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CHEMICAL AND RADIOLOGICAL COMPOSITION OF AIR AND GROUND WATER NEAR THE MINERALIZED URANIUM ORE DEPOSIT AT CRAWFORD, NEBRASKA

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Data on chemical composition of ground water collected from the "post-Middle Chadron" Oligocene strata are compared with those from water of the basal Chadron hydrological unit in an area near Crawford, Nebraska, where uranium mineralization is known to occur. Uncorrected Eh values, using a calomel reference electrode, indicated water from the "post-Middle Chadron" Oligocene unit was positive, whereas water from the basal Chadron was negative. The basal Chadron waters contained lower levels of calcium and uranium and higher concentrations of sodium, chloride, sulfate, and total dissolved solids, than did waters from the "post-Middle Chadron" Oligocene zone. Radon and radium concentrations in the water samples from the basal Chadron unit fluctuated widely. Radium levels in the oxidizing waters were consistently low (<2 pCi/l), while radium in several water samples from the reduced unit exceeded the maximum recommended level of 5 pCi/l for drinking purposes. These samples, however, are not used domestically.

Radon and radon daughter products in air were also evaluated on a limited basis. Their concentrations appeared similar to reported background levels in other midwestern regions.

† † †

INTRODUCTION

An announcement was made in 1981 of the discovery of a "significant" uranium deposit near Crawford (population 1,300) in northwestern Nebraska (Gigot, 1981). This ore deposit is sometimes called the Crow Butte deposit due to its proximity to neighboring Crow Butte, a landmark located a few kilometers southeast of Crawford. Wyoming Fuel Company, the discoverer of the ore deposit, has estimated a probable reserve in excess of 11,000 metric tons of U_3O_8. Extraction from the ore reserve is currently under study by the *in situ* (solution) technique. Because previous work indicated anomalous concentrations of radium in several flowing wells used for lawn and garden irrigation in Crawford, this study was undertaken to evaluate various water parameters from all known wells in Crawford serving drinking or lawn-garden purposes. Water from known flowing wells in and surrounding the mineralized zone was likewise analyzed. A further extension of the study evaluated radon and radon daughter concentrations in the atmosphere on a monthly basis at three selected sites for a one-year period. The data provide baseline information of water and air parameters in relationship to the underlying mineralized ore zone.

Geological Setting

The Crow Butte ore deposit lies immediately southeast of Crawford in a narrow band up to about 915 m wide and about 9.7 km long (Fig. 1). The ore deposit is contained exclusively in the basal Chadron Sands of Early Oligocene age at depths from 90 m to 250 m (Schultz and Stout, 1955; Collings and Knode, 1983). The confining bed below the basal Chadron Sandstone is the Pierre Shale. The Pierre is a Late Cretaceous marine deposit with relatively uniform composition throughout. It crops out north of Crawford but is not considered an aquifer as demonstrated by the need to pipe in water from other formations for drinking and livestock uses. However, water may be occasionally obtained from wells penetrating the Quaternary sediments lying on top of the Pierre.

The White River Group of Oligocene age consists of the Chadron and the Brule formations (Schultz and Stout, 1955). The Chadron is sometimes separated into three members, but for interpretation of data for this study, the White River Group is separated into the basal Chadron Sandstone and the "post-Middle Chadron Unit."

