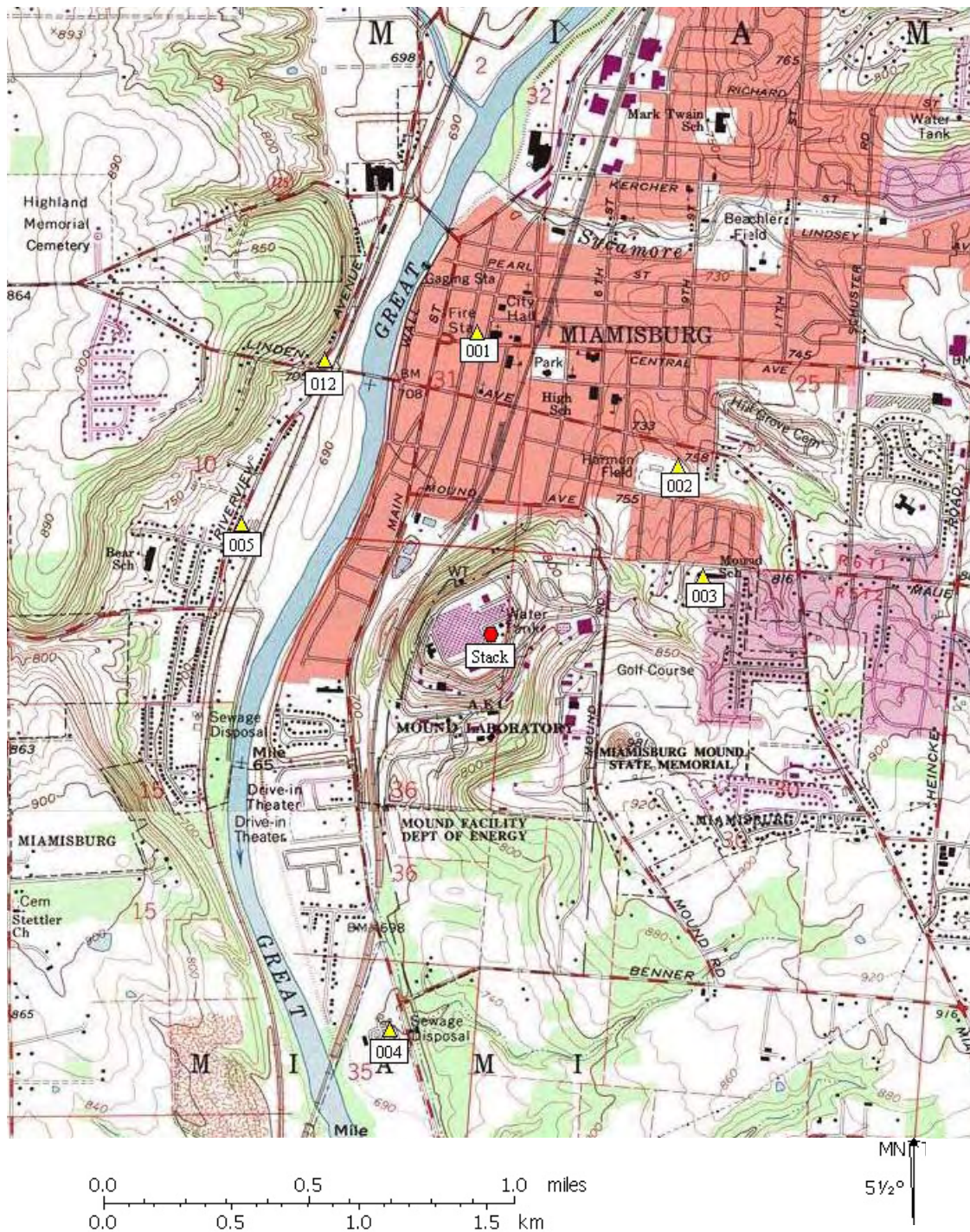
Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
001	57.2
002	53.6
003	307.5
004	29.7
005	18.8
012	28.0

Figure 4: Average Concentration of tritium oxide in sample collection solution - January through June 1974



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
001	20.6
002	21.1
003	22.5
004	11.2
005	21.5
012	16.4

Figure 5: Average Concentration of tritium oxide in sample collection solution - January through June 1975



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
001	11.7
002	13.0
003	10.5
004	7.9
005	7.9
012	7.5

