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ABSTRACT

Groundwater, surface water, and soil in the Goose Lake oil field in northeastern Montana have been affected by Cl^--rich oil-field brines during long-term petroleum production. Ongoing multidisciplinary geochemical and geophysical studies have identified the degree and local extent of interaction between brine and groundwater. Fourteen samples representing groundwater, surface water, and brine were collected for Sr isotope analyses to evaluate the usefulness of $^{87}Sr/^{86}Sr$ in detecting small amounts of brine. Differences in Sr concentrations and $^{87}Sr/^{86}Sr$ are optimal at this site for the experiment. Strontium concentrations range from 0.13 to 36.9 mg/L, and corresponding $^{87}Sr/^{86}Sr$ values range from 0.71097 to 0.70828. The local brine has 168 mg/L Sr and a $^{87}Sr/^{86}Sr$ value of 0.70802. Mixing relationships are evident in the data set and illustrate the sensitivity of Sr in detecting small amounts of brine in groundwater. The location of data points on a Sr isotope-concentration plot is readily explained by an evaporation-mixing model. The model is supported by the variation in concentrations of most of the other solutes.

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1. Introduction

The Prairie Pothole Region of the northern Great Plains of the USA and Canada (Fig. 1) contains many small lakes and sloughs in glacial till and outwash deposited at the margins of continental ice sheets during the Pleistocene. Sloan (1972) describes the water budget of these lakes as input by direct precipitation, basin runoff, and seepage inflow of groundwater and output by evapotranspiration, and seepage outflow. The groundwater and surface water is naturally saline with large concentrations of Na, Mg, SO_4^{2-} and HCO_3^- and relatively small concentrations of Cl^-. Salinity in some of the lakes increases seasonally because of evaporation during summer months. Others have reached an approximate equilibrium with groundwater recharge and evaporation and are perpetually saline. For example, Little Manitou Lake near Watrous, Saskatchewan, a highly saline lake with a density of 1.06, has attracted visitors since the early 19th century because of the supposed curative powers of the water (Lewry, 2007). Other lakes in the region are of interest because they contain economic deposits of sodium sulfate or mirabolite (Kelley and Holmden, 2001). Further, the Prairie Pothole Region is a critical habitat for waterfowl and other terrestrial and aquatic species (Guntenspergen et al., 2006).

Many of the lakes have been drained for agricultural purposes, and many others have been affected by oil-field brines (Reiten and Tischmak, 1993; Preston, 2011). Much of the Prairie Pothole Region is coincident with two major sedimentary basins that have produced oil and gas for more than 60 a: (1) the Williston Basin centered in western North Dakota and extending into eastern Montana, southern Saskatchewan, and southwestern Manitoba (Fig. 1), and (2) the southern part of the Western Canada sedimentary basin in southern Alberta. The effects of releases and spills of highly-saline brines co-produced with oil during the past 60 a are widespread in Sheridan County, Montana (Reiten and Tischmak, 1993; Gleason et al., 2011; Preston, 2011). Reiten and Tischmak (1993) developed an index for identifying brine contamination (abbreviated here as CIx). This index is the ratio of Cl^- concentration to specific conductance, both readily measured in the field. Oil-field brines are dominated by Na and Cl in halite proportions and may contain significant concentrations of other anions and cations including B, Ba, Li, Mn, Sr, Zn, Br and I. More recent activity in the Williston Basin (Fig. 1) involves increased drilling and production from the Bakken Formation, a thin but widespread organic-rich shale and sandstone unit that forms an extensive continuous petroleum system (Polastro et al., 2008).

