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**Electronic supplementary material *for* Groundwater movement, recharge, and perchlorate occurrence in a faulted alluvial aquifer in California (USA)**

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# Groundwater movement, recharge, and perchlorate occurrence in a faulted alluvial aquifer in California (USA)

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Electronic supplementary material – Hydrogeology Journal

## 1. Field and laboratory methods, and quality assurance data

Most production wells were sampled from the surface discharge of the existing pump. Monitoring wells were sampled using a 4.5-centimeter (cm) diameter, positive displacement gas-reciprocating pump capable of lifting water from depths greater than 200 m below land surface. Additional samples were collected from WestBay installations using specialized equipment according to manufacturer's specifications. Production and monitoring wells were purged to remove at least 3 casing volumes prior to sample collection. Field parameters (pH, temperature, and specific conductance) were monitored during purging using a thermometer and portable meters. After field parameters stabilized (three repeated samples within a 15 minute period were within 5 percent), alkalinity was measured by titration and dissolved oxygen was measured using the indigo-carmin method (CHEMetrics, Inc., Calverton, VA).

### 1.1 Coupled well-bore flow and depth-dependent sample collection

Coupled well-bore flow logs and depth-dependent samples were collected from seven wells distributed along two sections through the study area: 1) Section A-A', extends along the axis of the eastern perchlorate plume within the Rialto-Colton subbasin, and 2) Section B-B' extends from the Chino subbasin to the Rialto-Colton subbasin perpendicular to the Rialto-Colton Fault and the two plumes (Fig. 1 in manuscript). Natural-gamma and caliper logs were collected from these wells under unpumped conditions to provide geologic information and to confirm well construction data. Fluid resistivity, fluid temperature, and well-bore flow logs were collected from these wells under unpumped and pumped conditions to provide information on well yield and water quality with depth in the well. Production pumps were removed from the wells prior to logging to allow entry for the geophysical tools and flowmeter.

One of two temporary submersible pumps, having a maximum capacity of either 9.5 or 16.5 liters per second (L/s) and capable of lifting from more than 200 m below land surface, was installed for data collection under pumped conditions (the 9.5 L/s pump was used as a backup if the 16.5 L/s pump was not operational). In most cases, data collected using the temporary pumps were considered comparable to conditions within the wells imposed by the higher production rates of the production pumps. Instantaneous and cumulative discharge from the temporary pumps was measured at the surface using a sonic flowmeter. In some cases, water pumped from the well during well-bore flow logging and depth-dependent sample collection was treated to remove perchlorate and organic contaminants prior to discharge. Permits were obtained from appropriate agencies prior to discharge of water.

Well-bore flow data were collected from production wells using an electromagnetic (EM) flowmeter (Young and Pearson, 1995). The EM flowmeter measures the voltage generated by an electrical conductor (water) passing through the inside of a hollow, cylindrical section within the flowmeter that is surrounded by electromagnets (Young and Pearson, 1995; Paillet, 2005). The EM flowmeter has no moving parts and a large dynamic range, capable of measuring velocities ranging from <0.1 to 82 meters per minute (m/min) (Newhouse, et al., 2005). Unpumped and pumped wellbore flow data were collected in the downward

direction at three trolling speeds (approximately 1.5, 3, and 4.5 m/min). Centralizers were used, but diverters were not used to allow passage of the tool around the temporary pumps.

Unpumped flow-log data, collected at each of the three trolling speeds, were plotted with depth to ensure the EM tool response increased proportionately with tool speed throughout the well. The average tool response in counts per second from an unscreened section of the well below the water table and above the uppermost screened interval where no flow was present was related to the trolling speed using linear regression (Fig. S1). The regression equation developed from these data was used to estimate unpumped flow throughout the well in m/min. Well diameter obtained from caliper log data was used to calculate the cross-sectional area of the well and convert well bore flow measured in length per time (L/t) to volume per time (L<sup>3</sup>/t). The precision of unpumped flow logs collected as part of this study was estimated as ± 1 standard deviation (σ) of the tool response measured in the unscreened section of the well.