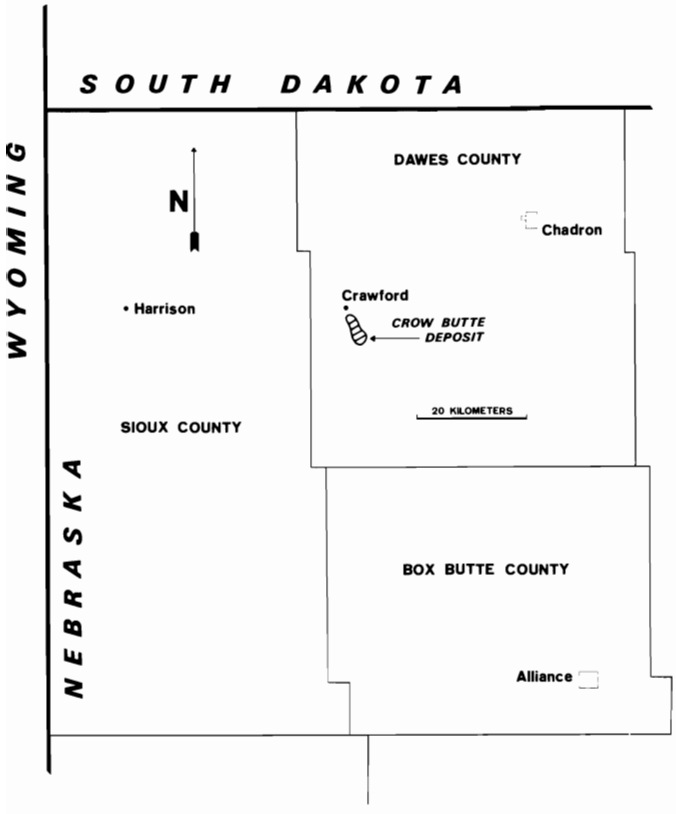

FIGURE 1. Location of the Crow Butte uranium deposit.

The basal Chadron is a depositional product of a large stream system which occurred about 36 m.y.B.P. to 38 m.y.B.P. (Collings and Knode, 1983). Regionally the basal Chadron varies in thickness from 0 to 107 m, but the thickness of the basal Chadron harboring the mineralized ore is about 12 m (Collings and Knode, 1983). Water from the basal Chadron in the Crow Butte study area is under artesian pressure. This pressure is sufficient to produce some flowing wells in the area. About 13 flowing wells emanate in the northern part of the study area (Fig. 2). Water from only two of these wells is used for drinking; the other wells are used for lawn or garden irrigation. Water from the basal Chadron is highly mineralized and considered of poor quality for drinking.

The unit immediately above the basal Chadron Sandstone in the study area is characterized by a distinct and rapid facies change (Collings and Knode, 1983). Brick-red clays interbedded with gray-green clays are reported in drill hole cuttings above the basal Chadron Sands. Above these clays are massive claystones and siltstones which range in color from dark blue-green to greenish-brown (Collings and Knode, 1983). Collectively this part of the Chadron is reported to be the Middle and Upper Chadron members in the study area; it ranges in thickness from about 40 m to 75 m (Collings and Knode, 1983). A few shallow wells penetrate the fracture zones and alluvial material and provide limited water for drinking and livestock uses.

The Brule Formation lies conformably on the Chadron Formation. It consists of two members, namely the Orella below and the Whitney above (Schultz and Stout, 1955). Most of the surface strata in the area of study are Brule. The Brule is not considered to be a reliable aquifer, but it contains low-yield shallow wells which often provide an adequate supply of water for drinking and garden-lawn uses. The Upper Brule sediments are considered to be in excess of 27 m.y.B.P. (Souders et al., 1980).

History of Uranium Investigations

Uranium ore potential in northwestern Nebraska has been recognized for several decades. In 1955, an area of the Brule Formation located about 20 km northeast of Chadron, Nebraska, was evaluated for its uranium potential (Dunham, 1955). Later, a more extensive study was made of the geology of uranium in the Dawes County and adjacent area (Dunham, 1961).

During the early 1970s, more extensive studies were conducted in northwestern Nebraska by various energy firms. Surveillance was largely by aerial and land-scintillation techniques seeking anomalous gamma-ray activities. On a reduced scale, ground and stream water samples were also analyzed for various constituents to determine their relationships to the uranium potential.

Later studies included the evaluation and correlation of various chemical and physical constituents, including uranium, in 139 well and stream water samples in northwestern Nebraska. Radon and radium were not evaluated in the study (Struempler, 1979).

In a more recent and extensive study, 543 ground water samples in the Alliance Quadrangle, Nebraska, were evaluated for their uranium potential as a part of the Natural Uranium Resource Evaluation Program (NURE) (Anonymous, 1980). The water samples were taken on a grid basis of about 8 km and included the Crawford area. Radium and radon were not determined in that study.

