

Interactions of Solutes and Streambed Sediment

1. An Experimental Analysis of Cation and Anion Transport in a Mountain Stream

KENNETH E. BENCALA, VANCE C. KENNEDY, GARY W. ZELLWEGER,
ALAN P. JACKMAN,¹ AND RONALD J. AVANZINO

U.S. Geological Survey, Menlo Park, California

An experimental injection was performed to study the transport of stream water solutes under conditions of significant interaction with streambed sediments in a mountain pool-and-riffle stream. Experiments were conducted in Little Lost Man Creek, Humboldt County, California, in a period of low flow during which only a part of the bank-full channel held active surface flow. The injection of chloride and several trace cations lasted 20 days. In this report we discuss the results of the first 24 hours of the injection and survey the results of the first 10 days. Solute-streambed interactions of two types were observed. First, the physical transport of the conservative tracer, chloride, was affected by intergravel flow and stagnant water zones created by the bed relief. Second, the transport of the cations (strontium, potassium, and lithium) was appreciably modified by sorption onto streambed sediment. In the stream the readily observable consequence of the solute-streambed interactions was an attenuation of the dissolved concentration of each of the tracers. The attenuation in the stream channel occurred concurrently with the storage of tracers in the streambed via both physical and chemical processes. All tracers were subsequently present in shallow wells dug several meters from the wetted part of the channel. Sediment samples collected approximately 3 weeks after the start of the injection contained increased concentrations of the injected cations.

INTRODUCTION

The sorption of cations onto solid surfaces is a well-recognized physical chemical phenomenon [Stumm and Morgan, 1981]. Solute sorption onto sediment is an important factor in determining the distribution of various solutes within a watershed [Kennedy, 1965] and [Kennedy et al., 1984]. The role of cation sorption in streams receiving anthropogenically released radionuclides [Cerling and Turner, 1982; Cerling and Spalding, 1982] and acidic deposition [Hall and Likens, 1981] has been previously observed. Within a stream channel the dynamic and tightly coupled relationships between physical and chemical mechanisms must be understood in order to interpret the transient behavior of many chemical substances.

Small, pool-and-riffle, gravel bed streams provide natural laboratories for investigations of the interactions of solutes and streambed sediments. The shallow but well-mixed cross sections in these streams allow for physical contact between the solutes and solids. The relatively long travel times permit slow reactions to proceed to an observable extent. Finally, low discharge makes it possible to alter the stream chemistry experimentally using moderate amounts of tracers.

In this report (part 1 of two papers) we present an interpretation of the data from an experimental injection of chloride and the trace cations strontium, lithium, and potassium into a mountain stream. The data support the conclusions that (1) there is significant hydrologic interaction between solutes in the stream channel and the surrounding streambed and (2) there is the potential for significant chemical interaction between solutes and streambed solids. In part 2 [Bencala, this issue] we will develop a dynamic analysis of the coupled

physical and chemical processes responsible for the observed tracer behavior.

FIELD WORK

Site

The stream used for the field experiment was Little Lost Man Creek in Redwood National Park near the coast (see Figure 1) of northwestern California. It is a pool-and-riffle stream draining a 9.4 km² mountainous area [Iwatsubo et al., 1975]. During late August and early September, when this study was done, streamflow normally has decreased to a low level which varies slightly during the day due to evapotranspiration.

The stream sediments are poorly sorted and commonly range from sand to boulder size, intermingled with woody fragments ranging from twigs to large logs. Debris dams formed of logs and/or gravel are common [Keller and Tally, 1979]. There is relatively little clay and silt deposited in the streambed, thus the debris dams, gravel bars, and riffle deposits are permeable, permitting a considerable amount of interflow through the sediments. During low flow a significant fraction of the stream discharge can be such interflow. The water percolating through the gravels has an opportunity for long and intimate contact with the sediment particles, which allows ample time for chemical interactions to occur.

The basin is underlain by highly lithified, but mostly unmetamorphosed, marine sedimentary rocks, largely graywacke, and sandstone with lesser amounts of mudstone and conglomerate [Janda et al., 1975; Harden et al., 1981]. During weathering, the rocks break down to form a sandy, gravelly soil with some silt and clay present. The sand and gravel-sized soil particles commonly are composed of mudstone or sandstone aggregates in which some permeability is present, mainly in a weathering rind. Thus the sand-sized and larger particles, when washed into the stream, offer considerable surface area for chemical reactions.

