Decision making — Is bioremediation a viable option?*

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Abstract

Bioremediation, as it pertains to hazardous wastes, is a process technology that uses microorganisms to degrade organic chemicals of interest. Often, the biodegradation can occur without transferring the contamination from one part of the environment to another, as is often the case with other remediation methods. Air stripping of contaminated ground water, incineration of contaminated material, and even carbon adsorption all contribute to cross-media pollution by producing hazardous residues, often more concentrated than the original form, that must be disposed. The diversity of bioremediation technologies ensures that one or more of them may be suitable for at least part of a remediation scheme. With our increased understanding of subsurface processes, we are shifting from the treatment of effects, i.e., treatment of contaminated ground water, to the treatment of contaminant sources.

Introduction

Bioremediation is a term bandied about with great frequency in the current parlance. Before we begin discussion of its viability, we should first address the question: What is bioremediation? Bioremediation, as it pertains to hazardous wastes, is a process technology that uses microorganisms to degrade organic chemicals of interest. Often, the biodegradation can occur without transferring the contamination from one part of the environment to another, as is often the case with other remediation methods. Air stripping of contaminated ground water, incineration of contaminated material, and even carbon adsorption all contribute to cross-media pollution by producing hazardous residues, often more concentrated than the original form, that must be disposed. As a sole treatment process, bioremediation may or may not be able to achieve the concentration levels, in either water or soil, specified by regulatory agencies. If one is under pressure to do something because a perceived or real risk exists, bioremediation will not provide a quick fix nor will it serve as a panacea for all or

^{*}Paper presented at the GCHSRC Third Annual Symposium: Bioremediation, Fundamentals and Effective Applications, Lamar University, Beaumont, TX, USA, February 21–22, 1991. ¹To whom correspondence should be addressed.

even most waste problems. Bioremediation is a technology that may be appropriate as part of a treatment train or site remediation protocol, depending on the objectives of the initiative. When trying to decide whether or not to exploit a bioremediation method, one will use a decision making process that is quite similar to any other decision making process, but the specific issues will differ. To impart a structure to the decision making process, we believe the following questions must be addressed: (1) What bioremediation methods are available for consideration? (2) Under what conditions should bioremediation be considered at a waste site? (3) Which bioremediation method is most appropriate? (4) Is bioremediation cost effective, especially compared to other nonbiological methods? After the bioremediation methods are described in more detail, the applicability of bioremediation in general and of the different treatment types in specific will be examined.

Bioremediation processes

What bioremediation methods are available for consideration? Bioremediation is a diverse technology that includes the following methodologies: *in situ*, bioreactors, and bioventing. Other classifications could, and, perhaps should, be used since all processes result in bioreactors and the differences between *in situ* and on-site are often vague. Nevertheless, these categories should suffice to focus this discussion. The analysis presented here will often be lacking in quantitative details because much of the literature about bioremediation processes is either proprietary or is published in the gray literature and has not been subjected to the peer review process for criticism of the experimental methods or the validity of the results obtained.

In situ biorestoration is a variation of pump-and-treat technology, with the biological treatment occurring in the subsurface environment. Ideally, contaminants dissolved in the ground water and present on the soil matrix are both degraded by the indigenous microorganisms; however, it is most effective for the biodegradation of dissolved contaminants. In situ biorestoration has been used primarily at fuel contaminated sites; the decrease in aqueous hydrocarbon concentration is well documented [1], but there is a paucity of information about its utility for decreasing the hydrocarbon concentration in subsurface solids. For *in situ* bioremediation, inorganic nutrients and an electron acceptor, usually oxygen, are added to the contaminated site with either injection wells or an infiltration gallery; ground water is recovered by production wells for recirculation to the treatment zone or disposal to the sanitary sever.