The purpose of this study was to evaluate the use of Sr isotopes for detecting small amounts of oil-field brine in groundwater (Peterman et al., 2010). Previously, this approach has been used successfully to track the release of formation waters associated with production of CH_4 from coal beds (Frost et al., 2002). Strontium is an alkaline-earth element that closely follows Ca in the hydrologic cycle. Combining the isotopic composition ($^{87}Sr/^{86}Sr$)

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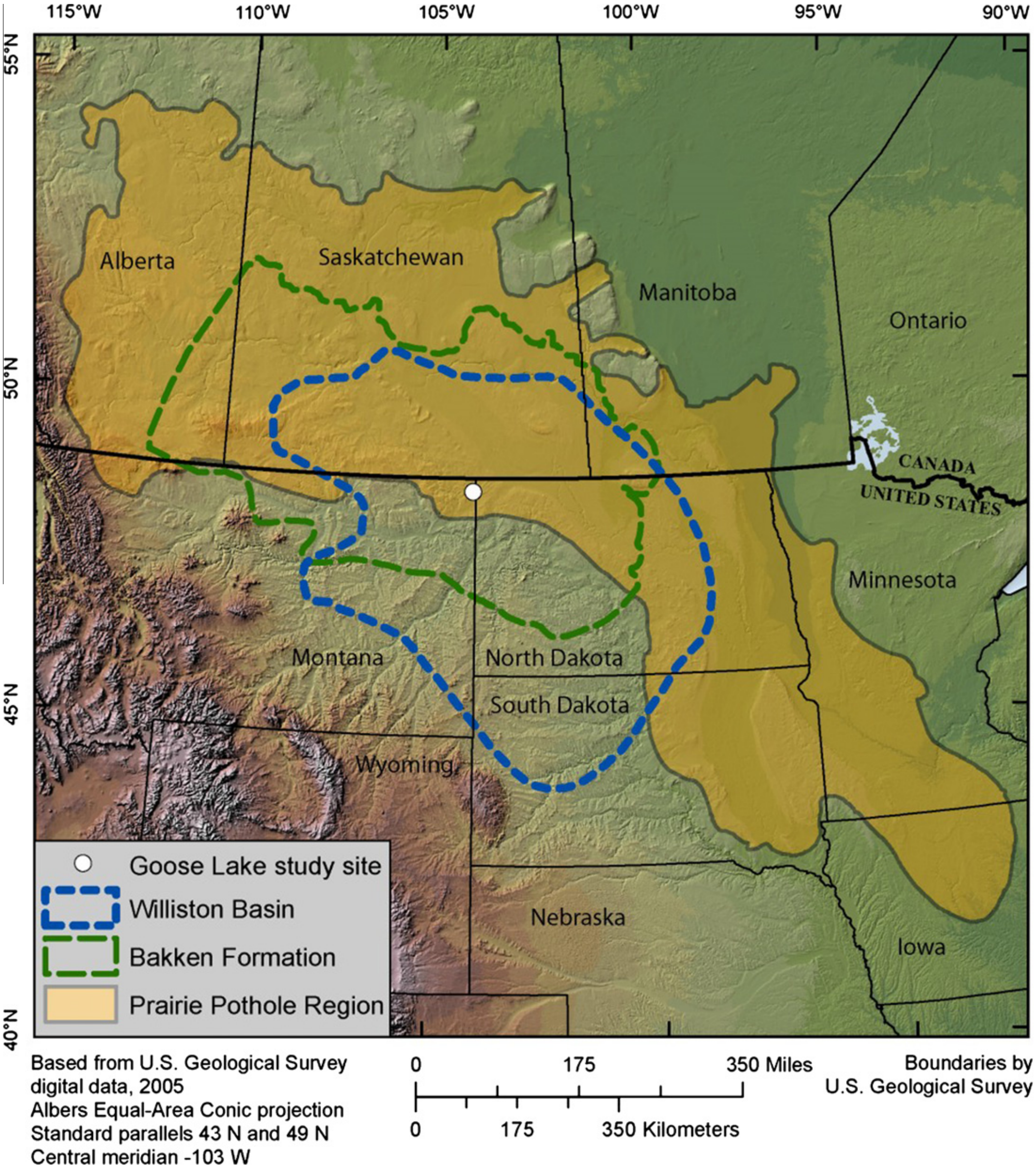

