

Biogeochemical Evolution of Domestic Waste Water in Septic Systems: 1. Conceptual Model

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Abstract

This paper presents a conceptual model, developed by synthesizing the results of many researchers, which describes the geochemical evolution of domestic waste water in conventional on-site septic systems as the result of the interactions of a few major constituents. As described by the model, the evolution of waste water is driven by the microbially catalyzed redox reactions involving organic C and N in waste water and occurs in as many as three different redox zones. Anaerobic digestion of organic matter and production of CO_2 , CH_4 , and NH_4^+ predominate in the first zone, which consists mainly of the septic tank. In the second zone, gaseous diffusion through the unsaturated sediments of the drain field supplies O_2 for aerobic oxidation of organic C and NH_4^+ and a consequent decrease in waste-water alkalinity. The NO_3^- formed by NH_4^+ oxidation in this zone is the primary adverse impact of septic systems at most sites and is generally an unavoidable consequence of the proper functioning of conventional septic systems. If adequate O_2 is not available in the drain field, aerobic digestion is incomplete, and the accumulation of organic matter may cause septic-system failure. In the third redox zone, NO_3^- is reduced to N_2 by the anaerobic process of denitrification. However, this setting is rarely found below septic systems due to a lack of labile organic C in the natural setting. Consideration of the changing redox and pH conditions in each zone aids our understanding of the fate of other constituents in septic systems.

Introduction

The United States uses individual on-site septic systems to dispose of approximately one-third of its domestic waste water (Canter and Knox, 1985, p. 1), and septic systems are also common in Canada. While sewage treatment plants have recently been subject to increasingly stringent treatment requirements and have incorporated sophisticated new treatment techniques, most new septic systems undergo little or no monitoring once they are installed, and their construction follows a basic design first used 100 years ago (Figure 1; Cotteral and Norris, 1969). Since the domestic waste water in septic systems contains many environmental contaminants, septic systems in the United States and Canada constitute approximately 20 million potential point sources for ground-water contamination.

The study of the subsurface behavior of septic-system effluent began many decades ago and has produced an abundant literature. Much of this work has focused on the ability of septic systems to degrade organic matter in waste water and on the prevention of septic-system failures. Recent findings, however, raise issues beyond those of improved septic-system functioning and point to the possible need to diminish our reliance on conventional septic

systems as a common means of waste-water disposal. The most compelling findings are those involving NO_3^- contamination. Nitrate-N has a maximum permissible drinking water concentration of 10 mg/l (Sayre, 1988; USEPA, 1992), but as explained below, properly designed and constructed septic systems frequently cause NO_3^- -N concentrations greater than 10 mg/l in the underlying ground water. Furthermore, septic systems also release other contaminants, such as PO_4^{3-} (Sawhney and Starr, 1977), human pathogens (Hagedorn, 1984), and toxic organic chemical from household products (Kolega et al., 1987). The dilution of NO_3^- and these other constituents in the ground water below septic systems is generally much less than assumed in previous decades. In recent times, the expectations for minimal environmental impact have become much stronger. Accordingly, the release of contaminants from septic systems may be less acceptable than when septic systems gained initial acceptance as the preferred means of on-site sewage treatment.

In general, the subsurface fate of waste water from septic systems has been studied constituent by constituent (Reneau et al., 1989). It is also important, however, to look beyond specific constituents and recognize that septic systems are true geochemical systems, in which the waste water constituents react with each other and with the subsurface gases and porous medium. As explained below, most geochemical changes in waste water occur as a result of the reactions of a few major components that strongly affect the master variables of the system: the redox level and pH. Understanding these major interactions improves our ability to understand the behavior of specific contaminants and to predict the impact of changes in septic-system design.

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This paper presents a conceptual model which outlines the biogeochemical processes that alter domestic waste water as it progresses through conventional septic systems and into the ground water. This model focuses on the reactions that determine the major geochemical characteristics of the waste water, and the unifying concepts of the model are illustrated with examples from the literature. In a companion paper, the model is applied to the evolution of domestic waste water at two extensively studied single-family septic systems in unconfined sand aquifers (Wilhelm and Schiff, in submission).

The Conceptual Model

The geochemical evolution of domestic waste water in a typical septic system is determined by the initial composition of the waste water, the physical arrangement of the septic system (Figure 1), and the composition of the subsurface. Domestic waste water contains relatively high concentrations of organic compounds containing reduced C and N. Microorganisms in septic systems fill their energy needs by catalyzing the oxidation of these reduced compounds, which requires the concurrent reduction of electron acceptors. Oxygen (O_2) is the most energetically favorable electron acceptor, and biogeochemical settings are usefully classified as aerobic or anaerobic according to the presence or lack of O_2 , respectively. The fairly high alkalinity and near-neutral pH of waste water are often altered by these redox reactions.

Septic systems typically develop two major redox environments in which the waste water is transformed. The septic tank, the first redox zone, is an anaerobic setting in

which the concentration of dissolved oxygen (DO) is very low but the concentration of organic matter is high. Microorganisms therefore use electron acceptors such as organic C, H^+ , CO_2 , and SO_4^{2-} to oxidize organic matter and produce CO_2 , H_2 , methane (CH_4), and sulfide (S^{2-}) [Figure 1; equations (1) to (6)]. (All equations are shown in Table 1.) Most large particles are removed in the septic tank, and most N is released from organic molecules as the reduced inorganic ammonium ion (NH_4^+).

The septic-tank effluent flows into the aerobic unsaturated sediments of the drain field, which constitute the second major redox zone [Figure 2(a)]. In this zone, microorganisms use O_2 as the electron acceptor in the oxidation of organic C to CO_2 and of NH_4^+ to NO_3^- [equations (7) and (8)]. In properly functioning drain fields, almost complete oxidation of the reduced waste-water components occurs because adequate O_2 diffuses into the unsaturated zone from the atmosphere. Ammonium oxidation in this zone creates NO_3^- concentrations in the effluent that are roughly two to seven times the drinking water limit. Depending upon the ground-water regime, a distinct plume of ground water with high NO_3^- concentrations may extend for considerable distance from the septic system. In the underlying saturated zone, the waste water typically undergoes very little aerobic oxidation because the slow diffusion rate of O_2 in saturated conditions restricts the availability of O_2 .