The clay mineralogy of the <2 μ m size fraction of sedi-

¹Now at Department of Chemical Engineering, University of California, Davis.

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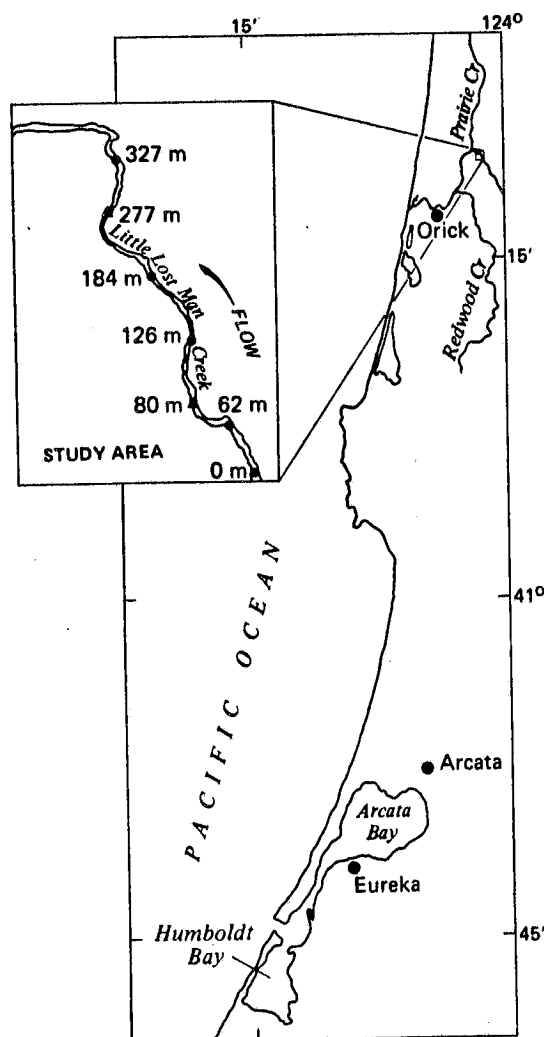


Fig. 1. Location of Little Lost Man Creek, Humboldt County, California. Location of study reach noted on map. Location of sampling points noted on insert.

ments from various streams in northern California has been studied [Griggs and Hein, 1980] and found to contain major amounts of chlorite + kaolinite and illite with minor amounts of smectite. Little Lost Man Creek is similar to the larger streams with about 55% chlorite + kaolinite, 45% illite, and a trace of smectite (J. R. Hein, personal communication, 1982).

Figure 2 is a sketch of the stream profile. Although it is evident from the stream profile that there is sufficient hydraulic head to drive water through the permeable gravels from pool to pool subparallel to the riffles, there are only a few places where the return of that interflow to the surface is readily visible.

At 80 m downstream of the injection point, water flows out of the stream gravels and mixes with a tiny rivulet coming down the hillside before joining the main stream. The flow at 80 m is separated laterally from the creek by a distance of about 5 m of permeable stream gravels and is approximately 1.3 m lower in elevation than the outlet of the next large pool upstream at 35 m. Examination of the downstream edge of the pool near 35 m showed that some water was flowing into several tiny pools which had no surface outlets, indicating that there was percolation into the gravels at that point. It seems evident that a part of the stream was flowing approximately

45 m through the gravels between the bottom of the large pool at 35 m and the flow at 80 m.

At 277 m downstream a trickle of water was observed entering from a side channel active during higher flows. The upstream end of this side channel appears to have once diverged from the main stream near 207 m but is now blocked by a large fallen tree near 207 m. The space between the present channel and the side channel is filled with unconsolidated gravels which would allow percolation of stream water. The elevation difference between 207 and 277 m is about 1.5 m, but the distance of intergravel flow is uncertain and probably quite variable, as the distance between the present stream and the side channel ranges from a few meters to several tens of meters.