Figure 6: Average Concentration of tritium oxide in sample collection solution - July through December 1975

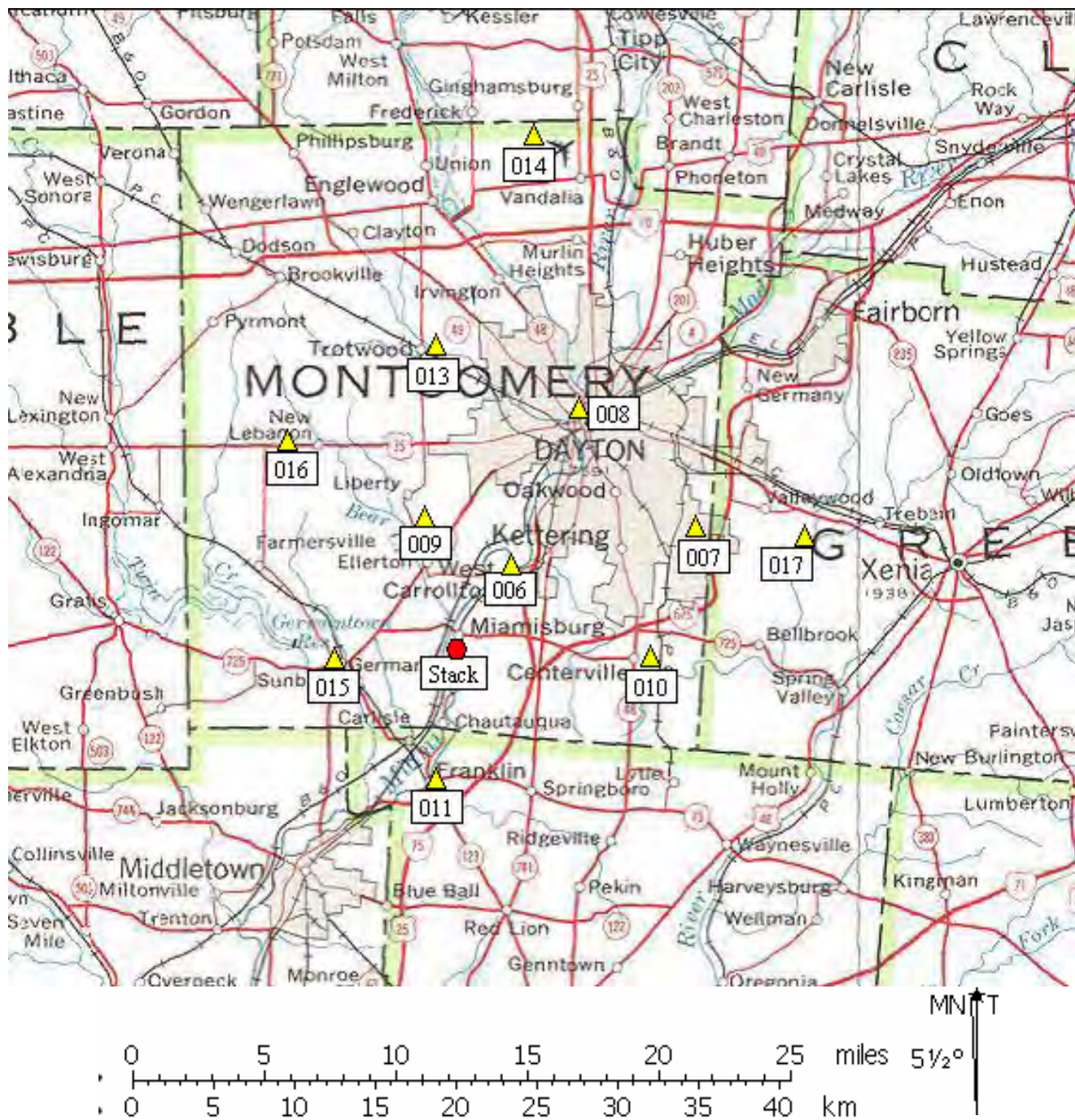