Pumped flow-log data, collected at each of the three trolling speeds, were plotted with depth to ensure the EM tool response increased proportionately with tool speed throughout the well. Two regression equations were developed relating tool response, in counts per second, throughout the entire well for trolling speeds of 1.5 and 3 m/min, and for trolling speeds of 1.5 and 4.5 m/min (Fig. S2a). For a linear tool response, both regression lines would have a slope of 1 and the difference between the two regression equations would be independent of the tool trolling speed. The EM flowmeter response was generally non-linear over the range of flows encountered in pumped wells, and the tool response in counts per second generally increases with increased flow. To correct for non-linearity, the difference between the two regression lines (1.5 and 3 /min and 1.5 and 4.5 m/min) at a given tool response was used to develop the regression equation used to estimate pumped flow throughout the well in meters per minute (Fig. S2b). Without using this approach, EM tool non-linearity would have resulted in as much as a 20 percent error in measured flow within the well. This approach also was used for unpumped flow in wells where an unscreened section above the well screens, having no flow, was not present.

For most wells, depth-dependent water-quality samples were collected from wells under pumping conditions using a 2.5-cm diameter gas-displacement pump using procedures described by Izbicki (2004). For Rialto-2, depth-dependent samples were collected using a commercially available wire-line bailer to ensure volatile organic carbon samples collected at this site were not stripped from the water during sample collection. The gas-displacement pump was flushed with sample water, and the wire-line bailer was rinsed with deionized water prior to sample collection. For depths where large volumes of water for analysis of chlorine and oxygen isotopic composition of perchlorate also were collected (not discussed in this paper) samples were collected under pumping conditions from new 5-cm diameter PVC pipes emplaced in the well at the sample depth. Sample water was pumped from the pipes using a 4.5-cm diameter, positive-displacement, gas-reciprocating piston pump.

The chemistry and isotopic composition of water entering the well between two sample depths was calculated on the basis of measured changes in flow ( $V$  in length cubed per unit time,  $L^3/t$ ) and concentration ( $C$  in mass per length cubed,  $M/L^3$ ) according to the following (Izbicki, et al., 1999; and Izbicki, 2004):

$$(C_3 \cdot V_3) = (C_1 \cdot V_1) + (C_2 \cdot V_2),$$

$$\text{rearranged to: } C_2 = ((C_3 \cdot V_3) - (C_1 \cdot V_1)) / V_2$$

Subscripts in the equations refer to sample depth; 1 is the measurement point at the bottom of the sample interval, 2 is the interval between the sample points, and 3 is the measurement point at the top of the sample interval.  $V_2$  is calculated from measured flow data as:

$$V_2 = V_3 - V_1.$$

## 1.2 Sample handling and preservation

Water samples for selected anions, cations, and nutrients were filtered in the field through 0.45 micrometer pore-sized filters, placed in plastic bottles, and chilled. Samples for cation analysis were preserved in the field using nitric acid. Samples for analysis of stable oxygen and hydrogen isotope ratios were unfiltered and

placed in glass bottles. Samples for tritium were unfiltered and placed in 1 liter (L) plastic bottles. Samples for analysis of carbon-14 and stable carbon isotope ratios were field filtered and placed in 1 L glass bottles. Nutrient samples were shipped from the field to the U.S. Geological Survey National Water Quality Laboratory (NWQL) in Denver, Colo. (<http://wwwnwql.cr.usgs.gov/USGS/>, accessed October 29, 2014) within 24 hours of collection. Major-ion, minor-ion, and trace-element samples from most wells were shipped to the NWQL for analysis within their respective holding times. Samples for nutrient, major-ion, minor-ion, trace element and perchlorate analyses for Rialto-2 were preserved on ice and delivered to U.S. EPA contractors on site and shipped within their respective holding periods to the U.S Environmental Protection Agency laboratory in Richmond, Calif.