Fig. 1. Map showing the prairie pothole lakes region in the USA and Canada, the outline of the Williston Basin, and the extent of the Bakken Formation, which is the target of intense petroleum exploration and development. Location of the Goose Lake study area is shown by the white dot.

with the Sr concentration is an effective way to evaluate and quantify mixing, especially when the end members differ significantly in both isotope ratio and concentration (Peterman and Stuckless, 1992; Shand et al., 2009). An area within the Goose Lake oil field in northeastern Montana (Fig. 2) was selected to conduct this test because of an ongoing multidisciplinary study (http://steppe.cr.gov/), which has identified and delineated brine plumes (Reiten and Tischmak, 1993; Preston, 2011).

2. Sample collection and analysis methods

Water samples were collected from monitoring wells, wetlands, and an oil/brine separation tank at the Goose Lake study site during June 2009 (Table 1). Monitoring wells were purged until at least three well volumes of water were removed and onsite measurement of pH, water temperature, and specific conductance had stabilized. Groundwater samples were collected using a 5 cm (2-in.) submersible pump.

Wetlands were sampled using a discrete sampling method modified from Knapton (1985). All of the wetland sites were shallow (typically less than 1.5 m deep) and tended to be well mixed by wind-generated turbulence, so a single sampling location was assumed to be representative of the entire wetland. Prior to sample collection, specific conductance was measured at various locations along the perimeter of the wetland to confirm adequate mixing. Samples were collected by wading to as deep a location as possible. An acid-rinsed 4-l polyethylene bottle was submersed upwind of the sample-collecting person and the lid was removed after submersion to fill the bottle. Brine samples were collected from a spigot located below the oil/brine separator tank. Each brine sample was collected in an acid-rinsed 4-l polyethylene bottle.

Quality-control data to document the reproducibility of analytical results and any sample contamination were provided by test samples that consisted of either a replicate sample or a field-blank incorporated into the sample set. Quality-control samples comprised about 10% of the total number used for analyses.