The oxidation of NH_4^+ in the aerobic zone releases H^+ [equation (8)]. In poorly buffered sediments, the addition of H^+ is buffered primarily by the alkalinity in the waste water and may cause significant decreases in effluent pH. In buf-

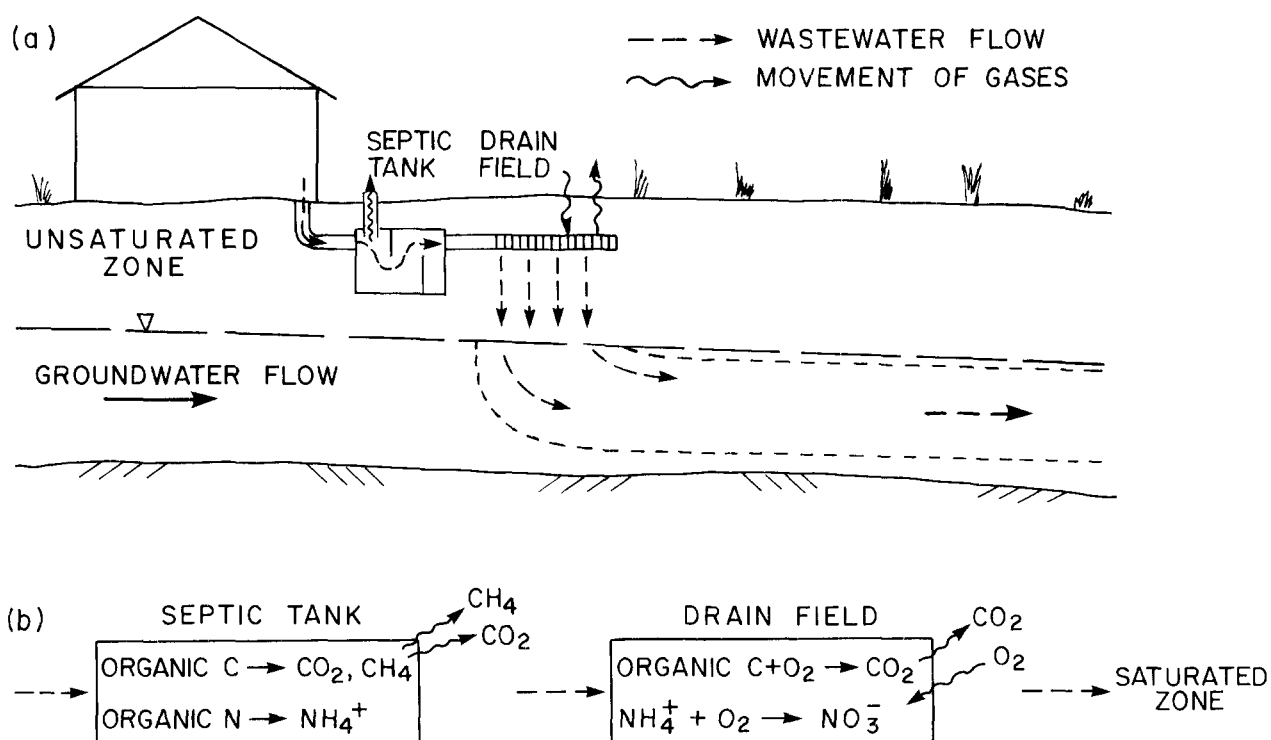
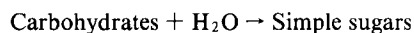
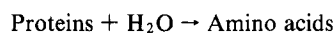


Fig. 1. (a) Schematic cross section of a conventional septic system, including septic tank, distribution pipe, and ground-water plume. (b) Sequence of simplified redox reactions in the two major zones of a conventional septic system: the septic tank and the drain field.

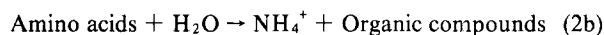
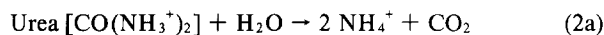
Table 1. Important Reactions in Septic Systems

Anaerobic zone (septic tank and biological mat):

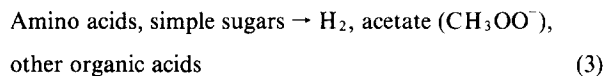
Organic molecule hydrolysis: (1)



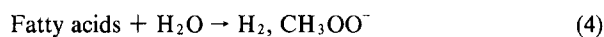
Ammonium release:



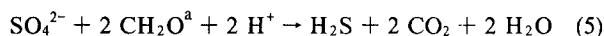
Fermentation:



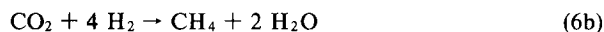
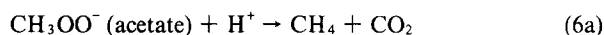
Anaerobic oxidation:



Sulfate reduction:

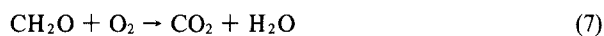


Methanogenesis:

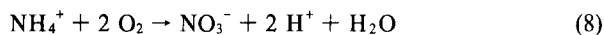


Aerobic zone (unsaturated zone, and saturated zone to lesser extent):

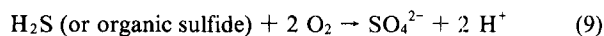
Organic matter oxidation:



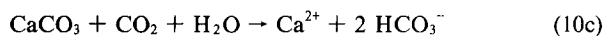
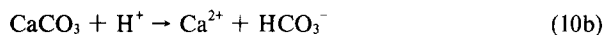
Nitrification:



Sulfide oxidation:

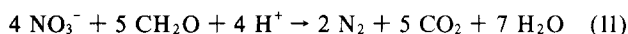


Carbonate buffering:



Second anaerobic zone (saturated or near-saturated conditions)^b:

Denitrification:



^aOrganic matter is simplified as CH₂O throughout. Actual organic matter contains C of various oxidation states and other elements such as N, P, and S, and therefore actual reaction products vary.

^bNitrate reduction can also be accomplished via oxidation of reduced sulfur compounds (Howarth and Stewart, 1992).

ferred sediments, minerals such as calcite dissolve in order to buffer the added acidity, and the pH and alkalinity of the effluent may not change significantly [equation (10)].

The aerobic unsaturated zone and the anaerobic septic tank below the distribution lines are the major areas of

waste-water treatment in septic systems. In properly functioning septic systems, aerobic oxidation causes the largest changes in waste-water composition: the waste water that reaches the water table is depleted in organic C and as a consequence contains NO₃⁻-N instead of NH₄⁺-N. Removal of organic matter is a primary goal of waste-water treatment, but its requirement for abundant O₂ makes the concurrent conversion of NH₄⁺ to NO₃⁻ unavoidable. Depending upon the setting, the pH and alkalinity of the septic-tank effluent at the water table may be lower than in the original waste water.

Some settings also include a third redox zone in which denitrification occurs [equation (11)]. Denitrification requires both anaerobic conditions and an electron donor such as labile organic C and usually occurs in saturated or near-saturated settings. During denitrification, microorganisms use NO₃⁻ as an electron acceptor in the oxidation of organic C. This reaction removes NO₃⁻ from the waste water and produces N₂, CO₂, and alkalinity.

Discussion

As presented above, this conceptual model represents the functioning of a conventional septic system as a straightforward sequence of redox zones and accompanying pH and alkalinity changes and best describes a properly functioning septic system which possesses an appropriate combination of O₂ availability and sufficient residence time in the unsaturated zone. These conditions provide for maximum degradation of organic matter and maximum conversion of NH₄⁺ to NO₃⁻. As documented in the literature, real systems vary greatly in their waste-water loads and physical settings and thus also vary in the degree to which these conversions occur.

