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KARST HYDROLOGY AND CHEMICAL CONTAMINATION*

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ABSTRACT

Ground-water flow in karst aquifers is very different from flow in granular or fractured aquifers. Karst ground-water flow is often turbulent within discrete conduits that are convergent in their upper reaches and may be divergent in their very lower reaches, simulating the flow pattern of surface water streams that are dendritic or trellised but with discharge to one or more springs. Significant precipitation events tend to flood karst aquifers quickly, causing a rapid rise in the potentiometric surface that may flood older, higher levels which discharge to a different set of springs. The epikarstic zone in karst terranes stores and directs infiltrating water down discrete percolation points. Chemical contamination may be fed directly to a karst aquifer via overland flow to a sinkhole with little or no attenuation and may contaminate down-gradient wells, springs, and sinkholes within a few hours or a few days. Contaminants may also become temporarily stored in the epikarstic zone for eventual release to the aquifer. Flood pulses may flush the contaminants to cause transiently higher levels of contamination in the aquifer and discharge points. The convergent nature of flow in karst aquifers may result in contaminants becoming concentrated in conduits. Once contaminants have reached the subsurface conduits, they are likely to be rapidly transported to spring outlets. Traditional aquifer remediation techniques for contaminated aquifers are less applicable to karst aquifers.

INTRODUCTION

Ground-water contamination from anthropogenic sources has become a serious issue in recent years. New techniques for evaluating chemical contamination of aquifers are developing almost daily. Unfortunately, chemical contamination of

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karst aquifers is not being given the specialized treatment that is required, mainly because most ground-water investigators lack an understanding of the principles of karst hydrology. The serious problems associated with chemical contamination of karst terranes and the general lack of knowledge concerning flow in them have been recognized by some ground-water professionals [1]. They share a growing concern that more attention must be given to karst terranes. Karst researchers have long known that special conditions exist in karst terranes and that these must be addressed.

Karst terranes are well known to be poor choices for industrial sites, yet many types of industrial developments frequently occur in them. These developments increase the probability that a release of chemical contaminants may ensue because of either natural circumstances or as a result of man's inefficiency and ineptness.

Five practical problems identified by LeGrand may be regarded as significant obstacles to industrial developments in karst terranes [2]. These five problems can have serious effects on attempts to prevent ground-water contamination from chemical releases. The five problems are: 1) poor predictability of ground-water flow, 2) scarcity of surface streams, 3) instability of the ground surface, 4) leakage from surface reservoirs or waste lagoons, and 5) an unsuitable waste-disposal environment.

The susceptibility of karst aquifers to chemical contamination is related to each of the five problems identified by LeGrand, to the processes of carbonate dissolution, and to the hydrologic characteristics peculiar to karst terranes. These problems, processes, and characteristics cannot be "engineered out" nor can they be ignored. Sound waste-management principles and the ability to recognize site problems and to adjust to them (even if it means relocating the site) are the only solutions to dealing with karst terranes.

Contaminant transport in karst terranes is highly complex because of the occurrence of both dispersion and convergence in karst terranes and the ability to store contaminants for long periods of time. The concept of long-term storage in a karst aquifer conflicts with commonly held perceptions while the notion of dispersion and convergence within the same aquifer seems to be contradictory. The purpose of this article is to review and compile the existing karst hydrological literature, relate it to contamination problems, and correct some of the misperceptions regarding chemical contamination of karst terranes.

KARST HYDROLOGY

Ground-water flow in karst terranes is very different from that of granular or highly fractured terranes. In general, the slow, dispersive, laminar flow defined by Darcy's law is rare in karst terranes. Most ground-water flow in most karst terranes is likely to be very rapid, convergent, and turbulent within discrete conduits [3, 4]. To understand why and how a karst aquifer is susceptible to chemical contamination, a basic recognition of the recharge, storage, and flow properties typical of most karst terranes must be established [5]. Figure 1

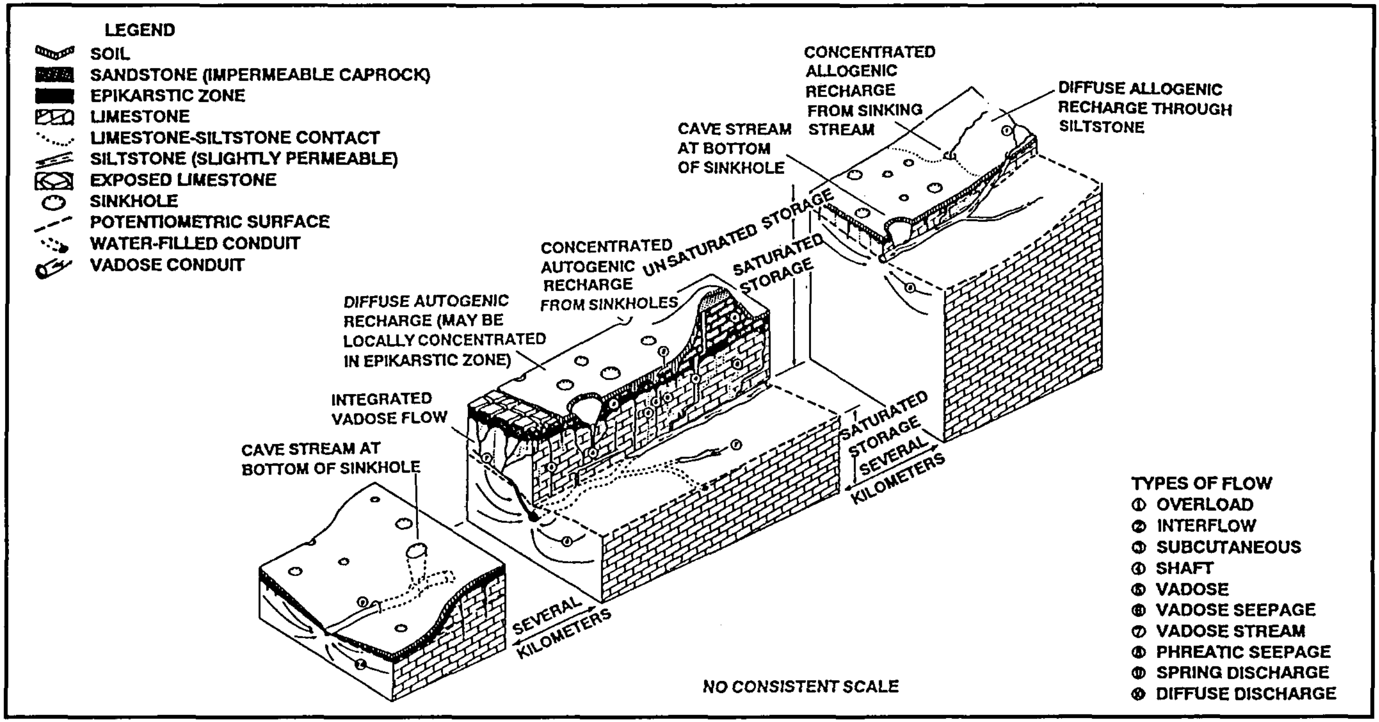

Figure 1. Block diagram depicting the hydrological relationships that exist in a maturely karsted terrane in gently dipping rocks of a low-relief landscape. The cave stream is fed by (1) sinking streams, (2) vertically percolating water infiltrating through soils and sinkholes, (3) tributary cave streams, and (4) seepage through conduit walls. Contaminants will enter the aquifer through the same mechanisms that control water inflow [27].

schematically depicts a typical karst terrane, but it should be remembered that actual geological conditions may be quite different. No generalized diagram can accurately describe each field condition likely to arise.