Stable oxygen and hydrogen isotope ratio samples were shipped to the USGS Reston Stable Isotope Laboratory (RSIL) in Reston, Va. (<http://isotopes.usgs.gov/>) for analysis. Tritium samples were shipped to the USGS laboratory in Menlo Park, Ca. (<http://wwwrcamnl.wr.usgs.gov/isoig/res/kendall.html>, accessed October 29, 2014) for analysis. Carbon-14 and stable carbon isotope ratio samples were shipped on ice to the NWQL for analysis by a commercial laboratory under contract with the USGS. Dissolved gas samples were collected in triplicate in 125 milliliter (mL) glass-serum bottles. The bottles were submerged in a clean bucket and sealed without headspace using rubber stoppers according to laboratory instructions (<http://water.usgs.gov/lab/dissolved-gas/sampling/>, accessed October 29, 2014)) and shipped to the USGS dissolved-gas laboratory in Reston, Va. (<http://water.usgs.gov/lab/dissolved-gas/>, accessed October 29, 2014) for analysis. Samples for analysis of nitrogen and oxygen isotopes in nitrate were filtered and collected in 125 mL bottles with KOH as preservative. Samples for analysis of sulfur and oxygen isotopes of sulfate were filtered in the field and collected in 1 L bottles with no preservative. Samples for nitrogen, sulfur, and oxygen isotopes of nitrate and sulfate were shipped to the USGS Reston Stable Isotope Laboratory (RSIL) in Reston, Va. (<http://isotopes.usgs.gov/>, accessed October 29, 2014) for analysis.

Samples for perchlorate analyses were field filtered through 0.2  $\mu\text{m}$  pore-sized filters, preserved on ice, and placed in 125 mL plastic bottles. Samples were shipped to Shaw Environmental, in Lawrenceville, New Jersey, for analyses. Samples for perchlorate from Rialto-2 also were analyzed by Shaw Environmental and are the values reported in this paper.

### 1.3 Laboratory methods

Samples for major-ion, selected minor-ion, and trace-element analyses were analyzed by various methods (Fishman and Friedman 1989; Fishman 1993; and Garbarino et al 2002 and 2006). Nitrogen and phosphate species were measured by automated colorimetric methods (Fishman 1993; Patton and Truitt 1992 and 2000), or Kjeldahl digestion with automated finish (Fishman 1993; Patton and Truitt 1992). Perchlorate was analyzed by ion-chromatography by Shaw Environmental, Lawrenceville, NJ using US EPA Method 314.0 (Hautman et al 1999). Concentrations of perchlorate below the detection limit for that method (0.5  $\mu\text{g/L}$ ), were analyzed at Texas Tech University in Lubbock, Tx, by Ion-Chromatography Mass-Spectrometry (ICMS) (US EPA Method 332.0) which has a detection limit of 0.1  $\mu\text{g/L}$  (Hedrick et al 2005).

The ratio of oxygen-18 ( $^{18}\text{O}$ ) to the more common isotope oxygen-16 ( $^{16}\text{O}$ ) was measured using the carbon dioxide ( $\text{CO}_2$ ) equilibration technique (Epstein and Mayeda 1953). The ratio of hydrogen-2, also known as deuterium ( $^2\text{H}$  or D), to the more common isotope hydrogen-1 ( $^1\text{H}$ ) was measured using a hydrogen gas equilibration technique at 30  $^\circ\text{C}$  (Coplen, et al., 1991). Oxygen and hydrogen isotopic results are reported in delta notation ( $\delta$ ) as per mil (‰) differences relative to VSMOW (Vienna Standard Mean Ocean Water) according to the following:

$$\delta^{18}\text{O} \text{ or } \delta^2\text{H} \text{ in per mil} = [(R_{\text{sample}} - R_{\text{standard}}) / R_{\text{standard}}] \times 1,000$$