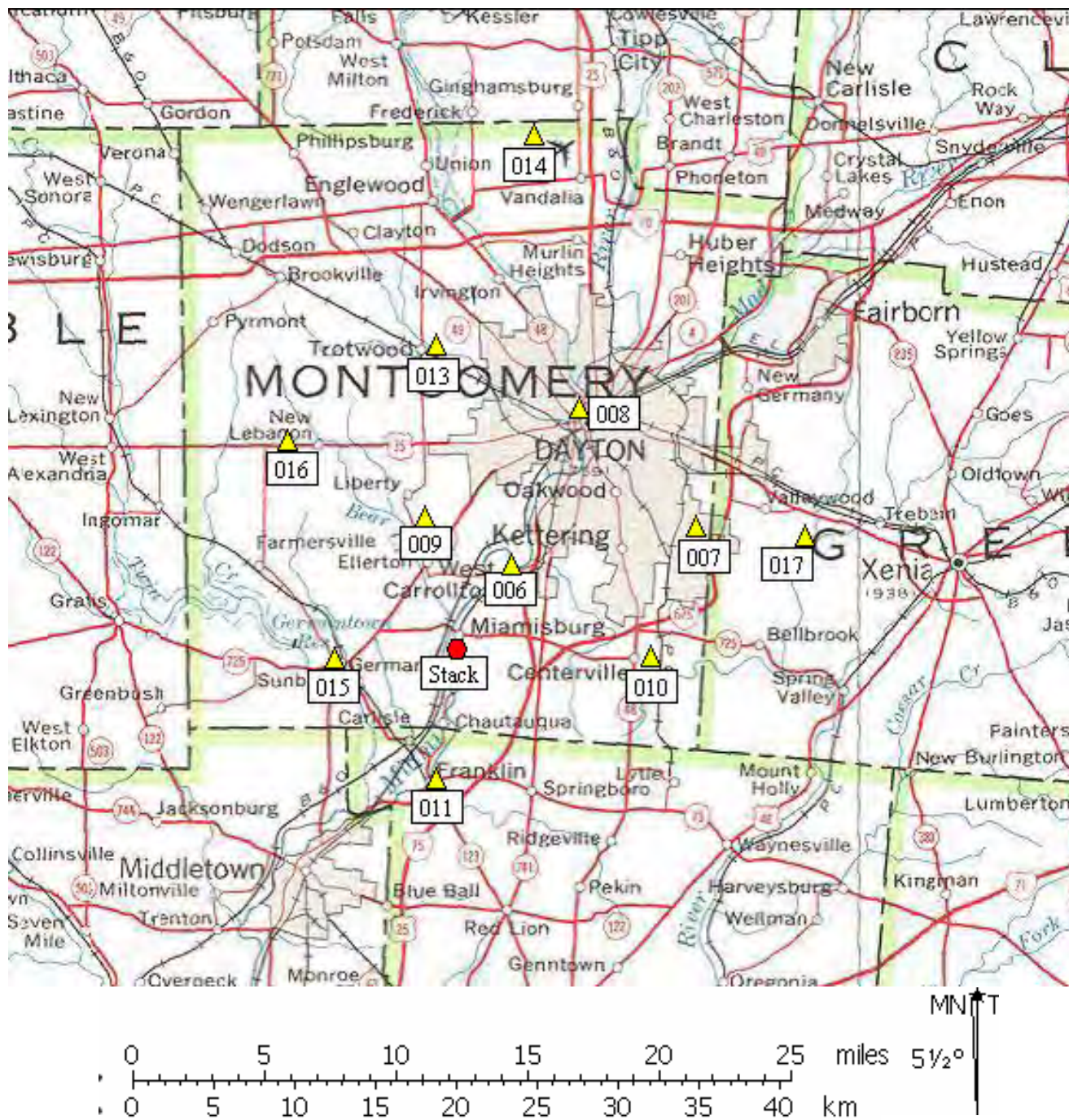
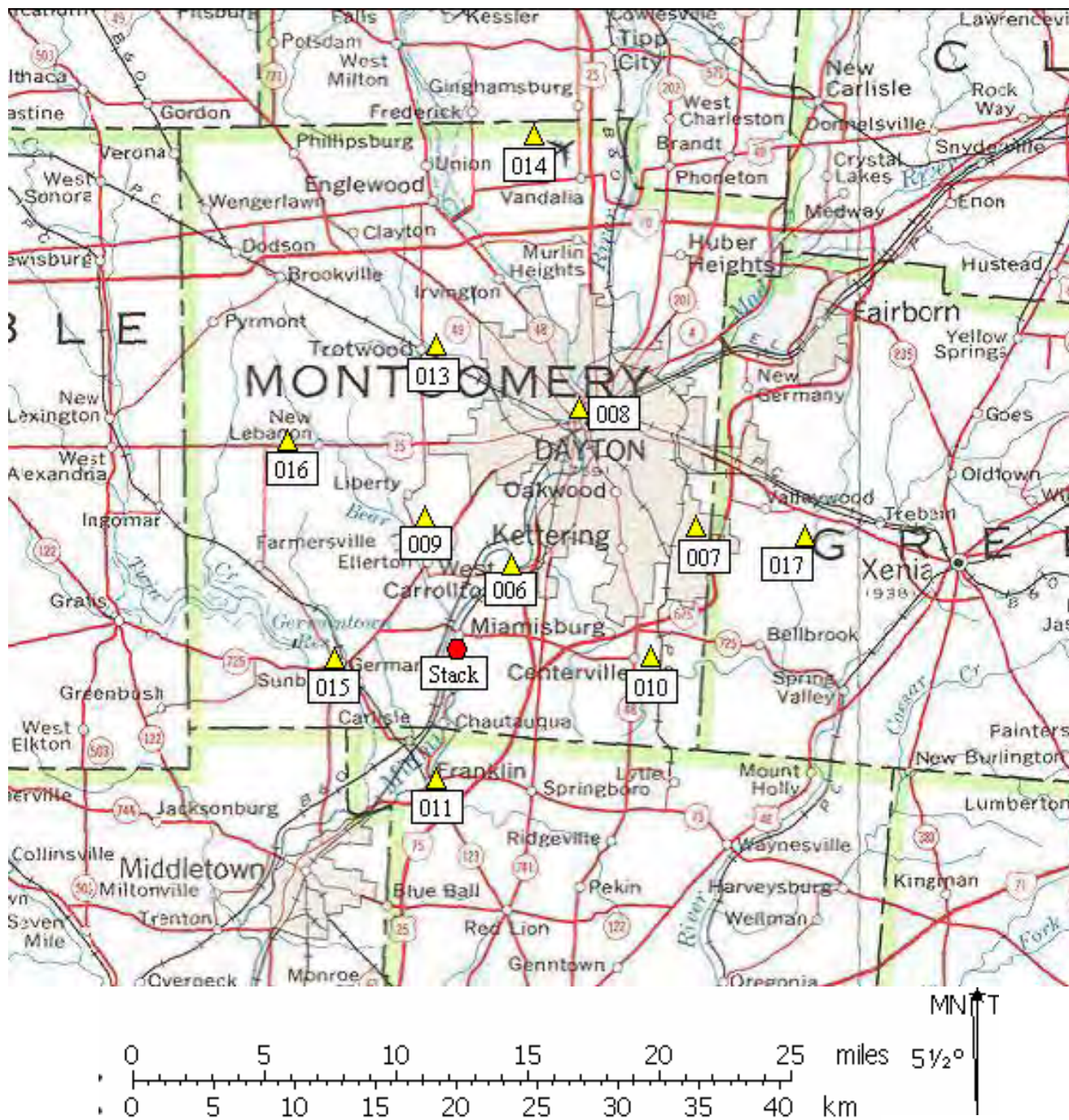