In 1979, the Wyoming Fuel Company began a regional drilling program which resulted in the discovery of the Crow Butte uranium deposit (Collings and Knode, 1983). Shortly after the announcement of the uranium discovery in the Crawford area, more than 600 wells in 12 townships surrounding the Crawford area were inventoried (Struempler, 1982, in an unpublished report by R. Spalding submitted to the Department of Environmental Control, Lincoln, Nebraska, for the University of Nebraska Conservation and Survey Division). In

addition, as a part of the study, 100 water samples were analyzed for various constituents. Only a few wells, however, from within Crawford, were included in that study. The study indicated that three flowing wells from Crawford contained radium-226 (Ra-226) in excess of the U.S. Public Health maximum recommendations for drinking, but they were not used for drinking.

Radionuclide Health Aspects

Uranium (U-238) is radioactive and decays through four nuclides to form radium-226 (hereafter called radium). Radium further decays to radon-222 (hereafter called radon). Radon gas further decays into the radionuclides polonium Po-218 (RaA), lead Pb-214 (RaB), bismuth Bi-214 (RaC), and polonium Po-214 (RaC′), all heavy metals (Rock and Beckman, 1979). In this decay series from radon to Po-214, a form of energy is given off as alpha or beta particles or gamma rays.

At this time, no maximum limit has been placed by health regulatory agencies within the United States for uranium in drinking water. Likewise, there is no established limit for radon gas in drinking water. The health implications of radium, however, make it the most important naturally occurring radionuclide that is monitored in public water systems. Radium may substitute for calcium in bone and accumulate with time because the half life of radium is about 1,600 yr. Estimates of risk indicate that continuous consumption of water containing radium at the maximum contamination of 5 picocurie per liter (pCi/l) may cause between 0.7 and 3.0 cancers per million exposed persons (Anonymous, 1976).

Even though no limit has been established on radon in drinking water, radon in air presents a different biological effect. It is not radon gas in the air but its decay daughters that cause concern. Radon daughters are commonly considered to be the first four elements (RaA, RaB, RaC, and RaC′) immediately following radon in the U-238 decay series. They have short half lives, averaging about 30 min. The daughters tend to attach to dust and other condensation nuclei, especially in the air of underground mines. Because of their high-energy alpha particles, >5 million electron volts (MeV), and short half lives, radon daughters can deliver extensive radiation damage directly to the epithelial lining of the lungs. It is estimated that between 25% and 50% of radon daughters are retained in the lungs (Rock and Beckman, 1979).

METHODS

Water samples were collected during the summer of 1983. Sample collections, preservation, and analyses, except those indicated, followed those procedures given in Standard Methods (Anonymous, 1981). All collections were fresh water samples.

Field analyses included pH, Eh, and H_2S. A Model 407-A Orion specific-ion meter coupled with a Model 96-78 redox electrode served for the Eh measurements (Orion Research, Inc., Cambridge, Massachusetts 02139). Thus, Eh is reported as uncorrected relative to the saturated calomel reference electrode potential. The remainder of the analyses were conducted in the chemistry laboratories at nearby Chadron State College.

Alkalinity was determined by titrating unfiltered samples (<8 hr old) with standardized H_2SO_4 to pH 4.5. Water for the remainder of the analyses was filtered through 0.45 μm porosity filters. Sulfate (SO_4^{-2}), nitrate nitrogen (NO_3-N), and specific conductance (SC) were analyzed via techniques, equipment, and reagents of the Hack Company (Loveland, Colorado 80537). Calcium (Ca^{+2}), magnesium (Mg^{+2}), and potassium (K^+) were determined by standard atomic-absorption procedures. Sodium (Na^+) was analyzed via flame photometry. Chloride (Cl^-) was determined by titration with mercuric nitrate using diphenylcarbazone as the indicator. Uranium (U^{+6}) was analyzed by fluorometry according to the procedure of Cagle (1982). Radon and radon daughters were measured by an EDA Model RDA-200 Radon/Radon Daughter Detector using their recommended procedures (EDA Instruments, Inc., 1 Thorncliff Park Drive, Toronto, Canada M4H 109). Appropriate factors were applied to convert alpha activity to pCi/l for radon and working levels (WLs) for the daughter products.