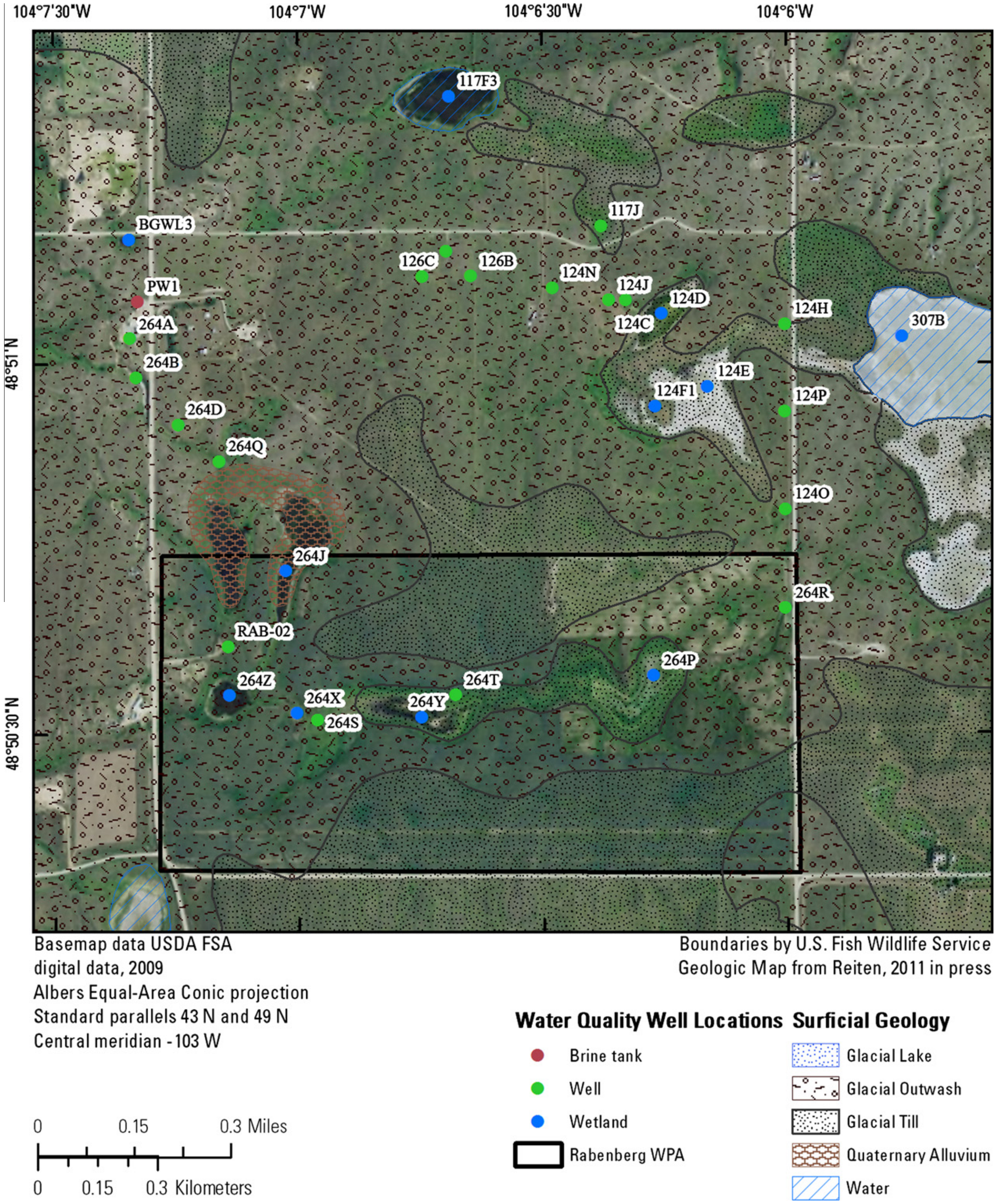

Fig. 2. Map of the Goose Lake study area showing sample locations and locations of oil wells.

Replicate samples were collected in the field by concurrently filling sample bottle sets.

Sample processing, filtration, and preservation were performed in the field using methods employed by the U.S. Geological Survey (variously dated). Samples of surface water and groundwater from shallow wells were collected in acid-washed polyethylene bottles following filtration to 0.45 μm.

In the laboratory, a sufficient volume of water to yield a few μg of Sr was evaporated to dryness in a Pyrex or Teflon beaker. The residue was dissolved in 3 N HNO_3 and passed through an Eichrom Sr-spec resin in Teflon columns to isolate the Sr from other elements, particularly Rb. The Eichrom Sr-spec resin, cleaned by rinsing with many, equal-volumes of purified water over a period of time (about 1 month), had a column blank averaging 0.1 ng. The sample solution collected from the column was dried and dissolved with a drop of 1 N HCL, which was evaporated onto Re filaments and analyzed on an upgraded (Spectromat) Finnigan MAT 262$^{®}$, thermo-ionization mass spectrometer (TIMS). To eliminate

Table 1
Strontium isotope ratios and selected solute concentrations for samples from the Goose Lake study area, Montana. Additional historical analyses are available at <http://steppe.cr.usgs.gov/data.html>. (Major dissolved ions were determined by the USGS National Water Quality Laboratory and trace metals and Sr isotopes, by the isotope laboratory of the former Yucca Mountain Project Branch.)