Domestic Waste Water

Septic systems receive all the liquid-transported wastes produced by a household, which average approximately 160 liters/person/day in the United States and Canada (Siegrist et al., 1976). The reduced organic compounds and alkalinity are the effluent's most important reactants in subsequent geochemical changes in the septic system.

Domestic waste water contains approximately 0.2 g/l to 0.6 g/l of organic matter by weight, which is contributed by human, kitchen, and cleaning wastes (Tchobanoglous et al., 1991). The organic matter is mostly composed of proteins and carbohydrates and smaller amounts of lipids (Tchobanoglous et al., 1991), but also includes two classes of contaminants: anthropogenic organic chemicals and microbial pathogens. Proteins and urea contribute over 97% of the 20 to 70 mg/l of N typically found in waste water (Laak, 1974; Tchobanoglous et al., 1991).

Together the organic C and organic N comprise most of the oxygen demand of the waste water. The standard tests which measure O₂ demand, the 5-day biochemical oxygen demand (BOD₅) and the chemical oxygen demand (COD) tests, measure primarily the O₂ demand of the organic C, which varies from 200 to 1000 mg/l O₂ (Tchobanoglous et al., 1991). However, the O₂ demand of reduced N in waste water can be an additional 90 to 320 mg/l. Organic S, which

is present in organic matter in a molar ratio of approximately 1 S:35 N (Howarth and Stewart, 1992), will also make a small contribution to O_2 demand. Sulfur is present in domestic waste water in both oxidized and reduced forms at total concentrations of 5 to 10 mg/l (Hypes, 1974; Tchobanoglous et al., 1991).

The pH of domestic waste water typically falls between 6.5 and 8.0 (Canter and Knox, 1985, p. 47; Hunter and Heukelekian, 1965). Generally the alkalinity of waste water is higher than that of the original water supply by 100 to 200 mg/l (expressed as $CaCO_3$; Tchobanoglous et al., 1991).

Anaerobic Zone

The anaerobic zone of a septic system consists of a septic tank and usually a layer of accumulated organic matter directly beneath the distribution pipes which is called the biological mat. This zone provides both physical and biochemical treatment of the waste water. Physical treatment in the septic tank involves the removal of the larger solid particles from the waste water: the denser-than-water particles settle to the bottom of the tank and form sludge while the floating particles are retained behind hanging barriers and form scum. Sludge and scum accumulate in septic tanks at rates of approximately 40 l/person/yr (Trojan et al., 1985; Winneberger, 1984) and must be periodically pumped from the tanks for disposal at landfills or sewage treatment plants. The detention of waste water in the tank also tends to homogenize the flow of waste water to the drain field (Baumann et al., 1978).

Biochemical waste-water treatment in the septic tank is constrained by a limited supply of O_2 . Septic tanks are usually vented and contain an air space within the tank, but the low diffusion coefficient of O_2 in water and a floating scum layer hinder the diffusion of O_2 into the waste water. In a California study, DO concentrations in the waste water of 12 septic tanks ranged from less than 0.1 mg/l to 2 mg/l and averaged 0.3 mg/l (Winneberger, 1984).

In the absence of O_2 , microorganisms obtain energy from fermentation and anaerobic oxidation of organic matter (Gujer and Zehnder, 1983). As observed in sewage treatment plants, microorganisms first hydrolyze large organic molecules in the waste water and in the sludge into simpler molecules such as amino acids, sugars, and fatty acids [equation (1); Mosey, 1983; Tchobanoglous et al., 1991]. Next these smaller molecules are transformed through two different pathways. Amino acids and simple sugars undergo fermentation, in which organic C is both oxidized and reduced, to produce intermediate organic acids, acetate, and H_2 [equation (3); Gujer and Zehnder, 1983]. Fatty acids and intermediate organic acids undergo anaerobic oxidation, in which protons accept electrons and form H_2 [equation (4); Gujer and Zehnder, 1983]. Acetate is also formed by this process. In addition, if SO_4^{2-} is available, microorganisms will use it as an electron acceptor to oxidize organic C and produce CO_2 and S^{2-} [equation (5); Mosey, 1983]. In the final steps of anaerobic digestion, methanogenic bacteria use acetate or CO_2 and H_2 to produce CH_4 and CO_2 [equation (6)]. The occurrence of these reactions is confirmed by the presence of their products in septic-tank gases. Gas

bubbles collected within the waste water of five septic tanks consisted on average of 73% CH_4 , 12% CO_2 , 13% N_2 , and trace amounts of H_2S , O_2 , and H_2 (Winneberger, 1984).

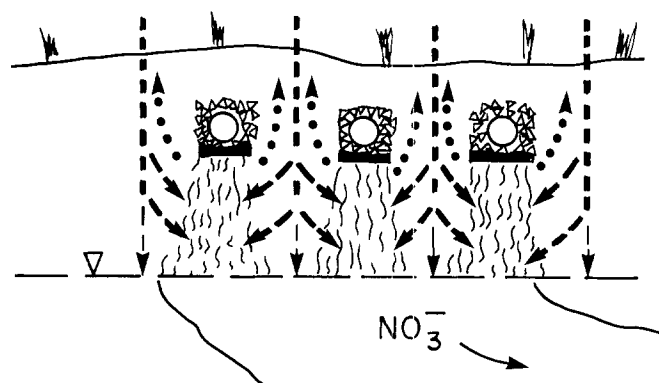
The combination of anaerobic digestion and the removal of solid particles reduces the O_2 demand of the waste water; reported values of BOD₅ reduction in septic tanks range broadly from 7% to 46% (Lawrence, 1973; Trojan et al., 1985; Viraraghavan, 1976). The total N content of the waste water may be reduced by roughly 10% to 30%, mostly due to organic N storage in the sludge (Laak and Crates, 1978; Winneberger, 1984). The degree of treatment is especially affected by waste-water residence time in the tank; removal by both settling and digestion will increase with increased residence time (Winneberger, 1984). Septic tanks are designed to maintain detention times greater than 24 hours even with large accumulations of sludge and scum (Canter and Knox, 1985, p. 15); in a 14-site study, detention times were estimated to range from 0.5 to 8.3 days and averaged 3.1 days (Trojan et al., 1985).

The organic matter that leaves the septic tank differs in structure and composition from the original matter in the waste water. Some of the organic matter may be transformed into larger humic-like organic molecules which are less available for biodegradation. Organic matter in secondary sewage effluent contained over 40% unidentifiable humic-like molecules (Hunter, 1971; Manka et al., 1974), but a similar analysis has not been performed on septic-tank effluent. Most N is stripped from organic molecules in septic tanks [equation (2)]. Roughly 90% of the incoming N, but only 25% of the outgoing N, is found in urea and other organic molecules; the remainder in each case exists as NH_4^+ (Brown et al., 1984; Laak, 1974; Magdoff et al., 1974b).