Experimental Procedures

The experiment described herein was done as part of more extensive research concerned with the interaction of strongly sorbing cations and nutrients with stream sediments. A solution of chloride, nitrate, and the sorbing cations, lithium, potassium, strontium, and sodium was injected into Little Lost Man Creek at a known constant rate, and water samples were collected at various sites downstream and also in shallow wells. The injection started at 1000 August 21, 1978, and continued for 20 days. Concentrations of the solutes of interest are listed in Table 1.

Standard methods, previously outlined [Bencala et al., 1983], were used for the analysis of dissolved chloride and cation samples and for the determination of cation sorption onto sediment. The shallow "wells" referred to in this report were small pits dug into the unwetted part of the bank-full channel. These pits were 0.1–1.0 m deep and located between 0.5 and 5 m from the edge of the active surface flow. The sediment samples were collected by wet sieving in the stream over a period of several hours. The well and sediment samples provide indications of the trends and processes occurring in and adjacent to the stream system but do not permit quantitative evaluation of tracer distributions.

EXPERIMENTAL RESULTS

We present here data collected during the field experiment. The main purpose of this report is to document, with field data, the extensive hydrologic and chemical interactions between water in the stream channel and the surrounding

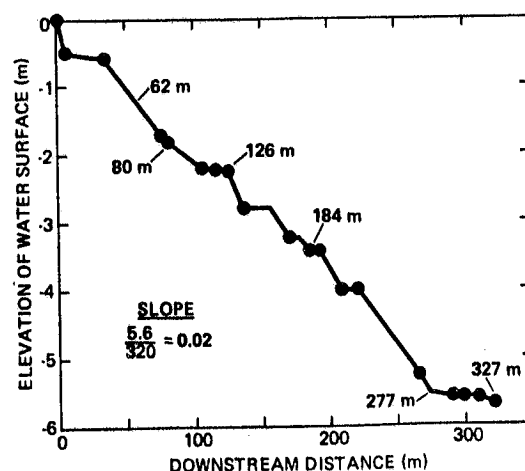


Fig. 2. Profile of study reach of Little Lost Man Creek.

streambed. This report also provides the data base for the analysis of the dynamic stream processes which follows in the next report [Bencala, this issue].

First, summary chloride and cation data are presented from the initial 10 days of the experiment. As will be discussed below, this was a period of relatively steady flow and background chemical conditions. Also, during this period, chloride concentrations reached plateau levels throughout the study reach; thus indicating that the major transport processes had reached a steady state. Next, data collected during the initial 24 hours of the experiment are presented. This is the most important period of the experiment. As a result of the injection, the chemistry of the stream is in a transient state. It is in this period that we can best observe the process dynamics of the system. As stated above, limited data from the shallow wells and sediment samples are also presented. In retrospect, it is clear that both the intensity and timing of these data collections were less than ideal. However, clear patterns of behavior are evident. Data from the wells are presented for the first and tenth days. The sediment data are presented for the end of the experiment.

Solute Data: Initial 10 Days

The chloride data shown in Figure 3a provide the framework for the analysis of the solute transport experiment. Chloride data for the first 10 days of the experiment are shown at three monitoring locations. The sampling frequency varied during the experiment and from location to location. Frequent samples less than 15 min apart were taken over the tracer arrival period at several locations. Later in the experiment, sampling was limited to irregular intervals 5 times a day. The first location was immediately upstream of the solute injection point. Relative to the magnitude of the solute injection there is no significant variation in the background chloride concentration entering the study reach. The second location was 5 m downstream of the injection. This location was at the end of a narrow (approximately 0.2 m wide) mixing channel assembled from streambed cobbles. The data from this location show two distinct patterns. First, there is a diurnal cycle of up to 2 mg L^{-1} . Second, there is a slow trend of increasing concentration of several milligrams per liter extend-

TABLE 1. Little Lost Man Creek Water Constituents Before and During Injection

Constituent	Concentration Before Injection, mg L^{-1} *	Concentration During Injection,† mg L^{-1} *
Chloride	6.2	21.6
Lithium	<0.005	1.9
Sodium	6.4	8.5
Potassium	0.6	2.5
Strontium	0.1	2.0
pH,* pH units	7.4	7.2
Calcium	5.4	N.D.
Magnesium	2.0	N.D.
Silica	7.8	N.D.
Sulfate	2.6	N.D.
Fluoride	<0.1	N.D.
Alkalinity (total lab as CaCO_3)	28.	N.D.