Site name	Latitude NAD83	Longitude Projection	Site type	Sample date	Field pH	Specific cond. (μS/cm)	Ca (mg/L)	Mg (mg/L)	K (mg/L)	Na (mg/L)	Alkalinity as $CaCO_3$ (mg/L)	Br (mg/L)	Cl (mg/L)	F (mg/L)	SO_4 (mg/L)	I (mg/L)	B (μg/L)	Ba (μg/L)	LI (μg/L)	Mn (μg/L)	Rb (μg/L)	Sr (μg/L)	Contam. Index	^{87}Sr/^{86}Sr Atom ratio
117F	48.8560	−104.1115	Wetland	9/17/2009	9.07	10750	992	459	71.7	1044	84	4.05	2941	0.13	1775	0.034	1103	106	310	19	5.9	3970	0.27	0.71006
117F	48.8560	−104.1115	Wetland	9/17/2009	9.07	10690	967	452	69.4	1010	84	–	2957	0.14	1787	0.034	1037	107	311	37	6.0	3980	0.28	0.71004
124D	48.8511	−104.1042	Well	5/14/2009	8.43	10600	284	171	61	1520	160	4.78	3280	0.17	605	0.2	7150	136	721	48	31.6	4320	0.31	0.70858
124E	48.8495	−104.1027	Wetland	5/14/2009	8.93	22000	306	298	97.9	3910	800	12.5	7020	0.09	729	0.449	14,200	149	1870	37	23.8	5640	0.32	0.70861
124F	48.8490	−104.1045	Wetland	5/14/2009	8.65	14700	332	134	54.5	2150	256	6.98	4870	0.3	299	0.107	7810	222	1140	69	6.7	3170	0.33	0.70884
124J	48.8514	−104.1061	Well	5/14/2009	7.37	17900	463	197	64.8	3060	257	10.7	6150	0.31	137	0.368	10,100	363	1400	1450	84.6	7610	0.34	0.70853
126C	48.8520	−104.1124	Well	5/15/2009	7.23	3630	372	183	13.5	47.3	242	1.87	964	0.15	220	0.039	64	185	121	2100	2	1590	0.27	0.70935
264A	48.8506	−104.1224	Well	5/13/2009	6.92	91300	1370	543	402	18,600	381	57.1	36,300	0.27	512	3.66	71,400	–	9340	6520	476	36,900	0.40	0.70828
264J	48.8453	−104.1171	Wetland	9/17/2009	7.92	9830	345	419	32.2	1076	449	3.32	2811	0.16	693	0.058	608	240	244	243	3.1	1300	0.29	0.71097
264K	48.8425	−104.1190	Wetland	9/17/2009	7.81	13980	648	512	65.6	1508	378	7.25	4738	0.21	418	0.075	1410	453	525	649	6.0	3340	0.34	0.71048
264M	48.8421	−104.1167	Wetland	9/17/2009	8.49	4500	110	128	30.2	551	108	1.19	1270	0.06	242	0.007	768	78	144	81	2.9	992	0.28	0.70991
264Y	48.8420	−104.1125	Wetland	5/12/2009	7.89	1080	85	52.1	19.9	45	190	0.06	120	0.05	181	0.015	199	74	40	17	2.7	431	0.11	0.71042
307B	48.8506	−104.0961	Wetland	5/15/2009	8.83	66600	489	785	263	23,000	800	30.6	4050	0.05	45,300	0.291	5380	585	2050	50	20	6360	0.06	0.70922
BGWL	48.8528	−104.1224	Wetland	5/15/2009	8.43	387	43	8.04	16.7	22	190	0.02	9	0.05	0.83	0.015	258	35	9	2	1.7	130	0.02	0.70898
PW1	48.8514	−104.1222	Brine tank	5/15/2009	–	223000	5030	961	2040	70,500	142	240	121,000	2.65	1230	29.5	230,000	856	31,900	101	4370	16,800	0.54	0.70802

instrumental fractionation, ^{88}Sr/^{86}Sr values were normalized to a value 8.37521, and ^{87}Sr/^{86}Sr were corrected accordingly by a power law. The normalized ^{87}Sr/^{86}Sr ratios were adjusted to a value of 0.709200 for modern sea water based on replicate analyses of Sr in a modern *tridacna* shell (EN-1) collected live from Eniwetok Lagoon in the western Pacific (Ludwig et al., 1988). The average measured ^{87}Sr/^{86}Sr value of EN-1 is 0.709177 ± 0.000003 ($n = 77$), and the value for SRM-987 is 0.710280 ± 0.000007 ($n = 19$). Uncertainties are 2-sigma of the mean. The numerical difference in ^{87}Sr/^{86}Sr between these two standards is 0.001103 ± 0.000008. This difference is close to the value of 0.001074 (no uncertainty given) reported by Chapman et al. (2012). Strontium concentrations were determined on an inductively coupled plasma mass spectrometer (ICPMS) and are accurate to ±10% or better of the value reported.