Anaerobic treatment of septic-tank effluent continues to a lesser degree in the biological mat. If the native sediments are finer-grained than the gravel surrounding the distribution pipes, a 2- to 5-cm thick mat forms over time below the gravel (Anderson et al., 1982). As the waste water enters the finer-grained sediments, suspended particles and organic matter are strained out. The high O_2 demand and moist conditions in this layer lead to the development of locally anaerobic conditions, and FeS deposits often give the biological mat a characteristically black appearance (Jones and Taylor, 1965; McGaughey and Winneberger, 1964). According to a six-year study using soil lysimeters, the rate of biological-mat formation was correlated with the amount of suspended solids and O_2 demand in the effluent (Siegrist and Boyle, 1987).

The biological mat also decreases the seepage rate of waste water in the drain field, especially in coarser-grained sediments. As material accumulates, the hydraulic conductivity of the layer decreases (Jones and Taylor, 1965) which often causes the ponding of effluent above the biological mat. In two field studies, effluent ponding was observed in all 18 septic systems investigated, aged 0.5 years to 13 years; 12 of these systems were located on sands and sandy loams (Bouma, 1975; Walker et al., 1973). In many cases the hydraulic conductivity of the biological mat, not of the soil, becomes the controlling variable in waste-water infiltration rates (McGaughey and Winneberger, 1964).

(a) WELL-AERATED DRAIN FIELD



LEGEND :