N.D., not determined.

*Except as noted in constituent column.

†Approximate values; a diurnal fluctuation in streamflow and constant injection rate caused diurnal variations in concentration of the injected species.

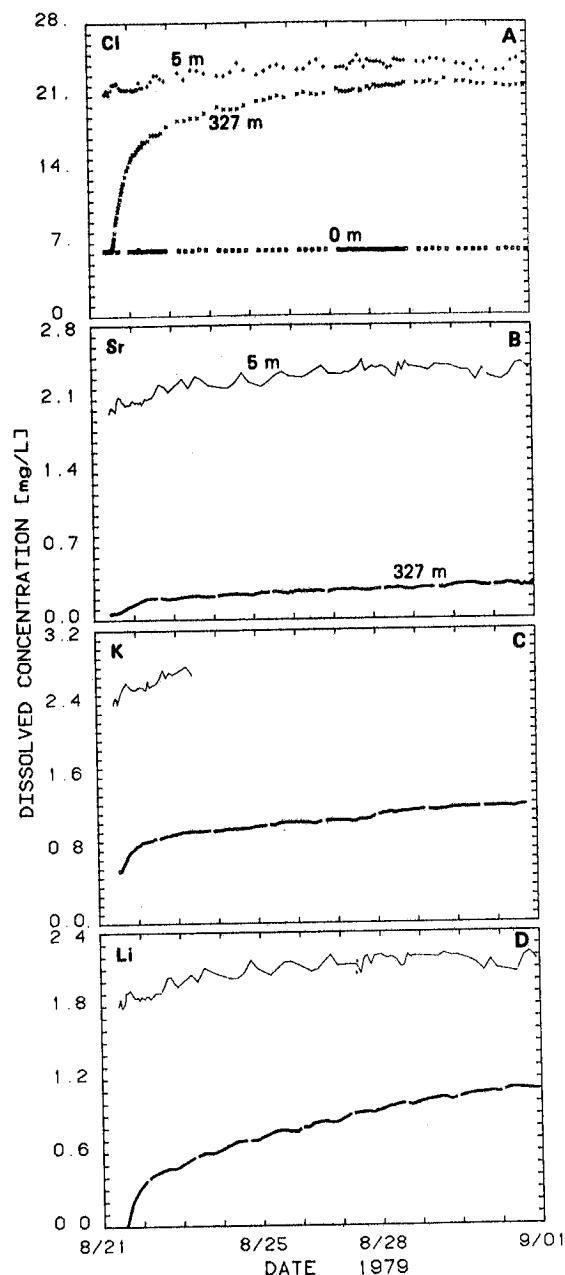


Fig. 3. Observed tracer concentrations for steady injection at Little Lost Man Creek. Data presented for the first 10-day period (plateau period) of the injection (a) Chloride. Sampling frequency was irregular. Individual symbols are shown to indicate the sampling intensity. (b) Strontium. (c) Potassium. (d) Lithium.

ing over the 10-day period. Both these patterns result from changes in stream discharge; as discharge decreases, chloride concentration increases. Discharge peaked in the early morning and slowly decreased through the late afternoon. There was no rain during the 10-day period, and average daily discharge was slowly decreasing. The third location, 327 m, was at the end of the study reach. The solute pulse at this location can be viewed as occurring in three phases; the arrival phase from midday on August 21 until midday on August 22, the storage phase until approximately August 29, and finally a steady phase through August 31. The arrival of the pulse is distinct and, at least initially, appears to follow the form of a standard convective-dispersive pulse [see Fischer et al., 1979].