Figure 7: Air Monitoring Stations more than 1 mile from Mound Laboratory



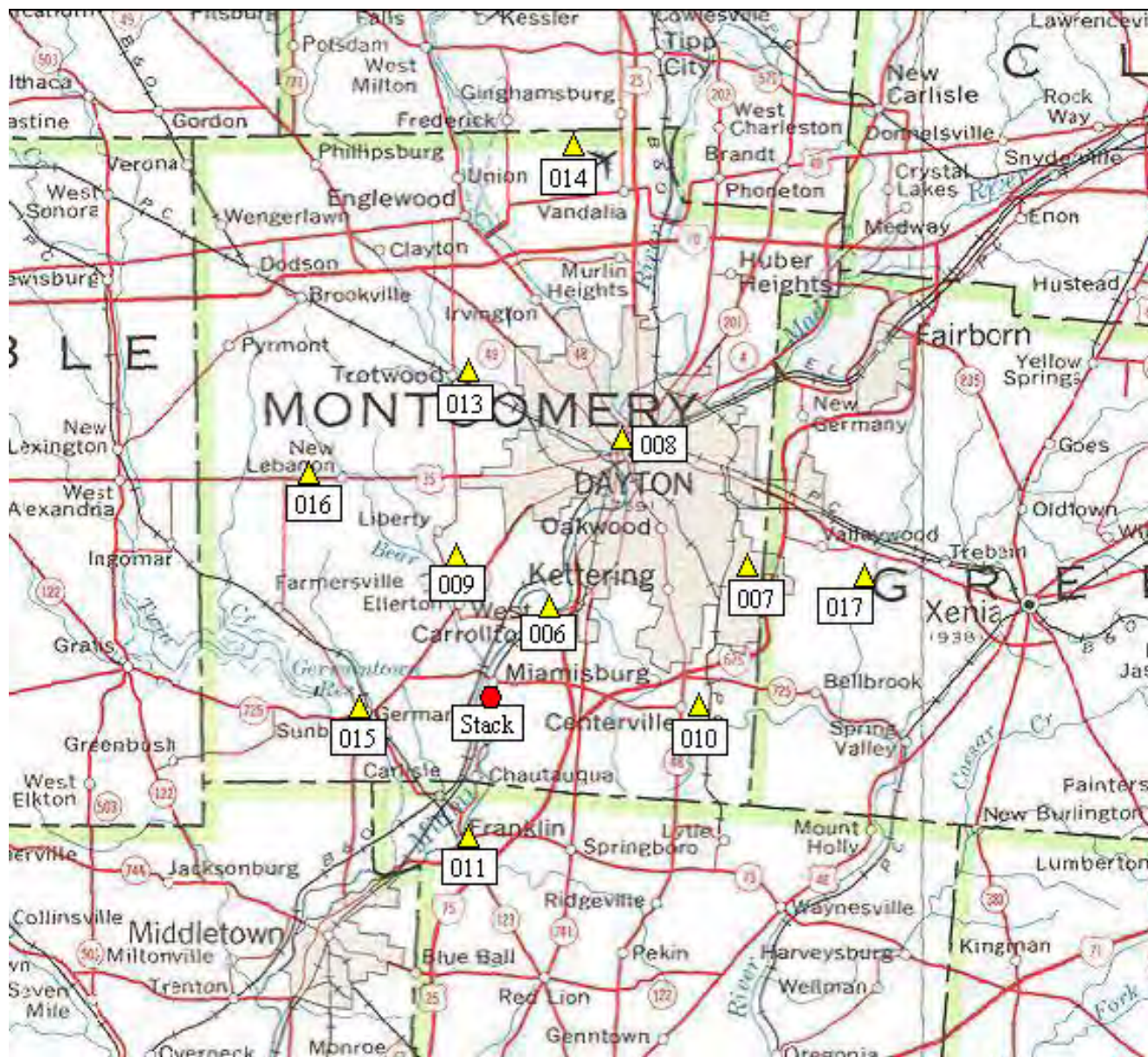
Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
006	80.1
007	56.6
008	18.4
009	60.7
010	15.5
011	30.8
013	56.7
014	49.4
015	47.3
016	67.8
017	49.4

Figure 8: Average Concentration of tritium oxide in sample collection solution - January through June 1973



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
006	14.0
007	15.9
008	8.2
009	11.5
010	14.4
011	11.0
013	9.3
014	10.2
015	8.2
016	7.5
017	10.2

Figure 9: Average Concentration of tritium oxide in sample collection solution - July through December 1973



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
006	29.4
007	34.3
008	14.8
009	15.2
010	19.5
011	7.3
013	10.9
014	8.4
015	26.1
016	8.6
017	14.3