CROSS-SECTION OF DISTRIBUTION PIPE IN GRAVEL TRENCH WITH BIOLOGICAL MAT AT LOWER LEVEL

~~~~~ WASTEWATER FLOW

--- O<sub>2</sub> DIFFUSION

..... CO<sub>2</sub> DIFFUSION

- · - CH<sub>4</sub> DIFFUSION

THICKNESS OF LINES INDICATES MAGNITUDE OF FLUX

(b) POORLY-AERATED DRAIN FIELD

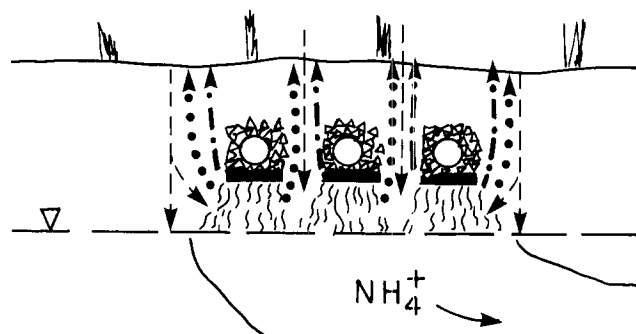


Fig. 2. Gas and water movement in the drain field of septic systems. (a) Well-aerated system, in which adequate O<sub>2</sub> enters the drain field and CO<sub>2</sub> and NO<sub>3</sub><sup>-</sup> are produced. (b) Poorly aerated system, in which adequate O<sub>2</sub> does not enter drain field and CO<sub>2</sub> and CH<sub>4</sub> are produced.

Anaerobic digestion includes reactions which increase as well as decrease the alkalinity of waste water. Production of organic acids by fermentation and H<sub>2</sub>-forming oxidation decrease alkalinity [equations (3) and (4)] while SO<sub>4</sub><sup>2-</sup> reduction and the methanogenic consumption of acetate increase alkalinity [equations (5) and (6)]. In a stable system in which both processes are occurring, the pH will remain close to neutral. However, methanogenesis is inhibited by lower pH, so if organic acid production significantly exceeds acetate consumption, the pH of the waste water will drop precipitously (Grady and Lim, 1980). In addition, the production of CO<sub>2</sub> during anaerobic digestion may lower the waste-water pH if the CO<sub>2</sub> remains in solution.

### Aerobic Zone

The domestic waste water undergoes its most significant geochemical changes in the drain field, where it flows downward from the biological mat to the water table. The effluent load to the drain field is typically 1 to 5 cm/day distributed over the absorption area of the field (Canter and Knox, 1985, p. 31; Walker et al., 1973). The thermodynamic instability of organic matter and NH<sub>4</sub><sup>+</sup> in an oxic setting

drives these changes, and the key factor in the functioning of this zone is the availability of O<sub>2</sub>.

When O<sub>2</sub> is available, microorganisms are capable of almost completely oxidizing the reduced waste-water components in the unsaturated zone [Figure 2(a)]. Most studies have investigated the oxidation of organic C and N in this zone [equations (7) and (8)], but the oxidation of organic S to SO<sub>4</sub><sup>2-</sup>-S is also possible [equation (9)]. The unsaturated zone's ability to provide aerobic oxidation is illustrated most clearly by two laboratory column experiments whose results will be cited in the next two sections. The first experiment included an investigation of the effect of sediment composition on waste-water treatment by applying 1.5 cm/day of septic-tank effluent to columns of limestone sands and silica sands with varying clay contents (Willman et al., 1981). The septic-tank effluent contained an average of 178 mg/l NH<sub>4</sub><sup>+</sup>-N, less than 3 mg/l NO<sub>3</sub><sup>-</sup>-N and NO<sub>2</sub><sup>-</sup>-N, and 296 mg/l COD. In the limestone and shale sand columns, 77% to 92% of the NH<sub>4</sub><sup>+</sup>-N was converted to NO<sub>3</sub><sup>-</sup>-N by the bottom of the 60-cm long columns, and the COD was reduced to less than 50 mg/l. In the silica-sand columns the transformations were less complete, producing 20 to 100

mg/l  $\text{NO}_3^-$ -N and removing 60 to 230 mg/l COD; as discussed later, pH decreases in these columns are partly responsible for this decreased performance. The second experiment used laboratory columns to simulate mound-type drain fields by layering 60 cm of sand and sandy loam over 30 cm of silt loam (Magdoff et al., 1974a). Septic-tank effluent was applied at a rate of 2 cm/day and contained 44 mg/l total Kjeldahl N (TKN, which equals  $\text{NH}_4^+$ -N plus organic N) and 260 mg/l COD. In aerobic columns, the effluents at the bottom of the sand layer contained 41 mg/l  $\text{NO}_3^-$ -N and only 1 mg/l  $\text{NH}_4^+$ -N. The final column effluents contained less than 30 mg/l COD.

Observations of operating septic systems also provide some indication of the extent of nitrification in the unsaturated zone. In three Wisconsin septic systems, the effluent contained 75 to 85 mg/l TKN, while  $\text{NO}_3^-$ -N concentrations in the soil 15 cm below the biological mat were 5 to 15 mg/g (Walker et al., 1973), or roughly 50 to 160 mg/l  $\text{NO}_3^-$ -N in the soil solution, as calculated from estimated soil moisture. In three vacation homes on a barrier spit, septic systems released effluent with an average of 100 mg/l total N during summer occupation and were idle for the remainder of the year (Postma et al., 1992). Immediately preceding occupation, the ground water located 2 m downgradient of the absorption fields contained less than 2 mg/l of  $\text{NO}_3^-$ -N. These concentrations rose to averages of 20 to 50 mg/l  $\text{NO}_3^-$ -N when the homes were occupied, and concentrations as high as 115 mg/l  $\text{NO}_3^-$ -N were observed.

In oxygenated settings, these oxidation reactions occur within the region directly below the biological mat. Both column and field experiments indicate that almost complete  $\text{NH}_4^+$  oxidation can occur within 1 m of the distribution pipes and within a few hours of its exposure to  $\text{O}_2$  (Stewart et al., 1979; Walker et al., 1973; Whelan and Barrow, 1984). The location and degree of organic C oxidation is less easily observed because of the loss of  $\text{CO}_2$  to the atmosphere, but a similarly rapid oxidation is expected. In aerobic trickling filters in sewage treatment plants, aerobic heterotrophs are concentrated near the point of waste-water input, whereas nitrifiers are concentrated deeper in the filter bed (Higgins and Burns, 1975; McCarty and Haug, 1971). In septic systems most oxidation occurs directly below the bottom of the biological mat, and this separation of bacteria according to electron acceptor has not been observed.

As observed in some waste-water treatment settings, plentiful  $\text{O}_2$  in the soil atmosphere does not preclude the occurrence of anaerobic reactions. In particular, denitrification has been observed in reaction vessels with DO concentrations above 0.5 mg/l, even though bacteria generally cease denitrification when directly exposed to greater than 0.2 mg/l  $\text{O}_2$  (Rittmann and Langeland, 1985). In soils, denitrification has been observed in anoxic microzones within soil aggregates (Tiedje et al., 1984; Sexstone et al., 1985). Similar anoxic microzones may also develop beneath septic-system drain fields.

If dissolved  $\text{O}_2$  is present in the effluent when it reaches the water table or if the background ground water contains appreciable dissolved  $\text{O}_2$ , aerobic oxidation may occur in the saturated zone. Signs of aerobic oxidation have been

reported beneath a large septic system in Ontario (Robertson and Cherry, 1992). In this system, DO in ground water decreases from greater than 3 mg/l to less than 0.5 mg/l within 1.5 m of the water table. In this same interval dissolved organic C (DOC) decreases from approximately 5 mg/l to less than 3 mg/l. Although this study suggests the occurrence of aerobic oxidation in the saturated zone, such oxidation can only treat a small fraction of the waste-water  $\text{O}_2$  demand because of limited  $\text{O}_2$  availability.

The pH and alkalinity of the effluent are altered by these aerobic redox reactions. The oxidation of  $\text{NH}_4^+$  affects both pH and alkalinity by releasing 2 moles of  $\text{H}^+$  for every mole of N oxidized [equation (8)]. Oxidation of organic C produces  $\text{CO}_2$ , which decreases the waste-water pH if it remains in solution [equation (7)] but does not affect the alkalinity unless it causes  $\text{CaCO}_3$  dissolution. Oxidation of the 20 to 70 mg/l of reduced N typically found in domestic waste water removes 2.8 to 8.6 meq/l of alkalinity from the solution. This amount often outweighs the alkalinity available for buffering in the waste water, which is typically 100 to 200 mg/l  $\text{CaCO}_3$  or 2 to 4 meq/l alkalinity. In poorly buffered sediments,  $\text{NH}_4^+$  oxidation can completely deplete the effluent's alkalinity and cause large decreases in the pH of the effluent [equation (10a)]. For example, the pH of a septic-tank effluent dropped from 8.0 to 5.4 during aerobic oxidation in a column of silica sand (Willman et al., 1981). In the short term, such large decreases may inhibit microbial activity, especially nitrification. In laboratory cultures, bacteria maintained maximum rates of nitrification at pH as low as 6.0, but activity was greatly reduced at pH 5.5 (McCarty and Haug, 1971). As mentioned earlier, only 10% of  $\text{NH}_4^+$  was oxidized in a sand column at pH 5.4, compared to roughly 60% oxidation in sand columns with pH of approximately 7 (Willman et al., 1981). Over longer time periods the microbial community may adapt to the pH changes.