3. Strontium isotope systematics of groundwater and surface water at Goose Lake

When precipitation enters the ground, it reacts with soil minerals and dissolves Sr with a certain ^{87}Sr/^{86}Sr (Blum and Erel, 2003). The isotopic composition will change by (1) further water–rock interaction during the subsurface residence of the water (McNutt, 2001), and (2) mixing with other waters that differ in ^{87}Sr/^{86}Sr ratios and Sr concentrations (e.g. Woods et al., 2000; Shand et al., 2009). Unlike O and H isotopic compositions, the ^{87}Sr/^{86}Sr will not change during evaporation although Sr concentrations will increase along with those of the other solutes (Peterman and Stuckless, 1992). Precipitation of Sr-bearing minerals will not appreciably change the isotopic composition of the water.

A plot of ^{87}Sr/^{86}Sr against Sr for a series of samples representing different proportions of two end members will define a mixing hyperbola (Faure, 1998, pp. 335–337). Fig. 3 illustrates the effect of different brine (b) and fresh water (w) Sr concentrations $[Sr_b]/[Sr_{fw}]$ on the curvature of the mixing relationships. Mixing of end-members with equal [Sr] produces a linear relationship, but the usual case is where $[Sr_b] \gg [Sr_{fw}]$, which imparts a strong curvature to the mixing line. In the case of $[Sr_b]/[Sr_{fw}] = 100$, not unusual for brine and fresh water in the Williston basin, the change in

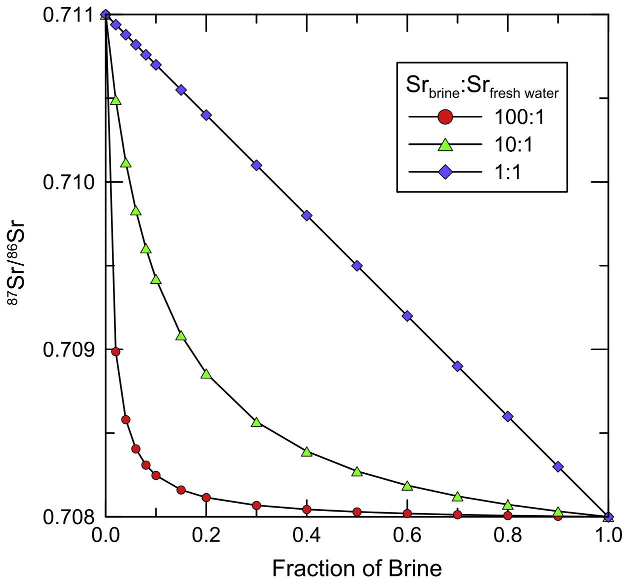

Fig. 3. Graph showing the effects of the Sr concentration ratios in end members on curvature of mixing lines.

$(^{87}Sr/^{86}Sr)_b$ with a large dilution of fresh water has little effect on $(^{87}Sr/^{86}Sr)_M$, whereas a very small amount of brine contaminant in the fresh water component has a large effect on the $(^{87}Sr/^{86}Sr)_M$. This relationship illustrates the value of Sr isotopes in detecting small amounts of brine.