Ground-Water Recharge

Recharge to a karst aquifer can take the form of concentrated input, such as flow down a sinkhole, or as diffuse recharge from seepage inflows over the entire extent of the ground-water basin. Flow down a sinkhole may feed directly to a significant conduit or, more commonly, may feed small conduits that are tributaries to the main conduit drain [6].

Both diffuse and concentrated recharge generally enter a section of the subsurface in a karst terrane termed the epikarstic (subcutaneous) zone. This is a highly fractured and weathered portion of the rock immediately underlying the soil zone and separated from the phreatic zone by an interval where water is channelled through discrete solutional openings. Williams [7] reported that 50-80 percent of carbonate rock dissolution occurs within the first 10 m of the bedrock surface; thus, the uppermost layer of rock beneath the soil zone is heavily corroded. Smart and Friederich emphasize that most dissolution occurs within a depth of 3 m, where fissuring has a uniformly high density and may even exhibit a rubbly nature [8]. Fissures are widened by solution at shallow depths but close rapidly with increasing depth because of overburden pressure and reduced dissolution [8], except for a few isolated preferential vertical flow paths termed subcutaneous drains by Smart and Hobbs [4]. Specific yield within the epikarstic zone is greatest [9, p. 138] because of the enhanced dissolution created by the relatively fresh infiltrating precipitation that is undersaturated with respect to $CaCO_3$. Williams described how the epikarstic hydrology is directly related to the subcutaneous drains [7]. These drains developed more extensively than adjacent fissures because they discharged larger quantities of water early during the process of karstification. The more fresh water discharged by the subcutaneous drains, the greater the extent of their dissolution.

Significant storm events tend to provide excess amounts of recharge to the epikarst, commonly resulting in a temporarily perched water table within the highly porous epikarstic zone. The perched water, accompanied by a buildup in hydraulic head, flows laterally toward the subcutaneous drains, which are the points of lowest head, as shown in Figure 2. A depression develops in the epikarstic water table over these subcutaneous drains as the epikarstic water flows down them in a turbulent manner. This depression is similar to the cone of depression that develops around a pumped well [7] except that it tends to be extremely elongated (Figure 3). Friederich and Smart [10] reported lateral flow rates on the order of 100 m/hr, which would indicate that Darcy's law is not always applicable to flow in the epikarstic zone. Vertical flow down subcutaneous drains was shown by them to be commonly in excess of 100 m/hr (at times, as high

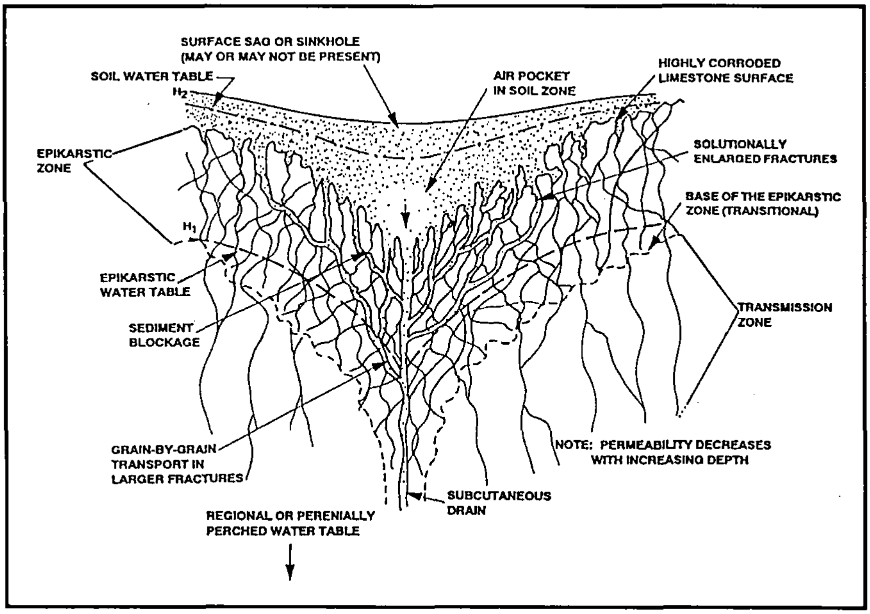

Figure 2. Cross section of the epikarstic (subcutaneous) zone during a period of substantial recharge. The overlying soil controls the rate of infiltration to the epikarstic zone. Infiltrating water becomes perched in the epikarstic zone (H_1) because of its high hydraulic conductivity relative to that of the transmission zone. Additional recharge may result in the formation of a soil water table (H_2) if the infiltration capacity of the epikarstic zone is exceeded. A cone of depression directed toward the subcutaneous drain is a result of the excess recharge being transported to the point of lowest hydraulic head.

as 600 m/hr), whereas vertical flow down the smaller, less developed fissures was less than 2 m/hr. They concluded that subcutaneous drains are responsible for the majority of aquifer recharge. It should be noted, however, that some of the water may not reach a karst aquifer until several months after a precipitation event because it has been stored in the epikarstic zone [9, p. 161].

As depicted in Figure 2, the lateral flow occurring in the epikarstic zone causes more corrosion at the subcutaneous drains because of their ability to accept more water than the adjacent fractures. The more corrosion that occurs, the more the vertical permeability becomes enhanced [7]. This greater corrosion and enhanced vertical permeability also leads to the redirection of soil water to the subcutaneous drains, commonly causing a catastrophic collapse of the overlying soil. Figure 4 schematically shows how a solutionally formed air-pocket stopes upward to the point where the ground surface can no longer be supported. If a subcutaneous drain exists beneath a hazardous-waste disposal unit, attempts to