Quality control procedures included the use of the following standard references: NO_3-N (Hack Company); total dissolved solids (TDS), specific conductance (SC), pH, Ca^{+2}, Mg^{+2}, Na^+, K^+, SO_4^{-2}, and Cl^- (U.S. Environmental Protection Agency, Quality Assurance Branch, Cincinnati, Ohio 45368); and U^{+6} (U.S. Department of Commerce, National Bureau of Standards, Washington, D.C. 20234). Radium quality control procedures included the participation in the Environmental Protection Agency Laboratory Interpretation Program (Environmental Monitoring Systems Laboratory, P.O. Box 15027, Las Vegas, Nevada 89114) with a deviation of 0.4, -1.3, and -0.9 sigma for three anaylses from the Environmental Protection Agency's expected laboratory deviation. Appropriate spiked samples were also a part of the analytical procedures.

RESULTS AND DISCUSSION

Water Studies

Thirteen flowing well samples were collected and were suspected to have issued from the basal Chadron (Fig. 2). The remainder of the samples was collected from non-flowing wells within Crawford, and all are believed to have issued from

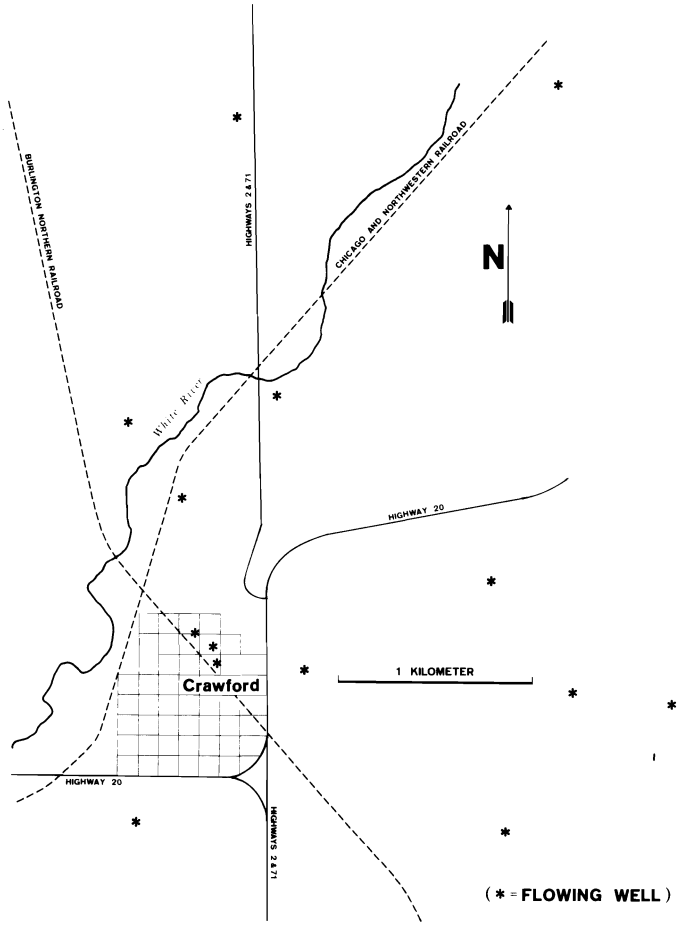

FIGURE 2. Location of the flowing wells sampled for analysis.

the "post-Middle Chadron" Oligocene strata, which include, for the purposes of this study, the Brule rocks. For evaluation, each water source is listed on the basis of its suspected producing horizon. In doing so, the very young rocks may have been combined with old rocks of the White River Group, but without well logs, which are largely unavailable, this classification scheme seemed best for this study.

The composition of well water from the "post-Middle Chadron" Oligocene rocks is shown on Table I. The Brule Formation appears to be the major ground water source; some wells, however, along the White River in Crawford appear to tap the alluvial fill associated with the paleostream channels and faults within the Upper Chadron.

The water from the "post-Middle Chadron" Oligocene rocks is oxidizing as manifested by the positive Eh values. Specific conductance and total dissolved solids are indicators of the total ionic content of the water composition. The major ions in ground water are calcium, magnesium, sodium, potassium, chloride, sulfate, and bicarbonate. These major ions comprise almost entirely the total dissolved chemical species in ground water regardless of the extreme range of total dissolved solids between high and low total dissolved solids waters (Freeze and Cherry, 1979). Concentrations of total dissolved solids fall within the range of previous studies from related host geological sources (Anonymous, 1980; Struempler, 1979; Struempler, 1982, in an unpublished report by R. Spalding submitted to the Department of Environmental Control, Lincoln, Nebraska, for the University of Nebraska Conservation and Survey Division). Nitrate nitrogen levels were below maximum limits of 10 mg/l for municipal or public supply established by the Environmental Protection Agency (Anonymous, 1976). No nitrate nitrogen level approached this limit and none should be of concern in drinking water. Hydrogen sulfide was not detected in any sample.