where  $R_{\text{sample}}$  and  $R_{\text{standard}}$  refer to the ratio in the sample and the standard, respectively. By convention, the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values of the standard VSMOW are 0 per mil. Results were normalized on scales such that the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values of Standard Light Antarctic Precipitation (SLAP) are  $-55.5$  per mil and  $-428$  per mil, respectively (Gonfiantini 1978; Coplen 1988). Tritium was measured by liquid scintillation (Thatcher et al 1977) with electrolytic enrichment (Ostlund and Warner 1962). Carbon-14 was analyzed by accelerator mass spectrometry (Beukens 1992). Carbon-13 analyses were done by mass spectrometry on dissolved inorganic

carbon (DIC) extracted from filtered water samples (Thatcher et al 1977). Dissolved nitrogen and argon gas concentrations were analyzed using a Hewlett Packard model 5890 gas chromatograph with helium as the carrier gas. The gas was injected into an Alltech CTR-III column at 55°C to separate argon, nitrogen, and oxygen, and concentrations were quantified with a thermal conductivity detector. Instrument calibration, analytical procedures, and reporting levels are described in [http://water.usgs.gov/lab/dissolved-gas/lab/analytical\\_procedures/](http://water.usgs.gov/lab/dissolved-gas/lab/analytical_procedures/) (accessed October 29, 2014).

Isotope ratios of nitrogen and oxygen in nitrate were determined by the bacterial reduction method in which samples are cultured with *Pseudomonas aureofaciens* to reduce nitrate and nitrite to nitrous oxide (N<sub>2</sub>O) for isotope-ratio mass spectrometry (Sigman et al 2001; Casciotti et al 2002; Coplen et al 2004). Replicates of each sample were analyzed in different batches with isotopic reference materials, giving 2 sigma reproducibility of less than ± 0.5 ‰ and ± 1 ‰ for δ<sup>15</sup>N and δ<sup>18</sup>O, respectively (typically 2–5 replicates). For samples having measurable nitrite (such as PW-5D), the measured apparent δ<sup>18</sup>O value may be incorrect because of a known calibration artifact of mixed (nitrate plus nitrite) samples (Böhlke et al, 2007). For sulfate isotopic analyses, samples were acidified with HCl and treated with BaCl<sub>2</sub> to precipitate BaSO<sub>4</sub>, which was filtered and dried. BaSO<sub>4</sub> was analyzed for δ<sup>34</sup>S by thermal reaction to SO<sub>2</sub> for mass spectrometry (Carmody et al 1998), and for δ<sup>18</sup>O by thermal reaction to CO for mass spectrometry (Böhlke et al 2003; Brand et al 2009). Isotopic ratios of nitrogen and oxygen in nitrate and sulfur and oxygen in sulfate are expressed in delta notation as δ<sup>15</sup>N-NO<sub>3</sub>, δ<sup>18</sup>O-NO<sub>3</sub>, δ<sup>34</sup>S-SO<sub>4</sub> and δ<sup>18</sup>O-SO<sub>4</sub>, respectively. Results were normalized to be consistent with reference values for δ<sup>15</sup>N-NO<sub>3</sub>, δ<sup>18</sup>O-NO<sub>3</sub>, and δ<sup>18</sup>O-SO<sub>4</sub> summarized in Böhlke and Coplen (1995) and Böhlke et al. (2003) and reference values for δ<sup>34</sup>S-SO<sub>4</sub> summarized in Coplen et al. (2002).

## 1.4 Quality assurance

Major-ion data (including ammonia and nitrate concentrations) had a mean positive charge imbalance of 1.8 percent. Given a ± 2 σ of 2.8 percent cation/anion charge imbalance this was not statistically different from zero. Analyses of major ions from five duplicate samples collected as part of this study agreed within 0.2 to 2.8 percent of the average of the five samples for each constituent. Variability in major-ion concentrations was least for chloride and greatest for alkalinity. Minor-ion and trace-element data for the five duplicate samples agreed within 0.5 to 12 percent. Variability was least for boron and greatest for lithium. With the exception of lithium, variability for minor ions and trace elements was less than 5 percent. Blank samples collected from sample pumps three times during the study showed no evidence of contamination in major-ion or minor-ion data that would affect data interpretation. Perchlorate concentrations in the sample blanks were less than detection limit of 0.5 µg/L using ion-chromatography. Blank samples were not analyzed for perchlorate by ICMS. As a consequence, low perchlorate concentrations between 0.1 and 0.5 µg/L within field blanks would not have been detected.