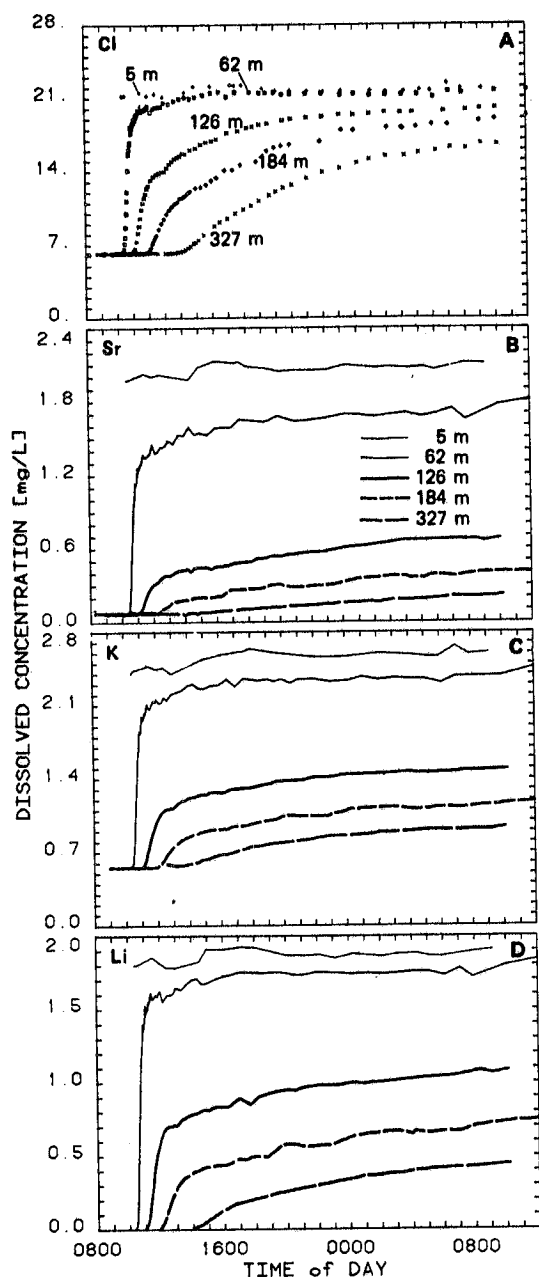


Fig. 4. Observed tracer concentrations for steady injection at Little Lost Man Creek. Data presented for the first 1-day period, transient rise period, of the injection. (a) Chloride. Individual symbols are shown to indicate the sampling intensity. (b) Strontium. (c) Potassium. (d) Lithium.

If, in fact, convection and dispersion were the only transport mechanisms operating in Little Lost Man Creek, we would expect the concentration-time history at 327 m to change sharply after the initial arrival phase and follow the pattern set by the data for the 5 m point. Instead, after midday on August 22 the concentration was slowly increasing relative to the concentrations for the 5-m location. This behavior is consistent with a transient storage mechanism in which solute is accumulating in "dead zones" [Fischer et al., 1979; Thackston and Krenkel, 1967] of slowly moving or stagnant water. In small mountain streams this mechanism may be a significant factor for solute transport. From approximately August 29 through August 31, the concentration at 327 m stopped rising and become stable relative to that at 5 m. There remains an ap-

proximate difference of 1 mg L^{-1} between the concentrations at the upstream and downstream locations. We attribute this steady state difference to dilution by background water inflows along the length of the stream.

Data for strontium, potassium, and lithium are presented in Figures 3b, 3c, and 3d, respectively. The strontium data at 5 m follow virtually the same patterns as do the chloride data. There is a small diurnal cycle imposed on a trend of slowly increasing concentration. At the downstream location, however, the strontium data are appreciably different from the chloride data. Over the initial 10-day period, the attenuation of the strontium concentration continued to be of the order of 2 mg L^{-1} . This attenuation reflects the significant capacity of the streambed sediment to sorb strontium [Cerling and Spalding, 1982] and thus influence transport.

The results for potassium and lithium are similar to those for strontium. The influence of sorption is greatest for strontium, followed by potassium and least for lithium. The significant observations for the potassium and lithium data are that (1) the responses for the 5-m location are qualitatively identical to that of both chloride and strontium and (2) the responses for the 327 m location are intermediate between the other injected constituents.