Significant decreases of pH may not occur if alkalinity from the water supply or from  $\text{CaCO}_3$  and other minerals in the sediments can supply additional buffering [equation (10)]. In the sediment-composition column experiment, aerobic effluent oxidation caused only a 0.3 unit decrease in pH in columns of limestone sand (Willman et al., 1981). The clay added to the silica-sand columns in this experiment was also able to partially buffer the added acidity. The pH of the effluent dropped from 8.0 to 5.4 in the sand columns without clay additions, but only to 6.6 in the column with 12% clay (Willman et al., 1981). Below a seven-year-old septic system, the dissolution of  $\text{CaCO}_3$  from the sediments maintained a soil pH of 9.0 during aerobic waste-water oxidation (Whelan, 1988). These sediments contained over 15%  $\text{CaCO}_3$ . If aerobic oxidation of waste water produces an excess of approximately 1 to 4 meq/l acidity, approximately 225 to 900 kg of  $\text{CaCO}_3$  could be dissolved below a typical septic system (640 l/day flowrate) over a 20-year period.

Organic C and N are also removed from waste water by retention in the sediments beneath the distribution lines. This retention does not transform these constituents, but it does complicate their behavior. In well-aerated settings, most of the reduced components are aerobically oxidized

rather than retained, but retention plays a more important role in poorly aerated systems, as discussed below. Organic C can be retained when organic particles are strained from suspension by the sediments or when dissolved compounds are adsorbed onto solid organic matter and sediments. Sediment retention may be a particularly important sink for organic C that is not easily biodegraded. In a two-year column study simulating an aerobic unsaturated zone, 77% of the influent organic C was oxidized, 9% was retained, and 14% was transported through the column of sand (Thomas and Bendixen, 1969).

Nitrogen is retained as both organic N and  $\text{NH}_4^+\text{-N}$ . In five operating septic systems, the sediments less than 20 cm below the biological mat contained roughly 20 to 300 mg organic N/kg sediment and 5 to 30 mg  $\text{NH}_4^+\text{-N}$ /kg sediment, concentrations which are 5 to 10 times higher than those in the deeper sediments (Walker et al., 1973). Organic N is retained together with organic C, as described above. Ammonium is retained on sediment grains by cation exchange reactions and together with potassium may be strongly "fixed" by vermiculite and similar clays (Nommik and Vahtras, 1982). Several experiments have demonstrated that approximately 50 to 85% of fixed  $\text{NH}_4^+$  may be unavailable or only slowly available to nitrifying microorganisms (Nommik and Vahtras, 1982). Cation exchange and fixation may retain a large fraction of  $\text{NH}_4^+$  when waste water is first introduced to the sediments, causing a delay in the delivery of N as  $\text{NH}_4^+$  or  $\text{NO}_3^-$  to the water table. In columns of shale sand,  $\text{NH}_4^+$  was retained without nitrification for the first seven weeks of effluent dosing, while columns of the limestone sand retained  $\text{NH}_4^+$  for four to nine weeks (Willman et al., 1981). These retention periods are typical of those expected from the concentrations of  $\text{NH}_4^+$  in waste water (20-70 mg/l) and the  $\text{NH}_4^+$  fixing capacities of soils (0.2-4 meq/100 g; Nommik and Vahtras, 1982). The ability of some septic systems or columns to remove N from the effluent may initially be quite high but may decrease over time as the exchange sites equilibrate with the  $\text{NH}_4^+$  concentrations. Once it is oxidized to  $\text{NO}_3^-$ , N is not significantly retained by most sediments (Bohn et al., 1985).

When  $\text{O}_2$  is limited or unavailable, the redox reactions and alkalinity-buffering reactions described above occur only partially or not at all, and organic C and  $\text{NH}_4^+$  loads to the sediments and ground water increase [Figure 2(b)]. An increased load of organic matter can cause system clogging and failure, as discussed in the next section. The lack of adequate aerobic oxidation in the unsaturated zone may be indicated by the presence of significant concentrations of  $\text{NH}_4^+$  in the ground water below the system. For instance,  $\text{NH}_4^+\text{-N}$  concentrations in the ground water downgradient of a large communal septic system were consistently above 40 mg/l, representing 70% of the original TKN concentration of the septic-tank effluent (Siegrist et al., 1983). In two studies which applied waste water to unsaturated laboratory columns, the major N species in the column outflows changed from  $\text{NO}_3^-$  to  $\text{NH}_4^+$  as the surface of the columns clogged and prevented aeration (Magdoff et al., 1974b; Stewart et al., 1979). Application of 2 to 8 cm/day of waste

water to three soil lysimeters caused saturated conditions in which  $\text{NH}_4^+$  moved as a front through the soils at rates of 0.07 to 0.27 cm/day, apparently as cation exchange sites equilibrated with the  $\text{NH}_4^+$  concentration (Brown et al., 1984).

In cases of alternating aerobic and anaerobic conditions, nitrification during aerobic periods may alternate with  $\text{NH}_4^+$  retention during anaerobic periods. This pattern is reflected in large fluctuations of the  $\text{NO}_3^-$  concentrations entering the ground water. Such alternating conditions were found below a leaching chamber which was used for six months and then rested for six months. After one application period began,  $\text{NO}_3^-\text{-N}$  concentrations in the soil solution 0.9 m below the chamber bottom climbed to a peak of 170 mg/l within 20 days, but then fell to less than 30 mg/l after 50 days (Starr and Sawhney, 1980). Since the effluent averaged 102 mg/l of total N, the additional  $\text{NO}_3^-$  was apparently created by nitrification of retained  $\text{NH}_4^+$  during the resting period.

In poorly aerated systems the degree of aerobic oxidation is lower than in well-aerated systems, and the addition of acid is correspondingly reduced. However, anaerobic digestion may cause substantial  $\text{CO}_2$  increases, which may lower pH of the effluent or dissolve  $\text{CaCO}_3$ . Furthermore, the high moisture contents associated with poorly aerated conditions may hinder  $\text{CO}_2$  diffusion to the atmosphere.

### **Oxygen Flux and Consumption**

In order to avoid situations of inadequate oxidation, a drain field must meet most of the  $\text{O}_2$  demand of the septic-tank effluent. Given the typical range of waste-water  $\text{O}_2$  demands (400 to 1500 mg/l) and application rates of waste water (1 to 5 cm/day), drain fields theoretically receive loads of approximately 4 to 75 g  $\text{O}_2$  demand/ $\text{m}^2$ /day. This demand is similar to the  $\text{O}_2$  uptake of surficial soils and much higher than usually found at depth in the soil horizon (Glinski and Stepniewski, 1985, pp. 5, 18). A sediment's ability to supply  $\text{O}_2$  is affected by many factors, including drain-field geometry, depth to ground water below the distribution lines, and the soil texture.

Adequate  $\text{O}_2$  can only be supplied to drain fields by the diffusion of  $\text{O}_2$  through the gaseous phase. Oxygen diffusion occurs much more rapidly in air than in water, due to both the larger diffusion coefficient of  $\text{O}_2$  in air than in water ( $1.9 \times 10^{-5} \text{ m}^2/\text{sec}$  vs.  $1.5 \times 10^{-9} \text{ m}^2/\text{sec}$  at  $10^\circ\text{C}$ ) and the higher equilibrium concentrations of  $\text{O}_2$  in air than in water in contact with the atmosphere (285 mg/l vs. 11.3 mg/l at  $10^\circ\text{C}$ ; Glinski and Stepniewski, 1985, p. 95). A simplified calculation of the  $\text{O}_2$  flux through a soil column 1 m in length whose pore space is either filled with water or air illustrates the huge difference in the ability of the two media to supply  $\text{O}_2$ . Estimating the diffusion coefficients in soil as 50% of the "pure-medium" coefficients at  $10^\circ\text{C}$  and assuming a gradient from saturation to zero across the columns, the calculated flux through air is 236 g/day/ $\text{m}^2$ , while the flux through water is only  $6.7 \times 10^{-4}$  g/day/ $\text{m}^2$ .

Researchers have observed that system clogging progresses faster in continuously inundated columns and septic systems (Jones and Taylor, 1965; Thomas et al., 1966) and

that most failed systems have a history of inundation and anoxia (McGauhey and Winneberger, 1964). The clogging is often due to the development of a biological mat whose hydraulic conductivity is so low as to prevent adequate drainage of the septic-tank effluent (McGauhey and Winneberger, 1964). In recognition of the failure of septic systems in saturated settings, most septic-system regulations require a minimum vertical separation of 0.6 to 1.2 m between the bottom of the gravel surrounding the distribution pipes and the water table (Canter and Knox, 1985, p. 26).