The bioreactors category consists of methods that use either the soil matrix, the ground water, or a combination of the two as the substrate and includes the following methodologies: conventional land treatment with and without excavation, composting of contaminated material, liquids-solids contactors, and withdrawal of ground water and treatment in a specialized reactor. In-land treatment (or farming), the soil is tilled and nutrient levels, moisture content, and pH are maintained at optimum levels. Land treatment may require a liner and leachate collection and leak detection systems to meet regulatory requirements. Land farming has been used to treat a variety of wastes, including fuels, polynuclear aromatic hydrocarbons (PAH) and pesticides. A joint project between two consulting firms and a county agency was conducted under the auspices of the Florida Department of Environmental Regulation to determine the utility of land farming diesel contaminated soil [2]. The state set a goal of 5 mg/l total petroleum hydrocarbons (TPH) in the soil. In three small test plots, the TPH concentration was decreased by an average of 2523, 2707, and 2930 mg/l in each plot at the conclusion of the six-week deadline. Although the cleanup goal was not met in the time allotted, the authors believed that it might have been, had the study continued. In another land treatment study, phenanthrene was reduced 79%, 86%, 92% and 78% in plots with no amendments, nutrient addition, nutrient and one microbial enrichment. and multiple microbial inoculations to yield final concentrations of 5.73 mg/kg, 2.71 mg/ kg, 5.75 mg/kg, and 5.28 mg/kg, respectively, after 94 days of treatment [3]. The compounds of primary concern, 2, 4-dichlorophenoxyacetic acid (2,4-D) and 4-chloro-2-methylphenoxyacetic acid (MCPA), at a site contaminated with pesticides, were reduced from 41.8 and 44.2 mg/kg, respectively to 4.0 and 1.2 mg/kg within 77 days, which was within the regulatory limit of 10 mg/kg [4].

Composting allows the degradation of waste material at the elevated temperatures produced by accelerated biological activity. The contaminated material is combined with a bulking agent to ease the mixing requirement and aid in oxygen transfer. Aeration and irrigation ensure adequate nutrient and moisture control; the reactor can be enclosed to minimize the emissions of volatile organic compounds.

Liquids-solids contactor reactors (LSC) treat a soil slurry in a batch process; the quantity of soil in the slurry depends on the soil type and the amount of aeration and mixing that can be produced by the system. The LSC maximizes the mass transfer rates and the contact between microorganisms and the contaminated material; it can attain high contaminant removal rates [5]. A LSC is a self-contained unit, thereby eliminating the concerns about leaching (and possible regulatory restrictions) that limit land treatment and can be designed to produce faster biodegradation rates than are possible with conventional land farming. Wood-preserving wastes, which are PAHs, and oil sludges have been treated successfully with LSCs [6].

The withdrawal of contaminated ground water followed by above ground biological treatment is reminiscent of standard aerobic wastewater treatment technologies such as activated sludge and fixed film reactors. Above-ground biological treatment of contaminated ground water can reduce aromatic hydrocarbons by 99 percent; high concentration influent can be reduced to $30-50 \mu g/l$ in the effluent and low concentration influent can be reduced to $5-10 \mu g/l$

l in the effluent [7]. The use of two reactors in series reduced an influent concentration of 300 mg/l methyl ethyl ketone in a contaminated ground water to an effluent concentration of $1-5 \mu g/l$, which was well within the required 1 mg/l limit for discharge to the local sanitary sewer [7].

Bioventing is a variation of vapor or vacuum extraction and is also an in situ technology. In bioventing, the degradation of fuel hydrocarbons located in the vadose zone is stimulated by the injection of air. Several possible configurations exist for the air injection and vapor extraction process [8]; the optimum design maximizes the oxygenation rate for biodegradation but allows sufficient retention time so that volatile organic compounds (VOCs) are biodegraded, rather than volatilized as a contaminated offgas. By biodegrading the VOCs before they are removed from the unsaturated zone, the need for treating the offgas is minimized; treating a contaminated offgas can account for 50% of the cost of a soil venting project [9]. Negative pressure may be provided by a vacuum pump; aeration through the vadose zone may occur passively or by injecting compressed air. A seven month field study of bioventing at a jet fuel (JP-4) contaminated site at Tyndall Air Force Base, FL, demonstrated that biodegradation was responsible for the removal of 32 kg of hydrocarbon (55%) and volatilization removed 26 kg of hydrocarbon in one test plot [9]. By optimizing the air flow rate in another test plot, the fraction of VOCs biodegraded was increased to 82%. Biodegradation rates through contaminated plots averaged 5 mg/kg·day; through an uncontaminated plot fed contaminated off-gas, the biodegradation rate was 1.34 mg/kg·day.