Solute Data: Initial 24 Hours

In this transient period the response of the system to the injection pulse becomes well established. As is clear from the previous set of figures, the system does not achieve a steady state over this period but does continue to respond (at least for 9 additional days) in accord with the patterns set. Detailed concentration-time histories are presented for chloride, strontium, potassium, and lithium in Figures 4a, 4b, 4c, and 4d, respectively. In addition to the upstream-downstream data shown previously for the locations at 5 and 327 m, intermediate data are shown at 62, 126, 184, and 327 m. The dominant

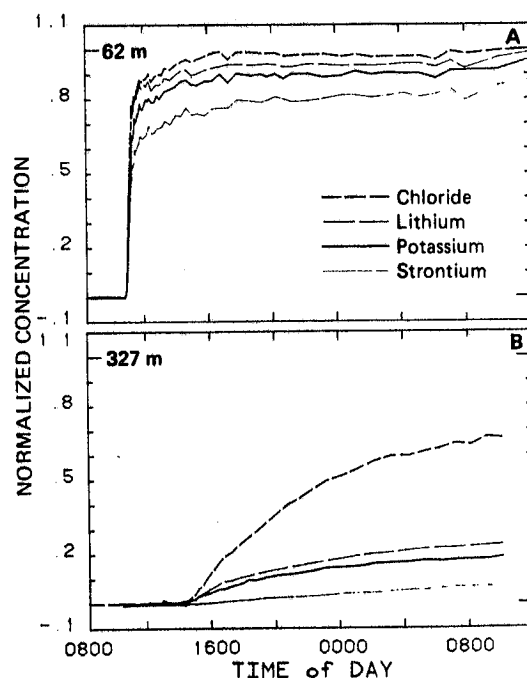


Fig. 5. Normalized tracer concentration data for chloride, strontium, potassium, and lithium. Normalized concentration = (concentration - background)/(injection level concentration - background). Injection level concentration is the concentration at 5 m. (a) Station at 62 m. (b) Station at 327 m.

features of these data are (1) the consistency as we proceed downstream (qualitatively, there are no significant spatial changes in processes) and (2) the attenuation of all solute concentrations as we proceed downstream (the transient storage and sorption processes are manifest in this initial stage).

Cation Attenuation

The influence of sorption on cation transport is clear in the preceding figures. However, the coupling of this geochemical process with the transient storage mechanism makes it difficult to identify the relative roles of these processes from plots of absolute concentration versus time. Thus we have attempted to place the concentration-time histories of all four solutes on one plot by normalizing the absolute concentrations for background and injection level concentrations. Detailed, normalized concentration-time histories are presented for 62 and 327 m in Figures 5a and 5b, respectively. In Figures 5a and 5b the ordering in the amount of sorption, strontium > potassium > lithium, is evident. From Figure 5b we can segregate the influence of storage and sorption mechanisms. The relatively slow rise of the chloride concentrations is the result of storage mechanisms which will act on any tracer in the stream. Comparison of the normalized cation concentrations to chloride shows the additional influence of sorption. During the early phases of the solute arrival the role of a physical process, transient storage, is very important in the attenuation of all tracers.

Return Stream Flow

In Little Lost Man Creek the interaction between solutes in the channel and the surrounding streambed appears to be substantial. In small, mountain streams, flow within a coarse gravel and cobble streambed may contribute to the total transient storage of solutes along the channel. An indication that there is in fact active communication between water in the channel and in the streambed can be observed by studying the

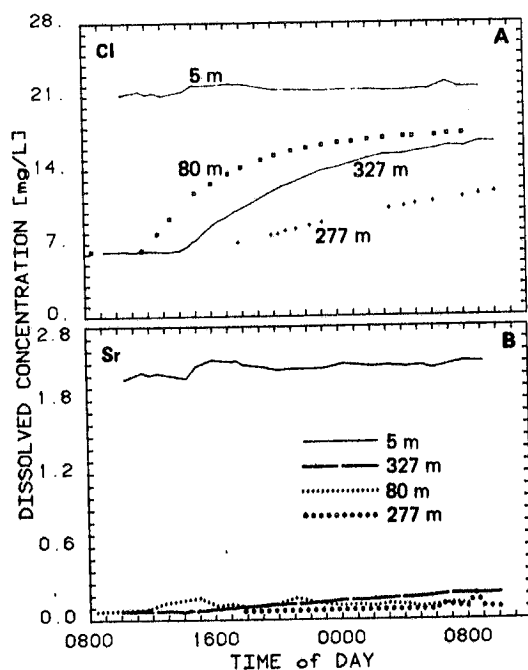


Fig. 6. Tracer concentrations at return stream flow locations (80 and 277 m) compared to concentrations in the stream (5 and 327 m). (a) Chloride. Individual symbols are shown to indicate the sampling intensity. (b) Strontium.