Dissolved-gas samples were collected in triplicate. Initially all three triplicate samples were analyzed, after 16 sample sets were analyzed, and the suitability of field sample collection procedures was established, only two of the three replicate gas samples were analyzed. If a problem was observed with the duplicate analysis, the third replicate was analyzed. For nitrogen gas analysis, the difference between triplicate (or duplicate) samples ranged from less than 0.05 to 3.5 percent of the average value, with 78 percent of samples having a percent difference of 1 percent or less. For argon gas analysis, difference between triplicate (or duplicate) samples ranged from less than 0.05 to 7.2 percent of the average value, with 83 percent of samples having a percent difference of 1 percent or less.

As part of sample collection for chloride and oxygen isotopic of perchlorate, 190 samples collected from the inflow to 28 resin columns (or manifolds for multiple columns) were analyzed for perchlorate and specific conductance. Wells were pumped from 2 hours to almost 100 hours, depending on the length of time needed to accumulate at least 5 milligrams of perchlorate on the column for isotopic analysis. (Samples from wells F-4, Chino-2, and F-18a that were pumped to tanks, and the water subsequently drained through resin columns, were excluded from this analysis.) The number of samples collected for each column (or manifold of several columns) ranged from 3 to 17, with a median of 6.5 samples per column.

Perchlorate concentrations in water from wells varied with time during sample collection in the perched and regional aquifers within the Rialto-Colton subbasin and in the Chino subbasin, with a median  $1\sigma$  variability of 47, 3, and 5 percent of the average perchlorate concentration for the column, respectively (Fig. S3). The median  $1\sigma$  variability in specific conductance was 12, 2, and 1 percent, respectively (not shown in Fig. S3). Variations in perchlorate concentrations were not statistically correlated with perchlorate concentration, and variability did not increase or decrease consistently as perchlorate concentrations increased or decreased, but were inversely correlated with specific conductance (Spearman rank correlation coefficient of  $r = -0.79$ ). Changes in perchlorate concentrations and specific conductance during pumping appeared random, with no wells showing consistent increases or decreases in perchlorate concentrations or specific conductance with time.

The median variability in perchlorate concentrations and specific conductance with time was greatest in the perched aquifer. However, the smaller number of sampled wells and columns collected in the perched aquifer limited the statistical comparison of those data with data from other areas. However, the greater variability in the perched aquifer may reflect conditions within the aquifer because it is closer to surface sources of contamination.

The  $1\sigma$  variability in perchlorate concentrations with time in water from wells within the Chino subbasin was statistically different from the  $1\sigma$  variability in water from wells in the Regional aquifer in the Rialto-Colton subbasin (Fig.S3). There was no statistically significant difference in the variability of specific conductance with time in water from wells in these aquifers. Wells in the Chino subbasin with the greatest variability in perchlorate concentrations were wells Chino-2 and F-26A (surface discharge samples only). Water from these wells had a  $1\sigma$  variability of 69 and 14 percent of the average perchlorate concentration (average of 2 columns from each well), respectively. Both wells are located between the Rialto-Colton fault and the western Rialto-Colton fault. Perchlorate data from the depth-dependent sample collected from Rialto-6 at 170.7 m below land surface were the most variable within the Regional aquifer in the Rialto-Colton subbasin, with a  $1\sigma$  variability of 10 percent of the average concentration.

## 2. Other material

Tables S1, S2, S3, S4, S5, and Figures S4 and S5 cited in the manuscript text are provided at the end of the Electronic Supplementary Material (ESM) within this document.

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