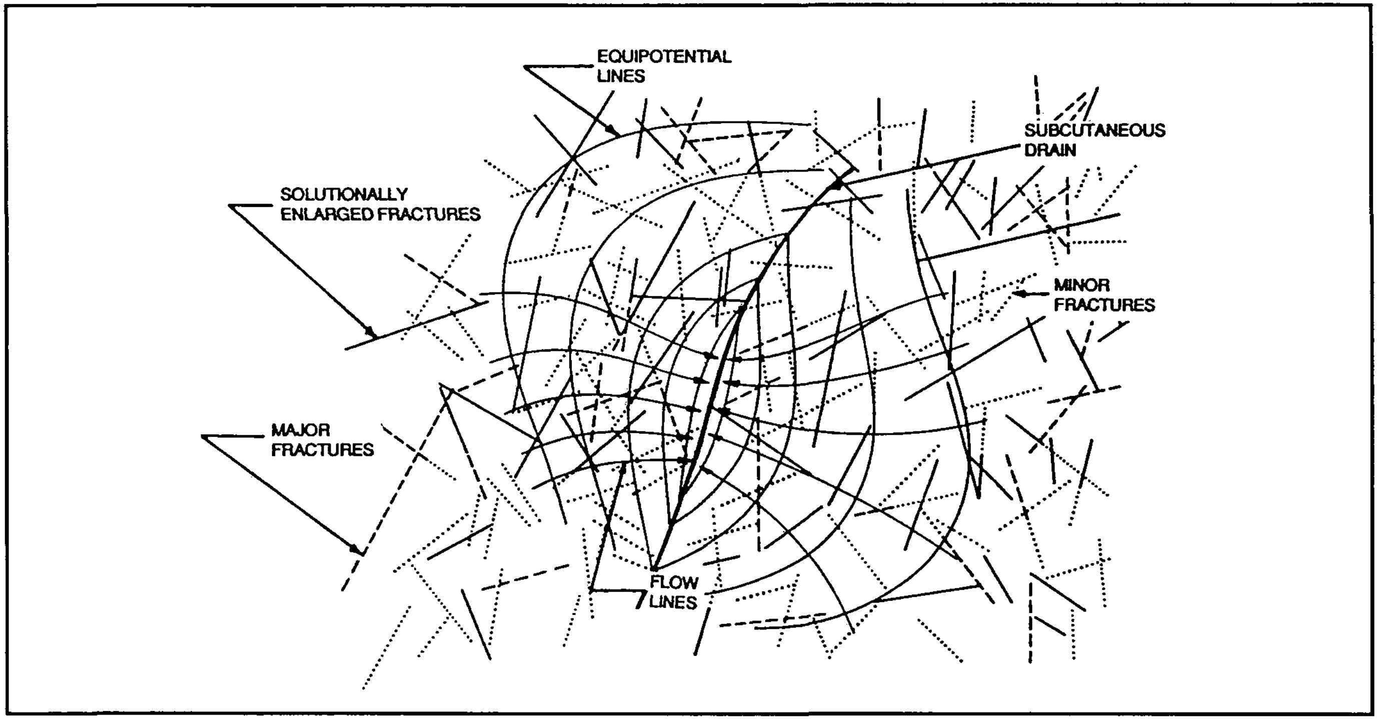

Figure 3. Plan view of the epikarstic zone during a period of substantial recharge. Elongation of the cone of depression is a consequence of the lateral extent of the subcutaneous drain. Although they are smooth lines, the equipotential lines and flow lines would actually be highly irregular because of the randomness of the fracture permeability and the lack of uniform permeability of the subcutaneous drain.

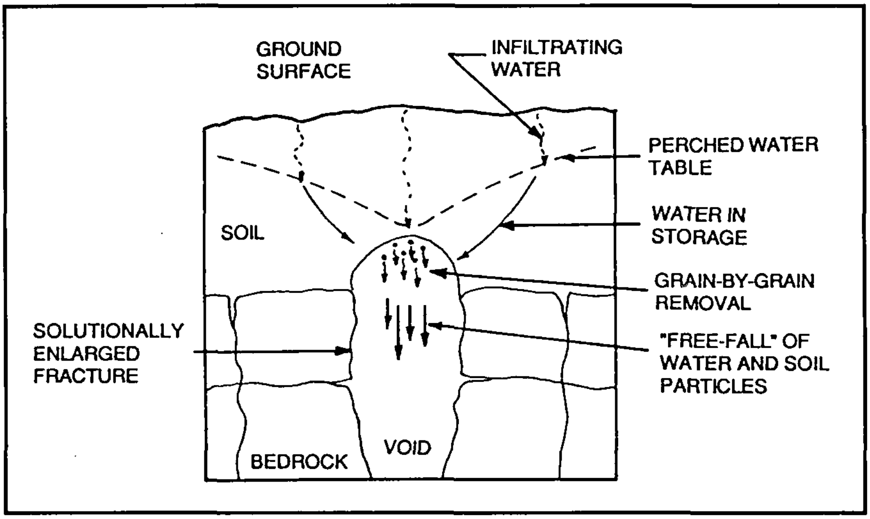

Figure 4. Development of an air pocket in the soil zone as a result of lateral through-flow in the epikarstic zone. Surface collapse occurs when supporting clastic material has been removed via spalling triggered by infiltrating water. Alternatively, an air pocket could be a response to lowering of thepotentiometric surface and consequent upward stoping and progressive failure of soil archces [49]. Horizontal scale is exaggerated by a factor of about three.

restrict infiltration around the unit may do little to prevent the development of a sinkhole beneath it because of the lateral flow component within the epikarstic zone.

Once a sinkhole has developed, point-source recharge during storm events from above may temporarily produce a recharge cone (mound) rather than a cone of depression [7]. This further results in directing recharge down discrete percolation zones such as vadose shafts, which may or may not be part of the epikarstic zone. Vadose shafts are vertical cylindrical openings ranging in diameter from a few millimeters to many meters and are produced by vertically descending water [11]. Palmer states that the vertically descending water spreads out in a thin film or spray which is responsible for enlarging the shaft [12]. Except for the flow of water as a film rather than as a cylindrical mass, vadose shafts are similar to pipes that transmit water through the vadose zone by the most efficient route. Vadose flow will descend along the steepest available openings which normally alternate along their length between vertical shafts and inclined canyon like passageways [12].

Below the epikarstic zone, preferential flow paths develop along major vertical joints, faults, and bedding-plane partings [8]. However, Palmer has pointed out that divergent flow is prominent in the vadose zone as shown by the large number of diversion passages for vadose cave streams [13]. Under the influence of gravity, subsurface vadose water moves progressively downward to lower flow-routes where the water may feed into several conduits over a large area. For this to happen, a vadose cave-stream must leak through underlying fractures or bedding-plane partings, some of which will become solutionally enlarged and thus eventually take all the flow. The important fact to recognize here is that as recharge reaches the phreatic zone, it has generally done so in a concentrated form but to several different conduits that may or may not become integrated along their length, thus causing some dispersion.

Ground-Water Flow

Ground-water flow in karst aquifers may be described in terms of conduit and diffuse flow, two end-members of a continuum [4, 14, 15]. Most flow in most karst aquifers is a mixture of the two in which one of the flow types predominates over the other [3]. Conduit flow occurs in large passages that allow relatively high ground-water flow velocities under turbulent flow conditions and an almost insignificant low-flow hydraulic gradient. The orientation of passageways depends almost entirely on the local stratigraphy and structure [16, 17]. Flow velocities are generally higher in open-channel vadose passages than in tube-full phreatic conduits [13]. Palmer also discusses the need to distinguish between currently forming and original vadose passages from phreatic passages because flow directions may be different in each zone. Careful leveling and mapping of beds along cave walls enabled him to demonstrate that most vadose passages in the Mammoth Cave area develop along the dip of the beds. In contrast, most phreatic conduits follow the strike of the beds except where fracturing is prominent. This difference can be explained in terms of flow in the vadose zone which is controlled by gravity and flow in the phreatic zone which is controlled by hydraulic efficiency.