Differences in both Sr concentrations and $^{87}Sr/^{86}Sr$ are optimal at the Goose Lake site for conducting this experiment (Table 1). Strontium concentrations range from 0.13 to 36.9 mg/L, and corresponding $^{87}Sr/^{86}Sr$ values range from 0.71097 to 0.70828. Significant isotopic and concentration contrast is provided by the local brine with 168 mg/L Sr and a $^{87}Sr/^{86}Sr$ value of 0.70802. Strontium isotope ratios are plotted against widely ranging Sr concentrations (Table 1) on a semilog plot (Fig. 4) to illustrate an evaporation-mixing model for Goose Lake samples. The model is illustrated by the graphical inset (Fig. 4). Evaporation will increase solute concentrations, including [Sr] but will not change the $^{87}Sr/^{86}Sr$ values. Any water composition along the "evaporation" line can be influenced by brine, salts derived from brine evaporation, or by previously affected groundwater, and its composition will be drawn downward along a hyperbolic mixing line towards the brine composition. This process can occur in multiple stages and combinations of freshwater recharge, evaporation, and brine input, but the net results are shown by the data in Fig. 4 where a mixing zone is populated by samples representing the resultant isotopic and solute variability.

Three mixing lines connect PW1, the brine sample, with data points that bound the array and extend to the horizontal line of $^{87}Sr/^{86}Sr = 0.71097$ (sample 264J). The distribution of data points in Fig. 4 indicates that in-mixing of brine will always lower the $^{87}Sr/^{86}Sr$ of water at the Goose Lake site. Thus, the value for 264J is selected as the least modified $^{87}Sr/^{86}Sr$ value, and this ratio is used to calculate the mixing lines. In reality, there may well be samples with larger $^{87}Sr/^{86}Sr$ values indicating less brine involvement. There may also be significant $^{87}Sr/^{86}Sr$ variability in the unaffected groundwater reservoir. Neither of these possibilities detracts from the concept of the evaporation-mixing model. The two solid curves bound the data array with PW1-BGWL intersection at 0.71097 at 0.043 mg/L Sr, and PW1-264A at 4.138 mg/L Sr. The dashed mixing line is calculated through PW1 and 264Y to intersect the line at 0.71097 at 0.351 mg/L Sr. All but BGWL, a sample collected from a wetland containing recently-melted snow that is very low in solutes, are contained within the dashed curve and the solid curve passing through 0.71097 and 4.138 mg/L Sr.

The $^{87}Sr/^{86}Sr$ ratio of PW1 is similar to Sr isotope values for produced water from the Weyburn field in southern Saskatchewan (Quattrocchi et al., 2006) and to brines that affected groundwater in the Ft. Peck Indian Reservation in eastern Montana (Peterman et al., 2012). All of these brines were produced from the Mississippian Madison Group carbonate rocks. Data for the Weyburn brines are plotted in Fig. 4. The differences in Sr concentrations between that of PW1 and the lower values for the Weyburn samples may be the result of dilution of the latter during previous flooding of the reservoir with water from the Cretaceous Mannville aquifer with only a small effect on the $^{87}Sr/^{86}Sr$ values of the brine (Quattrocchi et al., 2006).

The sensitivity of Sr isotopes in detecting small amounts of brine can be estimated by considering sample 264Y. As mentioned above, the dashed mixing line passing through PW1 and 264Y with 0.431 mg/L Sr intersects the horizontal line passing through 264J at 0.351 mg/L Sr. Only 0.05% brine (PW1) is required to move 264Y from the hypothetical original position at 0.351 and 0.71097 to its measured position in Fig. 4. Similarly, sample 264A could have been displaced from an original value on the 0.71097 line to the measured value of 0.70828 by only 20% brine.