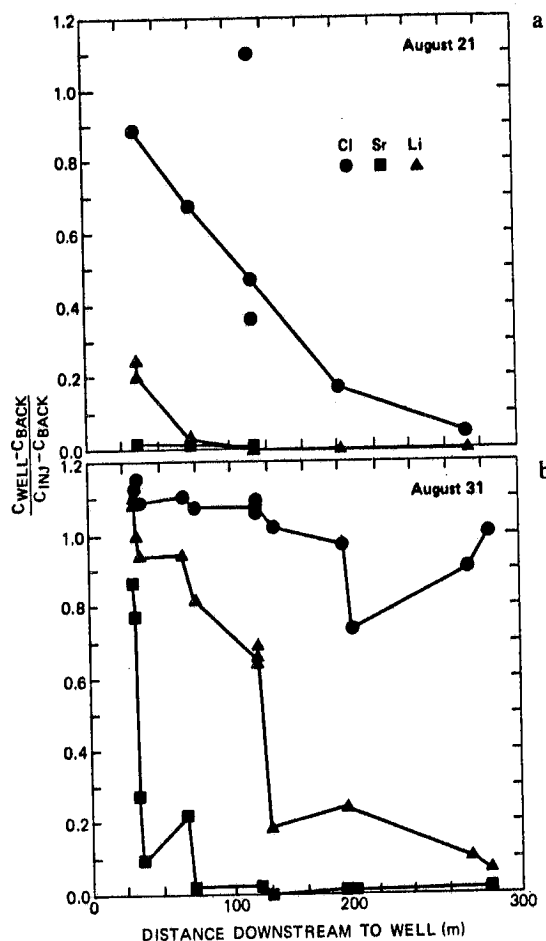


Fig. 7. (a) Tracer concentration data in wells on August 21. (b) Tracer concentration data in wells on August 31.

solute concentrations in the water inflows to the stream. Concentration-time histories of chloride and strontium are presented in Figures 6a and 6b, respectively at the two water inflows (80 and 277 m). The concentrations in the inflows are compared to the upstream-downstream concentrations. From the chloride data it is clear that solutes do enter the flows in the gravel beds and return to the main stream flow. In this flow the concentrations are attenuated and the transport delayed. The strontium concentrations are further attenuated compared to the chloride values. This is taken as evidence for interaction of the cations with the streambed materials. The flows at 80 and 277 m are return stream flows containing, at least in part, water which at one time was in the stream, entered the gravels, and later returned to the stream.

Wells

Tracer data from the shallow wells provide additional evidence for both flow into the surrounding streambed and cation sorption. Chloride, strontium, and lithium data (normalized for background and injection levels) are presented for August 21 and August 31 in Figure 7. (For August 31, several of the adjusted concentration values exceed 1.0. To be consistent, the values of the injection level concentration used for the August 21 adjustments were also used on August 31. The actual values of the injection level concentration for August 31 would be somewhat higher due to decreased stream discharge.) The chloride data are clear indicators of the transport of solute out of the stream channel into storage zones. This

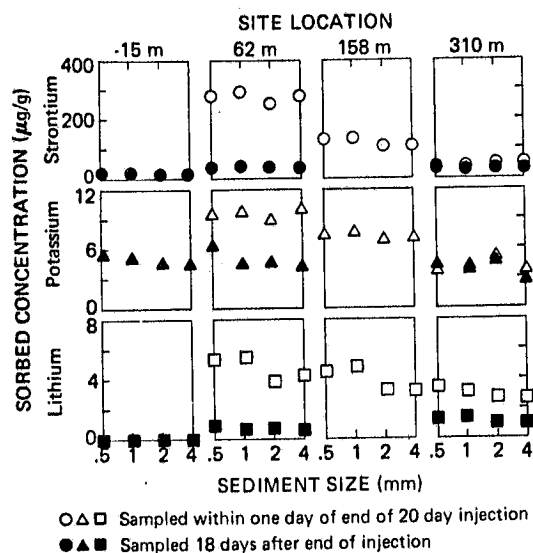


Fig. 8. Cation sorption onto sediment.

transport is slow compared with longitudinal transport in the stream. The chloride values on August 21 are markedly lower than those on August 31. The strontium and lithium values reflect chemical processes identical to those occurring in the stream. The cation values are appreciably attenuated relative to the chloride values and the attenuation increases in the downstream direction. Note that the increase of cation concentration is slow. On August 31 the normalized concentrations for lithium starting with the well at 126 m are all below 0.3, and for strontium starting with the well at 74 m they are all below 0.03. Over a 10-day period the sorption of strontium in the surrounding streambed continues to be significant along most of the study reach.