The concept of a continuous potentiometric surface becomes less appropriate as conduit flow becomes more dominant, according to Smart and Ford [18]. They observed the perching of some springs and the frequent overflow flooding that occurs in karst aquifers dominated by conduit flow as evidence that a continuous potentiometric surface does not always appear to exist in an obvious manner. This is not to say, however, that a potentiometric surface does not exist in a karst aquifer. Karst aquifers dominated by conduit flow still contain diffuse-flow in-feeders which, when tapped by observation wells, allow the construction of a well-defined potentiometric surface map [19, 20, pp. 181-183]. Also, the dynamic response of the potentiometric surface to storms is greatly amplified in the conduits of karst aquifers, allowing perched streams to develop in the vadose zone, a fact which may account for confusion over the existence of a true potentiometric surface.

Diffuse flow occurs within the tight fractures and small pores with relatively low flow velocities. It is laminar and behaves according to Darcy's law. A much more significant hydraulic gradient is in evidence in karst aquifers dominated by diffuse flow and can be mapped reasonably accurately if the influence of conduit flow is recognized. White [20, p. 182] points out that combining dye-tracing experiments with detailed mapping of caves provides important information on the conduit-flow portion of a karst aquifer, while mapping of the potentiometric surface enables one to interpret much about the diffuse-flow portion of a karst aquifer (as done by Quinlan and Ray [19], Crawford [21]). If conduit-flow analysis is combined with a potentiometric surface map of the area, anisotropic flow, as suggested by local non-orthogonality of flow lines to equipotential lines, may be present. In fact, it is possible that some flow lines may even appear to be parallel or sub-parallel to equipotential lines [22, pp. 29-31; 19, 21]. This is not what actually occurs, but the lack of closely-spaced data points may make such parallelism seem to occur in karst terranes. Accordingly, incorrect predictions of ground-water flow patterns are to be expected [23]. The possibility that flow lines may appear to be parallel or subparallel to the equipotential lines is so foreign to conventional wisdom that it is completely ignored in many ground-water investigations in karst terranes. Such ignorance can lead to devastating effects from chemical contaminant releases because their flow route is incorrectly predicted. When the potential result of incorrectly predicting contaminant flow routes is considered, this parallel flow factor takes on even greater significance.

In pollution monitoring of granular or highly fractured terranes, the importance of the hydraulic gradient in relation to Darcy's law is vital in identifying the flow rates and direction of the ground water. But Darcy's law is not valid when applied to karst aquifers because turbulent flow within discrete conduits is common in karst aquifers. Darcy's law assumes laminar flow in which individual particles of water move in parallel streamlines in the direction of flow with no mixing or transverse component in their motion [9, p. 132].

The importance of Darcy's law relative to turbulence may be misplaced, however. Darcy's law tends to break down when flow lines become distorted because of changes in the direction of motion. This occurs when the changes in motion direction are sufficiently great as to cause the inertial forces to become significant relative to viscous forces. The ultimate result is that turbulence will develop at a much higher Reynolds number than originally predicted, yet Darcy's law was violated at a very low Reynolds number [24, p. 164]. White explains that whenever turbulent flows occurs within a conduit, there will be a laminar flow boundary layer along the conduit wall [20, pp. 160-164]. The thickness of this boundary layer decreases with increasing Reynolds number, and turbulent flow occurs at a lower Reynolds number than expected. More clearly stated, Darcy's law will break down long before the critical Reynolds number of 2100 for smooth pipes is reached. Although the Reynolds number for the actual onset of turbulence in conduit flow is not known, it is believed to be much less than 2100 and may be as low as 10 [20, p. 164], mainly because of the flow-line distortion that develops from the change in motion direction and because of the relationship of surface irregularities to the thickness of the laminar flow layer that develops along the walls of a conduit. Darcy's law cannot be used to predict flow velocity in most karst aquifers, especially those dominated by conduit flow. To do so is to commit a grave error in judgment, but many ground-water professionals working in karst terranes still rely on Darcy's law.

The importance of the hydraulic gradient in a karst aquifer still remains crucial to understanding the migration of chemical contaminants. Palmer pointed out that the discharge of water through carbonate rock is more dependent upon variations in channel size than upon variations in hydraulic gradient [25]. He emphasized the importance of this concept by describing a scenario where, under phreatic conditions, two adjacent flow paths of differing sizes will exhibit unequal hydraulic gradients along their lengths. The larger flow path will have a much lower hydraulic gradient while transmitting greater amounts of water. If both flow paths discharge to a nearby river, the larger, more efficient opening will show evidence of a hydraulic head gradient that is almost flat in relation to the less efficient opening that will be required to maintain steeper hydraulic heads. So even a long circuitous flow route that started with a wide initial opening size may develop into the dominant flow path in spite of the fact that its hydraulic gradient is much less than the more direct but less efficient flow routes [12]. The consequence of this, especially for pollution monitoring, is that the water in the inefficient channels will possess a steeper hydraulic gradient toward the nearby, more efficient flow channel as contrasted with the more remote river. As one moves farther away from the efficient flow channel, the hydraulic gradient may falsely seem to suggest that flow is uniformly flowing to the now less remote river (in relation to the efficient flow channel) as shown in Figure 5. In reality, however, the majority of the flow will still be directed toward the more efficient flow channels (Figure 6). Without a very large number of observation wells over the entire drainage basin, with many of the wells in or adjacent to the more efficient flow channel draining the basin, this characteristic cannot be observed.

Many investigators fail to accurately define the true flow paths in karst terranes because of an unfailing reliance on measuring the hydraulic gradient in observation wells. Methodology described by Quinlan and Ewers suggest that dye-tracing studies to better define the true flow paths will allow accurate monitoring for pollution studies to be conducted at springs rather than wells [3]. Field [26] and Quinlan [27, 28] have now concluded that if dye tracing to wells proves positive, then the monitoring of these wells, plus spring monitoring, is appropriate. The rationale for this is that monitoring wells must intersect the major conduit(s) draining a site, but that the probability of a randomly drilled well doing so is commonly less than 0.04 percent [3]. Obviously, this probability is low and is an uneconomical goal, but on the slight chance that an intersection does occur, existing wells are highly desirable for monitoring sites for dyes in tracing studies which are necessary prerequisites for design of a monitoring system in a karst terrane.