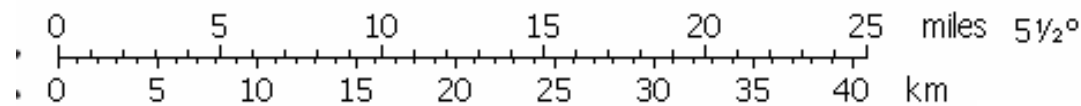
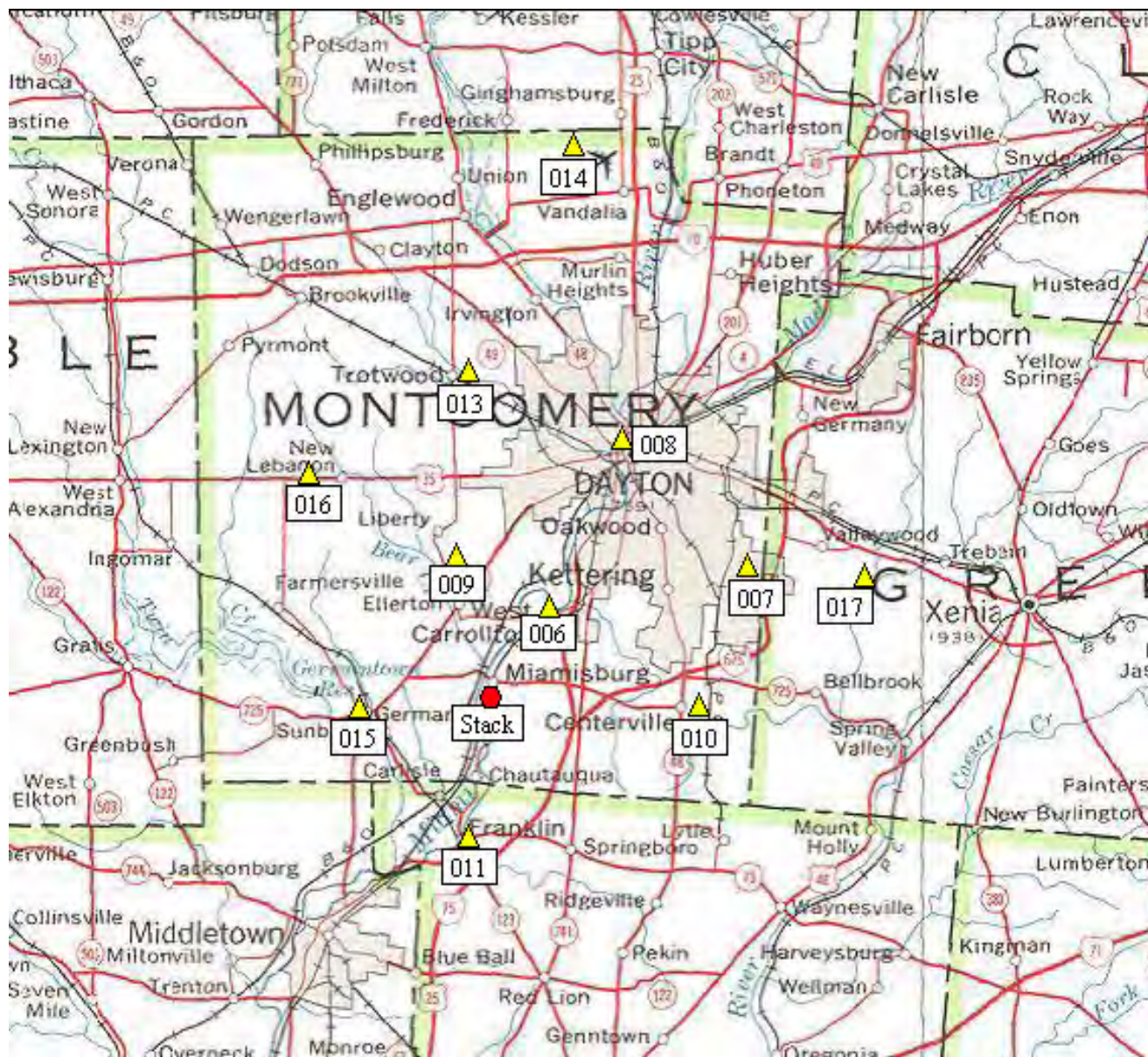


Figure 10: Average Concentration of tritium oxide in sample collection solution - January through June 1974



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
006	9.1
007	8.9
008	9.0
009	1.4
010	7.5
011	7.2
013	10.7
014	45.8
015	16.7
016	7.9
017	9.6

Figure 11: Average Concentration of tritium oxide in sample collection solution - January through June 1975



Sample Location ID	Average Concentration in pCi/cc of Ethylene Glycol Analyzed
006	4.3
007	3.0
008	5.7
009	9.7
010	4.4
011	4.7
013	7.2
014	8.6
015	4.8
016	5.6
017	4.8

Figure 12: Average Concentration of tritium oxide in sample collection solution - July through December 1975