Unsaturated conditions, however, do not necessarily provide adequate  $O_2$ . Effective gaseous diffusion requires continuous air-filled pore space, which corresponds to air-filled porosities of roughly 10% in soils (Bridge and Rixon, 1976; Glinski and Stepniewski, 1985, p. 55). The drainage of waste water through sediments hinders  $O_2$  diffusion by filling pore spaces with water and also by depositing organic matter, which increases the residual moisture content of the sediments and decreases soil porosity. For instance, the field capacity of a column of Ottawa sand, originally 1%, increased to 5% after it was seeded with municipal sewage and dosed with soluble substrates for more than six weeks (Pincince and McKee, 1968).

The composition of the soil atmosphere demonstrates the adequacy of aeration in drain fields. In the mound-system column experiment,  $O_2$  decreased from 18.5% to 1.7% in the atmosphere of the sand layer once a biological mat formed at the surface of the solid-walled columns. At the same time nitrification virtually ceased (Magdoff et al., 1974a). In similar columns,  $O_2$  in the sand layer remained above 20.1% and nitrification continued after a biological mat formed because the column walls had been perforated to allow lateral gaseous diffusion (Magdoff et al., 1974a). In the large communal septic system the gaseous  $O_2$  concentration fell below 3%, and  $CH_4$  concentrations were approximately 19% in the center of the 1200 m<sup>2</sup> drain field (Siegrist et al., 1983). The air-filled porosity in the field was estimated to be only 2% to 6% in sediments 10 cm below the infiltration surface and 15% at a depth of 15 cm to 20 cm.

### **Second Anaerobic (Denitrification) Zone**

The aerobic zone provides basic waste-water treatment by greatly reducing the  $O_2$  demand of the effluent, but the oxidation of  $NH_4^+$  to  $NO_3^-$  in this zone creates a potential health hazard. Excessive  $NO_3^-$  may also cause eutrophication in coastal marine surface waters such as estuaries (Ryther and Dunstan, 1971). Nitrate can be removed from solution by denitrification [equation (11)]. Denitrification reduces  $NO_3^-$  to  $N_2$ , with a small percentage of  $N_2O$  being produced as well (Seitzinger, 1988). Nitrogen gas is unavailable to most organisms. Bacteria capable of denitrification are commonly found in many subsurface environments, even when denitrification is not actively occurring (Ghiorse and Wilson, 1988; Keeney, 1986). These bacteria require anoxic conditions to reduce  $NO_3^-$ ; such conditions are best maintained in saturated or near-saturated sediments due to the limited  $O_2$  diffusion in such settings. In three alluvial soils, the denitrification rate decreased as the air-filled

porosity increased, and denitrification ceased as the environment became oxidizing at air-filled porosities of 11% to 14% (Pilot and Patrick, 1972). Stoichiometrically, denitrification requires nearly equal parts of organic C and  $NO_3^-$ -N, but additional organic C may be consumed in other reactions occurring simultaneously and may be incorporated into biomass (McCarty and Haug, 1971). As seen in equation (11), the alkalinity and pH of the solution increase during denitrification.

In ground-water settings, a lack of labile organic C is the most common limitation to denitrification (Keeney, 1986). Complete denitrification of the 20 to 70 mg/l  $NO_3^-$ -N expected in septic-system effluent requires at least equal amounts of organic C. Potentially labile organic C may be supplied by the waste water or by the porous medium. Unfortunately, most organic C in the waste water is consumed by aerobic oxidation at approximately the same time that  $NO_3^-$  is created, so that very little waste-water organic C remains for consumption in denitrification. Indeed, denitrification units in waste-water treatment plants commonly add methanol as a source of organic C to compensate for the low organic C concentration in the waste water (Tchobanoglous et al., 1991).

The natural setting may supply organic C from the ground surface or from solid organic matter in the aquifer. In agricultural fields with shallow water tables, the ground surface supplied organic C for denitrification of fertilizer  $NO_3^-$  in the ground water (Gillham, 1991), but this source of organic C is probably not constant and concentrated enough to denitrify septic-system effluent. As for solid organic matter, the amount of organic C in subsurface sediments is often very small, generally less than 0.5% (w/w) in sand aquifers (Mackay, 1990), and much of this solid organic matter may be very old and resistant to biodegradation.

In septic-system research, denitrification has been observed in only a few settings. In these settings, organic matter in the native soil supplied the labile organic C. When 3.3 cm/day of waste water was applied to a soil column, denitrification occurred in a saturated clayey-sand layer which contained 5% organic C (Stewart et al., 1979). Approximately 90% of the waste-water  $NO_3^-$  was removed in the first 42 days of waste-water application, but removal decreased to less than 30% after 95 days. Potentially long-term denitrification ability was observed in a young sand aquifer with an exceptionally high solid organic C content (2.5% w/w), which has been receiving and denitrifying septic-system effluent since 1971 (Robertson and Cherry, 1992). If  $SO_4^{2-}$  is present in the effluent,  $SO_4^{2-}$  reduction [equation (5)] may occur following the consumption of  $NO_3^-$  by denitrification.

### **Fate of Other Constituents**

Other waste-water constituents are influenced by the major changes in redox and pH conditions that occur in the reaction zones of septic systems. For instance, phosphate is present in septic-system effluent at average concentrations of 10 to 20 mg/l (Laak, 1974; Whelan and Titmanis, 1982), concentrations 1000 times higher than those found in natu-



ral surface waters. Fortunately  $\text{PO}_4^{3-}$  is often immobilized within a few meters of the distribution pipes in septic systems (Reneau et al., 1989), and significant movement of  $\text{PO}_4^{3-}$  from septic systems is relatively rare. In aerobic settings,  $\text{PO}_4^{3-}$  interacts with and is retained by iron and aluminum oxyhydroxides in acidic environments (Bohn et al., 1985) and by  $\text{CaCO}_3$  in basic environments (Doner and Lynn, 1989). In general,  $\text{PO}_4^{3-}$  retention is greater in acidic settings than in neutral or basic settings (Tofflemire and Chen, 1977), so the possible lowering of soil pH in drain fields is not expected to increase  $\text{PO}_4^{3-}$  mobility. Phosphate behavior under reducing conditions is less clear. Phosphate is not reduced itself, but the oxyhydroxides with which it associates in acidic environments may be reduced if conditions change from aerobic to anaerobic. Phosphorus release under anaerobic conditions has been observed in surface-water sediments (Fillos and Molof, 1972).

Since major cations do not undergo changes in oxidation state, their behavior in septic systems is only indirectly affected by the redox changes. As mentioned previously,  $\text{CaCO}_3$  is often dissolved in drain fields in order to buffer the acidity released during  $\text{NH}_4^+$  oxidation, which results in increased  $\text{Ca}^{2+}$  concentrations in the effluent. Other cations may also be released from the solid phase during buffering reactions such as mineral dissolution or cation exchange (Bohn et al., 1985).

Trace metal cations are more directly affected by the redox changes in septic systems as well as by the pH changes. Trace metals such as Cu, Cr, Pb, and Zn are found at concentrations in the range of 2 to 300  $\mu\text{g/l}$  in many domestic waste waters (Kaplan et al., 1987; Klein et al., 1974), but their behavior in septic systems has not been investigated. Studies of raw sewage in sewage treatment plants and thermodynamic calculations show that most trace metals are present in the particulate fraction of waste water and that many are likely to form insoluble sulfides (Chen et al., 1974; Kaplan et al., 1987; Morel et al., 1975). Thus, in septic systems, a large fraction of most metals should be retained in the septic tank. Metals that prefer dissolved forms, such as Mn and Ni, and small fractions of other metals will be released from the septic tank. The oxic conditions in the unsaturated zone may cause precipitation of some of these metals as hydroxides. However, if low pH conditions develop below a septic system, hydroxide formation will be less likely (Stumm and Morgan, 1981), and low pH may even mobilize metals present naturally in the sediments in much the same way that acid rain mobilizes aluminum (Nordstrom and Ball, 1986).