Uranium concentrations in the "post-Middle Chadron" Oligocene water range between 11 ppb and 67 ppb (parts per billion, or mg/l), with a mean of 35 μg/l. This range is typical of ground water from the Oligocene and is considered high relative to sea water (3 ppb) and most surface and ground waters (<10 ppb) (Barker et al., 1965).

Radon, a daughter product in the U-238 series, ranges between 280 pCi/l and 1,180 pCi/l. In a previous study in the Crawford area, the average radon concentration in well water from 27 wells from the Upper Chadron was 346 pCi/l; from 29 wells in the Brule Formation, the average was 273 pCi/l. These values may represent baseline concentrations of radon in water from formations in the Crawford area.

Radium, also a daughter product in the U-238 series, was not detected in excess of 2 pCi/l. Most samples contained less than 1 pCi/l, well below the maximum limit of 5 pCi/l, and should not be a concern for drinking, garden, or lawn uses.

By contrast, the basal Chadron waters are reducing (Eh<0.0) and demonstrate a marked change in major ion and trace metal concentrations (Table II) relative to water from the "post-Middle Chadron" Oligocene rocks. Hydrogen sulfide was detectable in some samples and may act as a reductant in the precipitation of uranium and other metallic elements. Specific conductance and total dissolved solids showed a marked increase due largely to increases in sodium, sulfate, and chloride. On the other hand, there is a pronounced decrease in uranium and calcium concentrations. Uranium ranges from 19 ppb to <1 ppb, which is notably less than those of the oxidizing waters of the "post-Middle Chadron" Oligocene rocks.

Radon exhibits a marked increase in concentration when compared to the "post-Middle Chadron" Oligocene waters and exceeded 65,000 pCi/l in two flowing wells. By contrast, many water samples from wells in the NURE Program in the east-

TABLE I. Well water composition from the "post-Middle Chadron" Oligocene strata.

No.	Use*	Depth (meters)	Temp (°C)	pH	Eh† (mv)	SC µmho/cm	ppm Ca	Mg	Na	K	HCO_3	SO_4	Cl	TDS	Major Ion (Bal%)	NO_3-N (ppm)	Rn (pCi/l)	Ra (pCi/l)	U (ppb)
1W	I		15	7.6	20	850	28	4	190	17	420	115	40	814	0.2	0.5	755	<1	43
2W	D&I	50	14	8.2	10	900	12	2	205	16	320	90	55	800	-2.2	0.5	510	<1	29
3W	D&I	30	13	7.2	200	1,250	66	16	160	42	420	150	75	929	4.1	0.5	345	<1	22
4W	I	15	13	7.3	75	1,190	51	8	230	26	610	110	95	1,130	-7.3	1.5	595	<1	44
5W	I	12	13	7.3	70	1,200	55	9	290	26	740	135	95	1,350	-4.9	1.5	770	<1	67
6W	I	14	12	8.0	50	1,080	30	3	220	13	455	130	70	921	-4.1	1.0	1,180	<1	48
7W	I		12	7.5	70	1,100	62	9	205	20	575	105	40	1,016	3.9	1.5	410	<1	46
8W	D&I	18	13	7.5	140	1,100	63	12	180	26	475	150	65	971	-0.9	0.5	470	<1	40
9W	I	15	13	7.5	40	1,080	62	12	155	26	400	160	80	895	-5.4	Trace	295	1	24
10W	D&I		13	7.6	90	990	63	11	150	25	405	130	80	864	-3.4	Trace	325	<1	26
11W	I	14	12	7.4	20	1,530	95	17	250	26	530	225	120	1,293	1.7	1.5	285	<1	60
12W	I	16	14	7.5	120	790	39	7	130	20	250	80	55	681	-3.0	0.2	415	1	19
13W	I	24	14	7.8	130	630	26	2	125	13	350	25	25	566	3.7	1.0	740	<1	23
14W	I	24	14	7.7	100	930	33	3	190	17	385	125	45	798	3.8	1.5	585	<1	30
15W	I		14	7.5	120	910	43	7	155	20	420	80	60	785	-2.6	1.5	290	1	30
16W	I		14	7.3	95	1,040	56	10	150	24	435	95	55	825	0.9	1.0	340	1	13
17W	D&I	12	15	7.4	95	1,060	60	10	170	25	420	130	70	885	2.4	1.5	570	2	11
18W	I		12	7.4		1,230	90	23	175	46	450	215	110	1,109	1.4	1.0	280	<1	41
19W	I	12	13	7.6		970	30	5	190	20	415	110	60	830	-1.0	1.0	580	<1	36
20W	I		13	7.5		1,550	90	17	250	28	420	320	145	1,270	-0.9	0.5	455	<1	49
21W	D&I	20	13	7.6		850	34	6	150	19	365	65	55	694	3.4	1.1	480	2	43
22W	I		13	7.6		870	50	9	155	24	365	125	60	788	2.9	1.0	455	<1	36
23W	I		13	7.8		560	16	2	120	16	335	18	20	527	2.4	0.2	380	<1	25
	Mean		13.2	7.5		1,029	50	9	181	23	442	127	68	789			500		35