The contamination index (CIx) is calculated for each sample in Table 1 using laboratory measurements of Cl^- and specific conductance. CIx values for earlier sample collections from many of these sites are given by Reiten and Tischmak (1993). They considered CIx values of less than 0.035 to indicate uncontaminated water. One sample in Table 1, BGWL, has a CIx value lower than 0.035. In

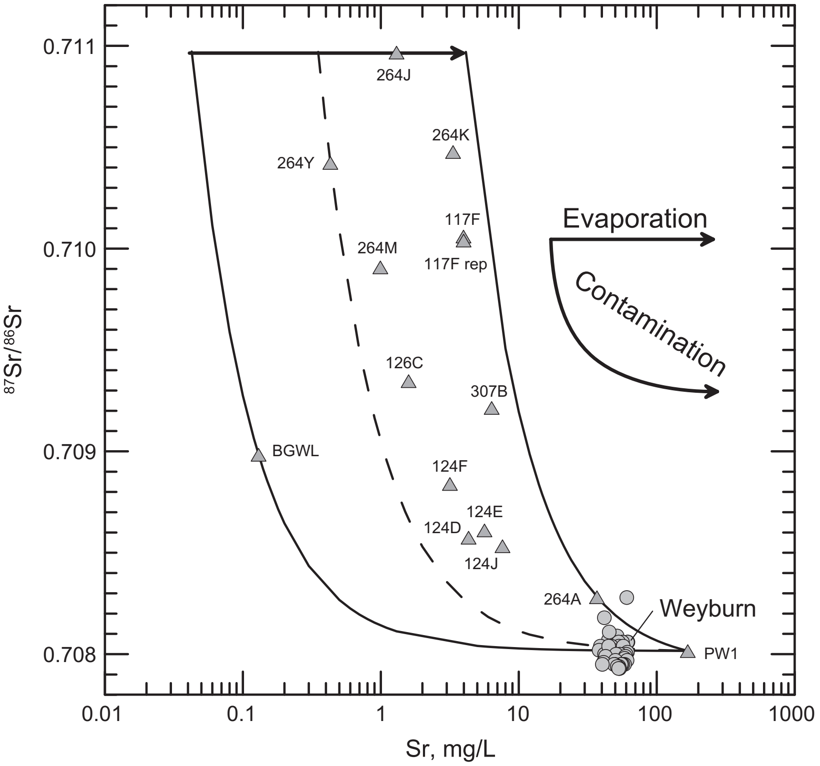

Fig. 4. Evaporation-mixing model for Sr isotope variation in surface and near-surface water at Goose Lake.

the binary mixing model described above, only 0.005% PW1 would be required to move BGWL from its hypothetical original composition at 0.043 mg/L Sr and a $^{87}Sr/^{86}Sr$ ratio of 0.71097 to its present position in Fig. 4. The relationship between CIx and the component of brine in other samples indicated by their location in Fig. 4 is complicated. The brine, PW1, has a CIx of 0.54, the largest value in the suite of samples. Sample 264Y, which has the largest component of brine based on the Sr model, has a CIx of 0.40. The cluster of samples 124D, E, F and J have similar CIx values of 0.31 to 0.34. A marked departure from the general relationship is shown by 307B with a CIx of 0.06. This sample is high in SO_4^{2-} and Na^+ resulting in a large specific conductance, but Cl^- concentration is not especially enriched relative to many of the other samples.

Concentrations of other dissolved ions are also given in Table 1. Except for HCO_3^- and SO_4^{2-}, the relationships of these concentrations to $^{87}Sr/^{86}Sr$ values are generally consistent with the evaporation-mixing model. Neither HCO_3^- nor SO_4^{2-} are necessarily conservative so departure from the model is not surprising.

4. Conclusions

As previous studies have shown, surface water and shallow groundwater at Goose Lake in northeastern Montana, have been variably affected by oil-field brines directly or through dissolution of salt deposits formed from the brines. Strontium concentrations in these waters and in oil-field brines differ by several orders of magnitude, and $^{87}Sr/^{86}Sr$ values are significantly different. An evaporation-mixing model is proposed. Seasonal evaporation increases solute concentrations but $^{87}Sr/^{86}Sr$ values of the water are not changed. An increasing brine component increases Sr and other solute concentrations and lowers the $^{87}Sr/^{86}Sr$ values. This mixing produces predictable relationships that can be used to detect very small amounts of brine.

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