Domestic waste water contains pathogenic bacteria and viruses which are capable of causing disease if ingested, and overflow and seepage of waste water is a major cause of disease outbreaks associated with ground water (Craun, 1985). The potential of a pathogen to cause disease once introduced into the subsurface environment is influenced by its abilities to survive and to move. Bacteria and virus survival in the subsurface is on the order of a week to several months and decreases with increasing temperature and decreasing soil moisture (Gerba and Bitton, 1984). In septic systems the steady input of waste water may provide enough

moisture to the soil that moisture content is always adequate for pathogen survival (Parker and Mee, 1982). Bacteria are retained in septic systems primarily by straining, and the biological mat seems to be most important in this process (Hagedorn, 1984). As variably charged particles, viruses are more affected by adsorption onto clay particles, and thus virus retention will decrease as the permeability of soils increases (Gerba and Bitton, 1984). In general, the mobility of bacteria and viruses is much greater in saturated flow than in unsaturated flow (Lance and Gerba, 1984; Rahe et al., 1978), making unsaturated conditions below septic systems desirable for both  $\text{O}_2$  supply and pathogen retention.

Organic chemicals in household products, such as toluene and methylene chloride, are often detected in domestic waste water in concentrations of 10 to 100's of parts per billion (Kolega et al., 1987; Viraraghavan and Hashem, 1986). The biodegradability of many organic contaminants depends on the redox conditions. In one group of experiments halogenated aliphatics were biodegraded under methanogenic conditions, such as exist in septic tanks, but persisted under aerobic conditions (Bouwer and McCarty, 1984). On the other hand, aromatic organics persisted under anaerobic conditions but were biodegraded under aerobic conditions (Bouwer and McCarty, 1984), such as exists in the unsaturated zones of septic systems. Since the sorption of hydrophobic organic contaminants is dominated by interactions with solid organic matter (Karickhoff, 1984), the accumulated organic matter in the septic tank and the drain field may be important retainers of anthropogenic organic contaminants. In addition the unsaturated conditions in drain fields should allow loss of the more volatile organic compounds through volatilization and diffusion.

## Implications

According to the conceptual model, septic systems and natural settings which receive septic-system effluent will provide one of three possible levels of treatment for the major waste-water constituents, organic C and N. These levels of treatment determine the geochemical conditions of the septic-system zones. Because the septic tank performs similarly in most systems, the degree of waste-water treatment is determined primarily by the waste water's reactions in the sediments below the distribution pipes.

The smallest degree of treatment occurs when the supply of  $\text{O}_2$  is limited in the drain field. In this case aerobic oxidation does not proceed. The solid organic C and adsorbed  $\text{NH}_4^+$  contents of the sediments may increase, and plumes of DOC and  $\text{NH}_4^+$  may form in the ground water. The increases in solid organic C may clog the soils and the distribution pipes sufficiently to prevent the downward flow of effluent. The waste water will experience little decrease in alkalinity, but the production of  $\text{CO}_2$  via anaerobic reactions may lead to pH drops and possibly  $\text{CaCO}_3$  dissolution.

The second possible level of treatment is that which probably occurs in most properly functioning septic systems. In these systems the availability of  $\text{O}_2$  in the unsaturated zone leads to the aerobic oxidation of organic C and  $\text{NH}_4^+$ . The effluent that reaches the ground water is depleted

in organic C, and N exists primarily as  $\text{NO}_3^-$  rather than organic N and  $\text{NH}_4^+$ . In well-buffered settings  $\text{CaCO}_3$  dissolves, and pH and alkalinity decreases are small; in poorly buffered settings the waste water experiences significant pH and alkalinity decreases. Given the original N content of the waste water, the  $\text{NO}_3^-$  concentrations in the ground water will often exceed drinking water standards beneath the distribution lines and even at considerable distance from the drain field in many cases.

The most complete treatment of waste water occurs when a natural anaerobic setting which is capable of denitrification follows the aerobic treatment zone. In this case, not only organic C but also  $\text{NO}_3^-$  is removed from the waste water. Denitrification also increases the alkalinity of the waste water.

If the first level of treatment occurs, problems with a septic system will arise quickly. In contrast, the second and third scenarios may attain quasi-steady-state conditions, in which organic matter and oxygen are converted to  $\text{CO}_2$ ,  $\text{NO}_3^-$  or  $\text{N}_2$ , and  $\text{SO}_4^{2-}$  for years or even decades. However, small changes over time may still affect the functioning of these systems. Three possible changes come to mind. First, a rising ground-water table or increased moisture in the drain field due to the buildup of organic matter may decrease the availability of  $\text{O}_2$ , thus increasing areas of anoxia and decreasing the ability of the system to provide aerobic treatment. Second, a gradual loss of buffering material in the subsurface and the accompanying drop in pH may increase metal mobility or temporarily inhibit microbial activity. Third, a system which is originally capable of denitrification may consume all the labile organic C and cease to remove  $\text{NO}_3^-$  from the effluent.

Current regulations on septic-system design and siting try to avoid inadequate waste-water treatment by emphasizing the hydrologic aspects of waste disposal. The main siting and design parameter is the percolation rate of the soil, a parameter whose actual physical meaning is difficult to define (Cotteral and Norris, 1969). Based upon this rate, waste-water loading rates are prescribed that should provide unsaturated flow. As other researchers have pointed out, this approach fails to recognize that the infiltrative capacity of the biological mat, not the soil's properties, usually defines the system's ability to transmit waste water (Cotteral and Norris, 1969; Thomas et al., 1966).

The regulatory approach indirectly recognizes the need for  $\text{O}_2$  in its requirements for unsaturated flow, but the geochemical perspective of the conceptual model highlights the importance of  $\text{O}_2$  in septic-system functioning. Without adequate  $\text{O}_2$ , anaerobic conditions in the drain field cause more severe clogging and poor waste-water treatment, especially of organic matter. Regulations that more directly consider  $\text{O}_2$  availability in septic-system design would favor designs that allow for more "sidewall" diffusion of  $\text{O}_2$  in the unsaturated zone. For instance, trench designs allow more diffusion than bed designs. In addition, consideration of the  $\text{O}_2$  supply suggests that waste-water strength as well as volume should be factored into design parameters. In particular, nondomestic facilities such as rural gas stations, restaurants, and even industries often rely on septic systems

and produce waste water with different  $\text{O}_2$  demands than domestic households.

The conceptual model also draws attention to the production of  $\text{NO}_3^-$  by septic systems and the unlikelihood of  $\text{NO}_3^-$  removal. As outlined in the conceptual model, conventional septic systems unavoidably create  $\text{NO}_3^-$  in the aerobic zone. In the past, advocates of conventional septic systems have relied upon "total mixing models" of ground-water dilution to ameliorate high concentrations of  $\text{NO}_3^-$  (Tolman et al., 1989), but recent findings about ground-water plumes from septic systems and the use of septic systems in more densely populated areas make this approach questionable. In settings such as sand and gravel aquifers and fractured bedrock, low dispersion often maintains high  $\text{NO}_3^-$  concentrations for considerable travel distances.

Unfortunately,  $\text{NO}_3^-$  removal from waste water by denitrification is rare in aquifers below septic systems. In particular, denitrification in most aquifers is limited by the lack of a long-term supply of labile organic C. Regulations should recognize the substantial addition of  $\text{NO}_3^-$  to ground water from conventional septic systems and work to protect valuable ground water from  $\text{NO}_3^-$  contamination. Either another form of waste-water disposal should be required, or alternative systems which provide the proper sequence of redox zones and organic C supply to remove  $\text{NO}_3^-$  (Ritter, 1988) should be installed in  $\text{NO}_3^-$  sensitive ground-water areas.

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