*I = irrigation, D&I = drinking and irrigation.

†Uncorrected Eh.

central Minnesota region showed radon concentration of 2,000 pCi/l (Morey and Lively, 1980). In 228 well samples in Maine and 26 wells in New Hampshire, radon concentrations averaged 53,000 pCi/l and 101,000 pCi/l respectively (Lucas, 1964).

In contrast to the oxidizing waters, radium in samples containing lower redox potentials showed a wide range of values from 174 pCi/l to <1 pCi/l. Five of the samples contained radium in excess of 5 pCi/l, the maximum limits established for drinking water. Only two samples were used domestically, and they contained <0.5 pCi/l of radium; this level falls far below the maximum health limit of radium for drinking water use.

Of interest is that the temperature of ground water from wells increased with the depth of the well. The temperature of water from the shallower non-flowing wells (Table I) averaged 13.2°C, whereas the temperature of water from the deeper flowing wells (Table II) averaged 16.4°C. This trend is in keeping with the observation that the temperature of water from wells and springs increases about 1.8°C for each 100 m down from the land surface (Heath, 1983).

Atmospheric Studies

Radon gas and radon daughter products were measured monthly from November 1982 until November 1983. Collection sites for outside air included the Nebraska Department of Road's building located on the highway in northeast Crawford and the Crawford municipal building. Air was also collected in the basement of the municipal building. No limit has been established for radon in air, but a 0.10 working level (WL) has been established as the need for more sampling to

TABLE II. Flowing water composition from the basal Chadron.

No.	Depth (meters)	Temp (°C)	pH	Eh* (mv)	H_2S†	SC µmho/cm	ppm Ca	Mg	Na	K	HCO_3	SO_4	Cl	TDS	Major Ion (Bal%)	NO_3-N (ppm)	Rn (pCi/l)	Ra (pCi/l)	U (ppb)
1A	66	13	8.2	-295	Trace	1,160	41	2	405	14	530	300	220	1,512	-4.5	1.5	175	< 1	< 1
2A	90	15	8.1	-310	ND‡	1,430	17	1	380	10	385	300	175	1,268	1.2	ND	750	1	1
3A	87	16	8.2	-200	ND	1,780	18	2	540	14	425	595	175	1,769	2.5	ND	175	< 1	< 1
4A	68	17	8.2	-120	ND	1,500	22	2	550	17	435	650	165	1,841	1.1	ND	515	< 1	< 1
5A	75	20	7.7	-180	Trace	2,500	28	5	630	19	460	835	195	2,172	-2.4	ND	2,090	9	< 1
6A		17	8.0	-200	ND	1,950	25	5	475	10	490	500	175	1,680	-4.5	ND	1,350	2	3
7A	84	16	8.2	-250	ND	1,730	15	3	415	11	360	170	170	1,449	-3.8	ND	24,315	38	16
8A	86	17	8.0	-240	Trace	2,100	18	4	520	15	410	640	175	1,782	-3.0	ND	68,570	174	19
9A	75	16	8.2	-280	ND	1,790	11	3	440	11	475	345	160	1,445	3.6	ND	69,825	86	5
10A	180	15	8.1	-300	⊕	1,370	14	2	390	8	480	300	170	1,364	-4.7	0.5	11,640	20	1
11A		15	8.0	-280	⊕	1,730	15	3	420	10	450	370	185	1,453	-3.9	0.5	530	< 1	2
12A		16	8.3	-310	ND	2,300	19	3	500	21	420	630	185	1,778	-6.9	0.5	380	1	1
13A		20	8.3	-320	Trace	2,400	20	5	480	24	420	620	160	1,729	-5.8	ND	580	< 1	4
	Mean	16.4	8.1	-253		1,826	20	3	473	14	450	496	178	1,415					