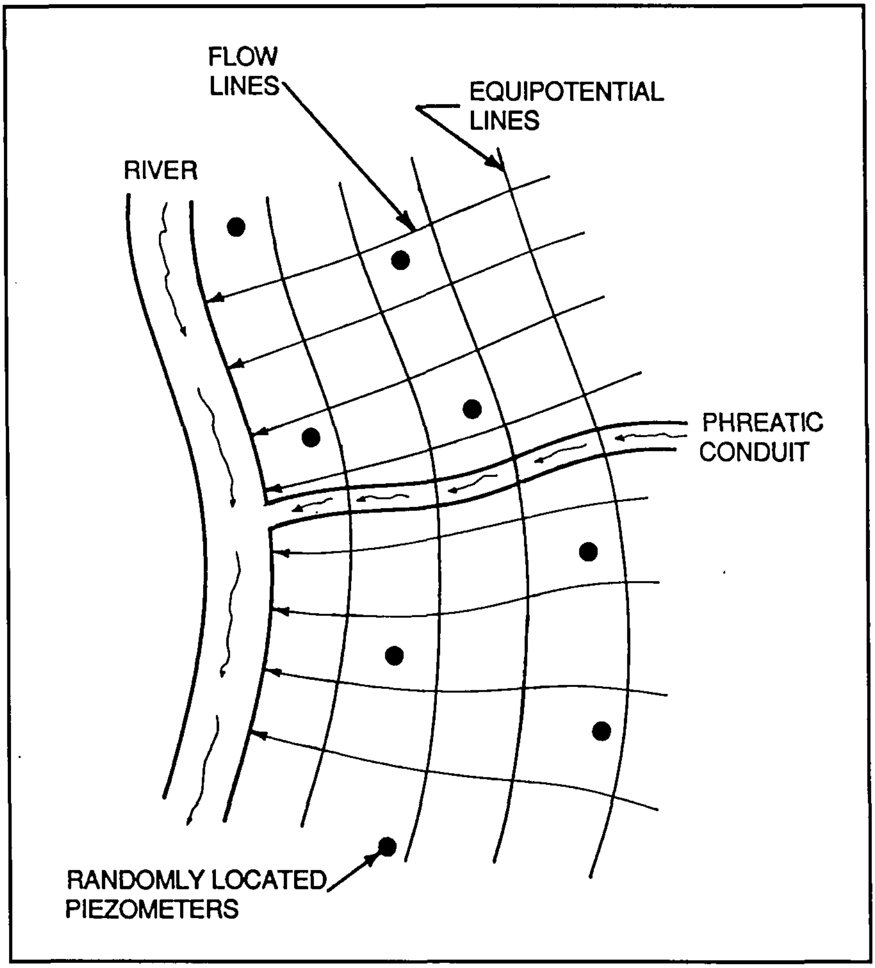

Figure 5. Incorrect flow net of a karst aquifer dominated by conduit flow. The flow net was derived from water-level measurements in randomly placed piezometers that were unable to unambiguously show hydraulic effects produced by the cave stream.

The incorrect conclusion that only a single conduit drains a ground-water basin when more such drains exist can be as serious as ignoring the existence of conduit flow altogether. Caves develop in stages, that is, through time the uplift of land surface and a lowering of the base level of nearby rivers will cause the ground water to progressively develop lower cave levels in an attempt to establish equilibrium with its surroundings. The result is a series of levels in the vadose zone leading to younger cave levels in the phreatic zone.

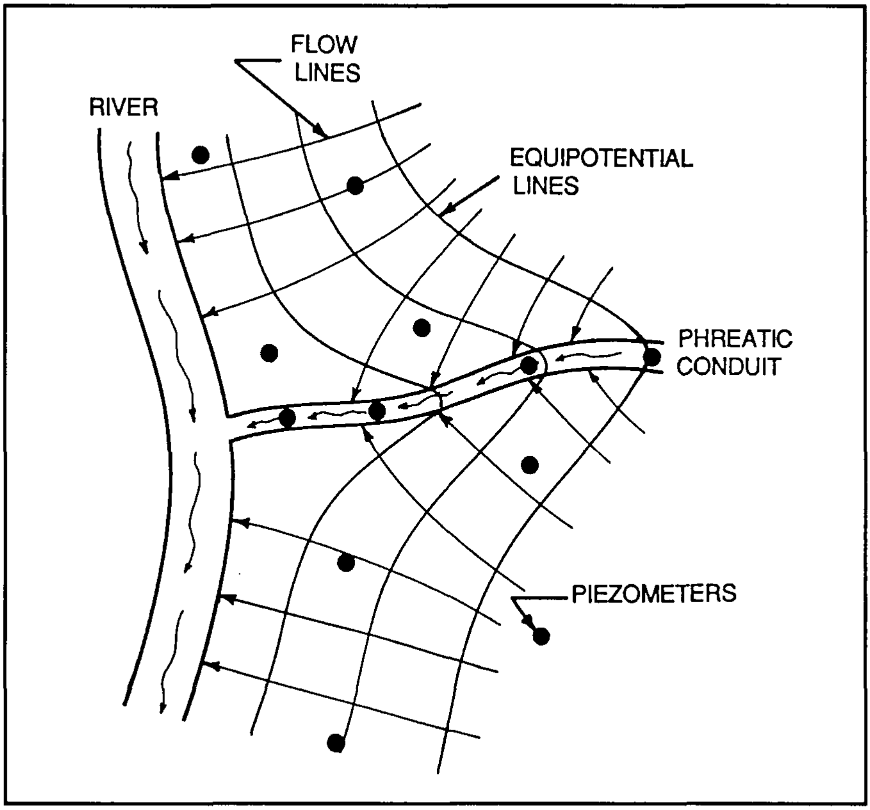

Figure 6. Actual ground-water flow in a karst aquifer dominated by conduit flow as shown by the same piezometers as in Figure 5 plus the fortuitously accurate placement of more piezometers than would be necessary in the cave stream. Water-level measurements in and near the cave stream indicate that the cave stream is draining the basin and probably discharges most of the ground water to the nearby river.

Head buildup in phreatic conduits from floodwaters entering the conduits can cause water level rises of 100 feet or more in observation wells in a few hours [3; 29, pp. 128-132] and can cause additional ground-water divergence [13]. Because head buildup in phreatic conduits is much greater than in the less efficient flow passages, the hydraulic gradient leading to the phreatic conduit reverses. Now, water is forced under very steep gradients back into all available openings in the rock formation and up into older, higher levels in what are usually vadose passages. This water is then discharged from ephemeral higher-level springs or forms a temporary bank storage for release to main conduits once the flow pulse begins to subside.

Independent crossing of two or more conduits without any mixing of the water is also a common occurrence although most such crossings occur in the vadose zone. Palmer states that such a crossing in the phreatic zone probably represents passages formed at different times with flooding causing a re-occupation of the older passages [13] (Figure 7).

Conduits are excellent traps for many things, the most common being sediment. The tendency for large amounts of sediment to accumulate in conduits has allowed a reasonably accurate interpretation of the conduit hydraulics in a manner similar to the interpretation of stream hydraulics based on sedimentological studies of surface streams [30]. More importantly, however, this relatively insoluble clastic sediment may shield the conduit floor from dissolution of the bedrock [12]. Chemical contaminants may flow along this bed of quartz sand and clay minerals [20, p. 400]. Adsorption and desorption of contaminants by the clay particles may have significant effects on the retardation of contaminants released into an aquifer.

Discharge of karst water may also occur through distributaries as a result of the periodic fluctuation of the potentiometric surface [3]. This is evident in the change that occurs from low flow to high flow. Rapid flooding during high flow, with a corresponding increase in hydraulic gradient (Figure 8), causes the water to follow alternative routes in an effort to find the most efficient flow route for eventual discharge [12]. As later pointed out by Palmer, clusters of springs usually represent multiple levels of overflow routes for water in the same catchment area [13]. Adjacent springs are not necessarily draining the same catchment area, however. Dye tracing easily confirms this fact.