Sorption

The sorption of trace cations, in particular strontium, has previously been observed on sand- and gravel-sized sediments in streams [Cerling and Turner, 1982; Cerling and Spalding, 1982] and aquifers [Jackson and Inch, 1983].

The effects of cation sorption in the Little Lost Man Creek experiment are most evident at the very end of the injection period when we can observe maximum concentrations on the sediment. In Figure 8, summary plots are shown of strontium, potassium, and lithium concentrations on sand- and gravel-sized sediments at four locations along the stream. The samples were obtained within 1 day before and after the end of the injection and then again 18 days after the injection stopped. One location was 15 m upstream of the injection, and thus for each cation the concentrations at this location represent the background values. Each sediment sample was sized into four subsamples (0.25–0.50, 0.5–1.0, 1–2, and 2–4 mm).

The data in Figure 8 show relatively little variation in the sorbed concentration as a function of sediment size. Each of the three cations exhibits similar behavior. The significant differences are between the sampling locations and sampling times. During the injection, cation concentrations at the 62-m location rose significantly relative to the background values at -15 m. At both 158 and 310 m the increases are smaller, reflecting the coupling between the sorption process along the length of the stream and the resulting lower dissolved concentrations in the downstream direction. (Recall from Figures 3b and 3d the continuing large upstream-downstream con-

centration differences after 10 days.) Eighteen days after the injection stopped, the sorbed cation concentrations dropped relative to the previous samples and returned to nearly the background values.

These data yield a qualitative indication of the significance of the sorption process. A more detailed analysis would require sediment sampling at various depths in the streambed and over a wider range of sediment sizes.

DISCUSSION

The transient storage mechanism resulting from dead zones in the stream and flow through the gravel bed is clearly seen in the slow rise in chloride concentration during the first 10 days. Additional new evidence of flow in the surrounding streambed is seen in the return stream flow and shallow well data. The low, but slowly increasing, cation concentrations from these locations show the great capacity of the streambed solids to remove and store cations from the stream.

The Little Lost Man Creek experiment continues studies of coupled transport and geochemical interactions begun with two experiments in Uvas Creek (Santa Clara County, California). In the first Uvas Creek experiment [Zand et al., 1976; Bencala, 1983], chloride and strontium were injected for a period of 3 hours. A longer injection period, 24 hours was used in the second experiment [Kennedy et al., 1984; Jackman et al., 1984]. The analyses of the Uvas Creek experiments demonstrated the role of transient storage zones in determining solute concentrations and the importance of considering cation sorption to streambed sediment as a kinetic process. Inferences concerning the role of transient storage zones were based on the behavior of the solute concentrations within the active stream channel. The Little Lost Man Creek experiment was designed to test further these observations in a second environment and with a more extensive experiment. The processes observed in Uvas Creek are also significant in Little Lost Man Creek. Monitoring in Little Lost Man Creek was done not only for solute concentrations within the stream but also in areas of subsurface flow and suspected return stream flow. Sampling these flows yielded direct evidence of interaction with the surface flow. The long period (10 days) of approximately constant streamflow and tracer injection in Little Lost Man Creek is also significant. The storage and sorption processes occur on long time scales. This experiment occurred on similar time scales and permits a clear observation of the processes.

There is a definite and complex coupling between solute transport processes in the stream and physical and chemical mechanisms occurring in the streambed. An understanding of these mechanisms requires an extensive data base, spatially and temporally, concerning solute concentrations in the stream, in the surrounding streambed, and on the sediment.

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- R. J. Avanzino, K. E. Bencala, V. C. Kennedy, and G. W. Zellweger, U.S. Geological Survey, 345 Middlefield Road, MS/496, Menlo Park, CA 94025.
- A. P. Jackman, Department of Chemical Engineering, University of California, Davis, CA 95616.

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