*Uncorrected Eh.

†Trace = <1 ppm.

‡ND = not detectable.

⊕ >1 and <5 ppm.

assess the potential hazard of radon daughters (Rock and Beckman, 1979).

Radon in outside air averaged 0.15 pCi/l and 0.26 pCi/l at the two sites (Table III). Radon gas in the basement averaged 1.15 pCi/l or about a five-fold increase over outside air. The outside radon gas concentration equaled normal background levels of about 0.2 pCi/l for an 8-mo period at four sites in another study in the Crawford area (*from* volume II of the unpublished application and supporting report for research and development source material license submitted by the Wyoming Fuel Company to the U.S. Nuclear Regulatory Commission on 11 February 1983). Although outdoor levels of radon can vary widely, a concentration of about 0.2 pCi/l appears to be a conservative value for outside air in the midwestern states.

In other areas, a value of 0.13 pCi/l is reported for radon in outside air in New Jersey. In Grand Junction, Colorado, in a more uranium-associated environment, radon in outside air averaged 0.73 pCi/l. Where uranium mill tailings were accidentally incorporated in residential structures in Grand Junction, radon in basement air manifested 57 pCi/l compared to 0.92 pCi/l in basements of residential buildings in which mill tailings were not incorporated (Spitz et al., 1978).

Radon daughters, however, have an established working level. No radon daughter concentration in this study approached the 0.10 working level commonly used as a reference base for further study. In a New Jersey study, the mean working levels have been between 0.002 and 0.010 (George and Breslin, 1978). In 343 Canadian homes, 75% had <0.01 working levels; the highest had 0.19 working levels (Taniguchi and Vasudev, 1978).

Based on limited sampling, atmospheric radon in the Crawford area appears to be similar to other reported pristine areas. Further studies with a larger number of samples would be required to confirm these preliminary results.

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TABLE III. Monthly radon (pCi/l) and radon daughter (WLs) concentrations at Crawford, Nebraska, 1982–1983.

	Nebraska highway building (outside air)		Crawford city hall (outside air)		Crawford city hall (basement air)	
Date	Radon	Radon Daughters	Radon	Radon Daughters	Radon	Radon Daughters
5 November 1982	0.29	0.004	0.21	0.005	0.31	0.001
27 November 1982	0.05	0.004	0.15	0.004	0.05	0.001
22 December 1982	0.16	0.002	0.16	0.001	0.05	0.005
1 February 1983	0.05	0.004	0.08	0.003	0.11	0.003
1 March 1983	0.11	0.001	0.05	0.001	0.74	0.002
30 March 1983	0.05	0.001	0.05	0.008	0.40	0.003
26 April 1983	0.05	0.001	0.48	0.001	1.82	0.003
2 June 1983	0.13	0.002	0.06	0.002	2.15	0.003
29 June 1983	0.05	0.001	0.56	0.001	2.62	0.003
29 July 1983	0.05	0.002	0.35	0.003	2.57	0.005
25 August 1983	0.56	0.002	0.19	0.002	0.16	0.002
14 October 1983	0.43	0.004	0.36	0.002	2.01	0.002
4 November 1983	0.05	0.002	0.61	0.001	1.91	0.003
Mean	0.15	0.002	0.26	0.003	1.15	0.003
S.D.	0.17	0.001	0.20	0.002	1.04	0.001

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