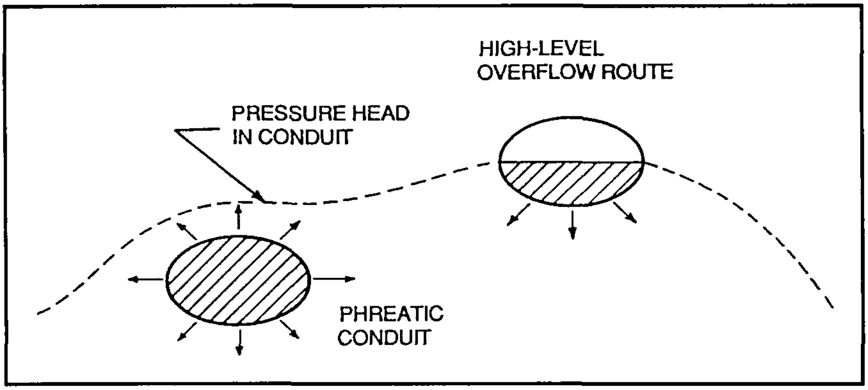

Figure 7. Re-occupation of older, higher previously dry cave levels during periods of flood flow. Water and contaminants are driven into the rock matrix in a form of bank storage. Passages are schematic but may be 10-20 feet in diameter and separated by hundreds to thousands of feet.

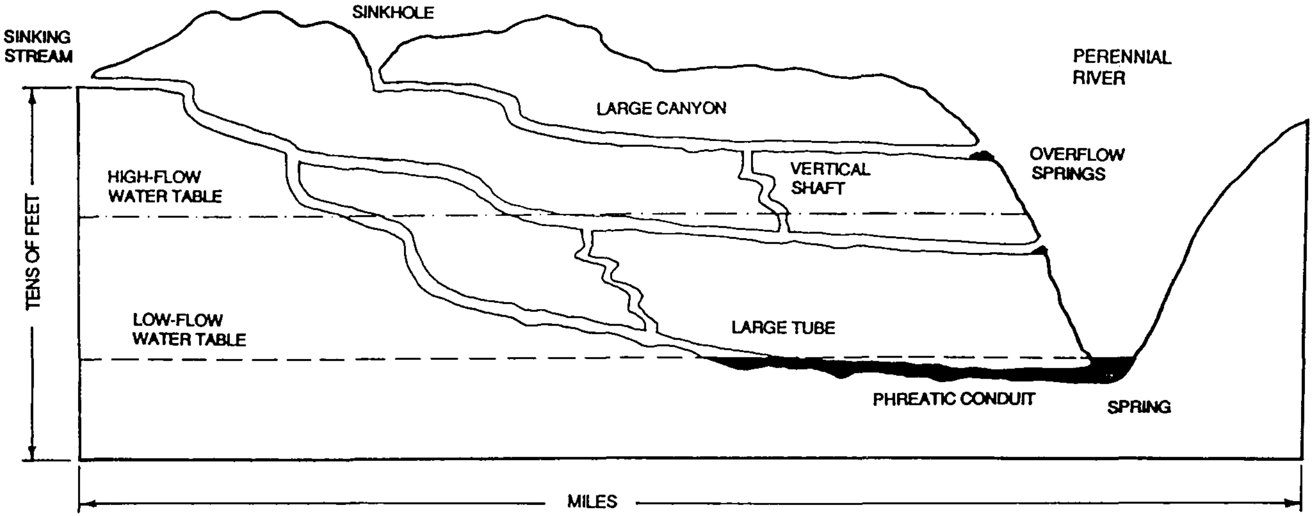

Figure 8. Flooding of older, higher flow routes induces flow of ground water and contaminants to springs other than those draining a basin during base flow conditions. Note vertical exaggeration is approximately ten times.

When evaluating karst ground-water flow, it is important to note that the bulk of the flow will be turbulent within discrete conduits that concentrate flow and emulate surface-water streams that are dendritic or trellised [3]. Discharge can be distributary at the very downstream ends and will be to distinct springs that may individually discharge many cubic meters per second. The discharge of springs is dependent mostly upon surface catchment area [29, p. 84] and the amount of precipitation an area receives. Recognition of this fact emphasizes the ability of a karst aquifer to concentrate flow and discharge to discrete points. As pointed out by Quinlan and Ewers mapping of drainage basins by dye tracing and water-table measurements is essential to accurately define the hydrology of a karst aquifer, which is a necessary prerequisite to determining the extent of contamination from spills in a karst terrane [3].

Ground-Water Storage

Although storage in granular or highly fractured terranes is always difficult to accurately assess, determining storage in karst terranes is far more complex. This is partially due to confusion regarding the hydrology of the phreatic zone, but it is also due to the complexity of the hydrology of the vadose zone. Essentially, five areas of storage exist in karst terranes: the overburden (soil zone), the epikarstic zone, the transmission zone, the phreatic conduits, and the saturated rock mass. The significance of these zones relative to each other varies from one karst terrane to the next [6].

Storage in the overburden (also termed soil moisture) has long been recognized, especially in the soil science literature, as an important factor in the hydrology of an area. Soil moisture is the water content of the soil zone that is retained against the force of gravity. The most important molecular forces in the soil opposing gravity are adhesion (adsorption) and capillarity [31, p. 209; 32, p. 150].

Adsorption is the attraction of water molecules to solid surfaces by surface tension that is held by the effect of Van der Waals forces. Air is also held in place by attraction to water molecules by the same forces. Because of the larger surface area afforded by clay particles, larger quantities of water are retained by adsorption in clayey soils. Capillary rise, caused by matric suction, is responsible for drawing water up into micropores that connect to form microtubes within the soil zone. Capillarity is dependent upon the pore size distribution. Clayey soil also enhances capillarity because pore size distribution is more uniform [32, p. 150]. Together, capillarity and adsorption in the soil zone can account for storage of significant amounts of water, some of which can never be removed.

During periods of high infiltration much of the water in the soil is displaced and continues downward as gravitational flow to the underlying aquifer. As the soil desaturates, some of the pores become air filled, hydraulic conductivity decreases, and tortuosity increases [32, p. 197]. Hence, large quantities of water will become stored in the soil zone which is the controlling factor on infiltration to the phreatic zone in most geological terranes. The release of the soil water is through the tortuous pathways of soil pores that are not completely blocked by soil gas and through macropores in the form of root zones or soil fractures. An exception to the moderating effect of the soil zone on recharge to the phreatic zone is the usually thin soil of karst terranes. As described above, recharge commonly occurs at point sources (sinkholes and other types of drains) that tend to diminish the importance of the soil zone in storage. This fact must not be ignored when investigating chemical contamination of karst aquifers. At the same time, it must be remembered that the soil zone still regulates significant amounts of infiltrating water and has a controlling effect on saturation of the epikarstic zone.

Storage in the epikarstic zone is usually large and can be easily accommodated with a specific yield of about 0.01 while the transmission zone (the next 6- to 10-m below the epikarstic zone) commonly has a specific yield of only 0.001, which is more than adequate to accommodate the balance of storage in the unsaturated zone [8]. The difference in specific yields is directly attributable to the difference in permeabilities. A relatively high permeability in the epikarstic zone is responsible for rapid drainage of stored water which causes high rates of recession and a flashy flow regime. Alternatively, the transmission zone drains much more slowly because storage occurs in fissures and fractures with little solution development. Water is held in the tighter fractures and fissures by capillarity in the same way that water is retained in the soil zone. Because the water passes through relatively few open conduits, the rate of recession is greatly reduced. Smart and Friederich substantiate this by observing the large differences in tracer concentrations between vadose shafts and epikarstic flows and the persistence of tracer in the feeders leading to the vadose shafts [8].

Naturally, storage in the epikarstic and transmission zones is unrelated to water-level measurements and aquifer testing in the phreatic zone, yet, as explained above, these two zones can account for very large amounts of storage in karst terranes. For chemical contamination investigations in karst terranes this is an essential factor to be realized. The retention of large quantities of water in the vadose zone very easily allows the possibility for storage of large quantities of chemical contaminants when they are released in karst terranes. Some of the storage mechanisms will be the same for chemicals as for water, but many other retention mechanisms unique to the specific chemicals released will also be responsible for chemical storage in the vadose zone.

Storage of water within phreatic conduits is often considered to be minimal, which may or may not be true. The problem of defining the amount of storage in phreatic conduits relates to the inaccessibility of perennially flooded conduits. If a pumping well were to intercept one of these phreatic conduits, storage would still be difficult to define because of the small diameter of the well bore relative to the phreatic conduit. A pumping well installed in a phreatic conduit will have almost no drawdown [20, pp. 186-189], whereas a pumping well installed just a few meters away from a phreatic conduit may yield only minimal amounts of water [29, pp. 235-237]. Low-flow conditions may decrease storage in conduits and blockage of them may increase or decrease their storage, but storage is only a small percentage of that of the vadose zone and rock matrix in the phreatic zone [33]. Bonacci [34, pp. 36-48] reviewed several methods for evaluating the storage of phreatic passages and also concluded that phreatic passages have no great capacity for storage.

Thrailkill has developed a method for estimating aquifer properties in what he calls shallow conduit-flow aquifers [35]. This method does not require a change in head. Rather, based upon a conceptual model of a shallow conduit-flow aquifer, the method relies on mass balance and flow equations. This method does not, however, provide any direct insight into the storage capacity of conduits themselves because it is extremely unlikely that the pumping well will intercept the phreatic conduit, and the minute feeders leading to the main conduits will not allow sufficient withdrawal of ground water from the phreatic conduit. This method shows great promise for defining storage and transmissivity values for the phreatic zone as a whole, but much work is still needed. Atkinson [33], utilizing a method developed by Ashton [36], estimated the amount of phreatic storage by comparing the flood pulse to flood water in the phreatic zone. This method indicated that the water stored within minute fractures was approximately twenty-nine times the amount of water stored in the conduits for the particular aquifer studied. The method allows at least some measure of storage capacity because the initial flood pulse will travel through the conduit system relatively rapidly, while some flood water will be driven into the rock matrix to create a form of bank storage. More specifically, the base-flow discharge from a karst spring will have a certain amount of dissolved $CaCO_3$. A large storm event will flood the aquifer and will increase the spring discharge. Water from the first part of the flood pulse will show unusually high levels of $CaCO_3$ because this water is being flushed out of storage. Next, the water will be highly undersaturated with respect to $CaCO_3$ because this is the part of the flood pulse resulting from direct recharge and represents flow and storage in the conduits. Finally, spring discharge will slowly decrease but will exhibit a steady increase in dissolved $CaCO_3$; this is the flood water that was held in storage and was slowly released from the fine fractures and intergranular pores of the rock matrix. Storage is most important in the openings in the rock matrix surrounding the main conduits, because a large majority of recharge will feed into the rock matrix (as opposed to feeding directly into the phreatic conduits) which in turn feed into more open fissures and fractures that eventually feed into the main phreatic conduits by diffuse flow. The small openings that eventually lead to phreatic conduits can account for much larger amounts of storage in the aquifer than described above even if the actual storage capacity cannot be accurately measured. This is possible where diffuse flow dominates over conduit flow. Adhesion to the walls of fissuers and the slow diffuse nature of the flow results in long-term storage relative to the phreatic conduits, but if low porosity and conduit flow dominate, then the overall phreatic storage will be low [20, p. 189].

The complex hydrology of karst terranes is further complicated by the effect of chemical releases to the environment. Some of the processes occurring will be similar to that of spills in granular or highly fractured terranes, but others will not. The differences between karst and non-karst aquifer types in effects created by chemical spills are related directly to the differences in their hydrology. The balance of this paper deals with chemical contamination and discusses the mechanisms related to chemical contamination of karst aquifers.

CONTAMINATION

Much of the previous discussion dealt with the hydrology of karst terranes because of the need to understand the basics of karst hydrology prior to understanding chemical contamination of karst aquifers. Chemical spills in karst terranes will be stored and transported in many ways that are very similar to the storage and flow of water in karst terranes, but it is necessary to relate the hydrology of karst terranes to chemical releases in order to establish the extent to which karst aquifers may become contaminated. Ford and Williams [9, p. 518] describe five specific karst-related phenomena responsible for the ineffectiveness of contaminant attenuation mechanisms in the subsurface. These are 1) poor adsorption or ion exchange of contaminants and little colonization of microorganisms because of a significant lack of available surface area relative to granular aquifers, 2) a reduction in the ability to evaporate highly volatile contaminants from the ground surface because of the extremely rapid infiltration rates possible, 3) little filtration of contaminants because of the thin soils typical of karst terranes and because of the solutionally enlarged fractures in the bedrock, 4) assistance in the transmission of contaminants by the turbulent flow regime typical of conduit flow, and 5) a reduction in adsorption-desorption processes and microbiological activities because of the rapid flow-through rates in conduits.

A popular misconception is that a karst aquifer will rapidly flush chemical contaminants out and thus cleanse itself. Large portions of chemical spills will typically be rapidly transported from an input point to a discharge point (on the order of several meters per day to several meters per hour), but this is not always true. Very large quantities of the chemical may be stored in the subsurface. According to work conducted by Kurtz and Parizek a disposal site in the Nittany Valley of Pennsylvania has released several organic contaminants that have become stored in the subsurface and are slowly being released from the unsaturated soil and bedrock and are likely to persist for many years [37].

Research into fate and transport mechanisms, such as the excellent work by Mackay et. al. [38] has dealt almost exclusively with fairly uniform aquifers, although other investigators are beginning to look at the species of karst terranes [37, 39, 40]. Attempts to apply the various transport mechanisms discussed by

Mackay et al. [38] to karst aquifers without realizing the differences in hydrology and geology can lead to serious errors in rate and direction calculations. Simmleit and Herrmann correctly point out that the complexity of contaminant transport and retention in karst terranes negate the use of simple empirical models and that extensive research is still necessary for even a basic understanding of the behavior of contaminants in karst terranes [39]. Kurtz and Parizek conducted extensive investigations of a chemically contaminated area only to conclude that transport mechanisms must be better understood [37].

Contaminant Infiltration and Transport

Initially, chemical releases will either infiltrate the soil zone (if present), flow overland into a sinkhole, or enter a nearby stream that sinks to the subsurface somewhere along its route. It released to a soil-covered area, the chemicals will be somewhat retarded in their downward migration, depending on their physicochemical parameters, but can still be expected to migrate to an underlying aquifer. Sorption to microparticles will actually do little to prevent downward migration of chemical constituents because transport of the microparticles themselves occurs very readily in all types of terranes but especially so in karst terranes. Lyman and Loreti [41] report in what they call "real-world applications" of their work on sorption coefficients that some highly sorbing chemical constituents may be very mobile because of the natural mobility of soil particles [41]. In karst terranes this effect is magnified many times. Cave explorers relate remarkable stories of finding old tires, picnic tables, and an incredible variety of debris that will fit into a cave opening. Filtration is a term to be avoided in serious discussion of karst aquifers.

If a soil zone is absent, then contaminants will readily flow down the exposed joints with essentially no retardation whatsoever. In such an area, the epikarstic zone is visible at the surface. Once in this zone, chemical constituents will become sorbed to a certain extent to the limestone walls and to any soil particles washed down from the overlying soil zone. This zone can effectively retain the chemicals for many years, slowly releasing small amounts during significant recharge events and continuing to supply contaminants to the underlying aquifer [42]. The bulk of the released chemicals will probably reach the aquifer through the same recharge mechanisms that affect the flow of water to the aquifer.

Upon reaching the epikarstic zone, chemical contaminants will either flow relatively freely or will be stored until a significant storm event flushes portions of them out into the flow of epikarstic water (if there is any water). Once in the flow of epikarstic water, these chemicals will flow toward subcutaneous drains for rapid transport to the underlying aquifer. If these chemical contaminants reach the transmission zone via some mechanisms other than by subcutaneous drains, then long-term storage characterized by the specific partitioning of the released chemical will dominate the rate of release. If, however, the chemical contaminants reach any vadose shafts underlying the epikarstic zone, then transport of the chemicals to the aquifer will be virtually unimpeded.

These same factors can be expected if the released chemicals flow overland to a sinkhole. Little retardation is likely to occur because sinkholes are drains into the subjacent aquifer. Sinkhole bottoms commonly have large quantities of soil which will help in retarding flow, but, more often, sinkholes are direct feeders to underlying cavernous features that include subcutaneous drains.

Spills to sinking streams will be diluted to a certain extent, but sinking streams also become vadose cave streams that generally become phreatic cave streams. Streams transport large amounts of sediment that will sorb some chemical constituents, but sorption to sediment particles does little to retard contaminant transport because the particles are themselves transported.

Immiscible Liquids

None of the foregoing discussion takes into account the behavior of immiscible contaminants in the subsurface in that they will probably behave in a manner very different from the subsurface water or that of miscible contaminants. Because of density differences, light nonaqueous-phase liquids (LNAPLs) will float on the water surface while dense nonaqueous-phase liquids (DNAPLs) will tend to sink to the bottom of the aquifer. In karst terranes, LNAPLs will not be very susceptible to the effects of dispersion caused by the bifurcation of vadose water along various routes, thus allowing contaminants to become concentrated in major active conduits. In water-filled passages that become constricted during floods, rises in the water surface will cause a decrease in velocity upstream, and globules of floating LNAPLs will be driven into the surrounding rock matrix and up into older, higher levels. DNAPLs will tend to sink rapidly and to collect as sludges on the bottoms of deep pools and to sorb onto microparticles. DNAPL globules will also be driven deep into the rock matrix during flooding and will thus be retained for long periods due to sorption effects. Natural concentration gradients will result in portions of these globules diffusing into the ground water.

Subsequent flooding will drive portions of these organic chemical globules out of the rock matrix and into the main conduit for rapid transport. Sampling experiments by Quinlan and Alexander showed that storm events flush contaminants out periodically for discharge, so sampling of spring waters should be accelerated during storm events and continued at a lesser rate during the flood hydrograph recession [43].

The Special Problem of Sinkholes

Special emphasis must be placed on the likelihood of sinkhole development beneath an existing disposal unit that is either unlined or lined with a synthetic geomembrane. Man-induced sinkhole development, mostly through pressure-head changes, is quite common in many karst terranes. Excessive ground-water pumpage [44] or the diversion of stormwater runoff into existing sinkholes [45] has been known to stimulate additional sinkhole development for many years. Loading, such as the construction of retention basins has caused substantial sinkhole development. It has also been demonstrated that even slightly permeable soils underlying lagoons may allow sufficient moisture retention to develop a pressure head in the soil. This condition can lead to soil piping and catastrophic collapse beneath the lagoon [46].

Sinkhole development cannot be prevented because water flows laterally in both the soil and the epikarstic zone, but it can be minimized by restricting storm water infiltration. Lateral through-flow can allow water to travel distances in excess of hundreds or thousands of meters.

Collapse of surficial materials beneath disposal sites usually results in rapid transmittal of potentially toxic wastes to underlying karst aquifers via subcutaneous drains while still leaving substantial portions of the waste in subsurface storage. Leaching tests and other evidence showing that chemicals cannot leach to the subsurface are inapplicable in these circumstances. In fact, leachate collection systems underneath land disposal units may induce sinkhole collapse beneath the unit if a leak develops in the system. The resulting discharge of leachate, besides contaminating the subsurface, will slowly transport the supporting clastic material away, thus causing a collapse to occur.

Another form of ground-water contamination associated with sinkholes is their use as disposal sites. Some farmers routinely dump empty pesticide containers, livestock carcasses, and other wastes into sinkholes because of the convenience. Large sinkholes have actually been used as landfills for municipal and industrial waste disposal. Direct disposal to sinkholes is equivalent to dumping potentially toxic materials into a funnel leading to a water supply pipeline. This harsh analogy is not unrealistic because of the speed and efficiency with which transport may occur in karst terranes.

REMEDIATION

In karst aquifers dominated by conduit flow, remediation is nearly impossible with current knowledge and technology. Only one spill response can be assured of being effective in karst terranes: Stop using water from affected water-supply wells or springs and provide alternative sources [47, 48]. Traditional aquifer remediation techniques such as ground-water extraction, containment, and biorestoration have little or no known value in conduit-flow-dominated karst aquifers. Extraction wells seldom intersect subsurface conduits, preventing significant withdrawals of contaminants. Even if extraction wells did intersect any conduits, their effectiveness would still be minimal at best. This is because of the inability of the wells to remove sufficient quantities of water from conduits and their inability to have any significant effect on water stored in the rock matrix. Grouting and biorestoration are equally ineffective for essentially the same reasons. Simply, the flow of a conduit-dominated system does not lend itself to remediation by any of the more conventional techniques.

Ground-water flow in a diffuse-flow-dominated aquifer is more amenable to restoration by extraction wells and other methods, depending on the extent to which conduits play a role in the subsurface. Vapor extraction of volatile organics in the soil and epikarstic zones may be fairly effective provided the more contaminated zones can be readily located. The efficacy of vapor extraction wells in the soil zone and particularly in the epikarst zone must not, however, be overstated.

Crawford describes the remediation of a contaminated karst aquifer by removing the source and waiting for the conduit system to flush itself, but he recognizes that this is not enough [50]. This procedure does remove part of the problem, but in no way does it account for chemical contaminants retained in the epikarstic zone, the transmission zone, or the rock matrix feeding the conduits.

A detailed hydrological and geological investigation of a contaminated area may provide sufficient insight into the karst hydrogeology to allow some form of aquifer remediation to be effectively implemented. Evidence for this possibility does exist for some sites, but the actual remediation of a contaminated karst aquifer will always face certain limitations that are uniquely inherent to karst terranes.

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