Biological considerations

Is the waste known to be biodegradable? Clearly, if the contaminants present are not biodegradable, bioremediation is not the proper course of action. Bioremediation requires microorganisms with suitable degradative capabilities. When in situ treatment is used, adapted microorganisms are often, but not always, present at the contaminated site. Typically, a laboratory screening of the waste material will indicate whether biodegradation is possible. If the indigenous population is unable to degrade the targeted chemicals, the potential exists for adding acclimated or specialized (genetically engineered) microorganisms to the waste material. Acclimated microorganisms have been beneficially employed in bioreactor technologies. For example, treatment of ground water contaminated with tetrahydrofuran and isopropanol in an activated sludge unit provided an adapted inoculum for subsequent injection to the subsurface in a combined above-ground bioreactor and in situ bioremediation project [10]. In a laboratory study, inoculation of soil with Arthrobacter sp. resulted in accelerated rates of pentachlorophenol degradation compared to uninoculated controls [11]. The white rot fungus (Phanerochaete chrysosporium), when under nitrogen limitation, secretes an enzyme system capable of degrading a number of otherwise recalcitrant compounds, such as pentachlorophenol, 3-5 ring polynuclear aromatic hydrocarbons, and DDT [12-14]. The addition of white rot fungus to a land treatment project is a potentially promising technology. Many microbiological preparations are commercially available which contain either bacteria or bacteria and a nutrient mix for application to contaminated material, but quantitative evidence of the effective-ness of these preparations is generally lacking.

Can the process be stimulated? Enhancement of natural degradative processes is critical to the success of bioremediation. Some transformations will not occur without amendments; enhancing the process generally increases the rate, reduces the risk of an exposure, and saves time and money. Stimulation of biodegradation is accomplished by increasing temperature, adding nutrients, a terminal electron acceptor, a carbon source, or, some combination of these treatments. Temperature affects the growth rate of bacteria; within a certain range, enzyme activity doubles with each temperature increase of 10°C [15]. Increasing the temperature to increase the rate of biodegradation has been incorporated into bioventing projects at hydrocarbon contaminated sites located above a shallow water table aquifer in New Mexico [16].

Mineral nutrients, in particular nitrogen and phosphorus, are often limiting in the subsurface environment and must be supplied to ensure degradation of a large contaminant mass. For example, Mulkins-Phillips and Stewart found that phosphorus limited the rate and extent of growth of a Nocardia sp. on 1% v/v Bunker C fuel oil [17]. Prior to beginning an in situ bioremediation project in Ambler, PA [18], laboratory experiments indicated that the native microflora could be stimulated by the addition of inorganic nitrogen, phosphate salts, and air [19]. The carbon: nitrogen: phosphorus ratio necessary to enhance bioremediation can vary from 100:10:1 to 100:1:0.5, depending on the methodology used and the location of the contaminant (liquid or solid phase) [5]. Nutrient addition is not always necessary, however. Miller et al. [9] found that nutrient addition did not significantly affect the rate of biodegradation in test plots undergoing bioventing because the soil matrix contained adequate amounts of nitrogen and phosphorus. Aerobic biodegradation is usually preferred because it is faster than anaerobic processes. As an example, the biodegradation rate constant for carbofuran in soil is 0.047/day aerobically and 0.026/day anaerobically [20]. In aerobic bioremediation, the ratio of oxygen mass to hydrocarbon mass required for complete degradation to CO₂ (mineralization) has been estimated to range from the 3:1 ratio used in the BIOPLUME II model [21] to 1.08-1.7 when the simultaneous production of cell mass is considered [22].

Methods for increasing oxygen availability include air sparging, addition of pure oxygen, and addition of hydrogen peroxide (H_2O_2) during *in situ* bioremediation; tilling during land treatment; forced aeration or mechanical rotation of a compost pile; and aeration during bioventing. The success of oxygen addition varies with the treatment method. The low solubility of oxygen in water limits the amount of oxygen that can be provided during *in situ* biorestoration. Raymond observed stimulation of hydrocarbon-degrading bacteria and suggested that 1000 bbls (159 000 l) of gasoline in ground water were degraded by supplying compressed air [18].

The use of hydrogen peroxide as an oxygen source has had mixed results. By using H_2O_2 as a supplemental oxygen source, Yaniga and Mulry successfully reduced the absolute concentration of gasoline sorbed in a sandy aquifer from the 3700 mg/l to 7200 mg/l range to the 2300 mg/l to 2900 mg/l range [23]. At a field experiment at Eglin Air Force Base, FL, using H_2O_2 at an initial concentration of 500 mg/l, problems with off-gassing and flow impedance were observed and attributed to microbial degradation of H_2O_2 [24]. The patented method for H_2O_2 amendment in bioremediation calls for pretreatment of an aquifer with phosphate compounds to minimize mineral decomposition of peroxide in the formation and a gradual increase in H_2O_2 concentration to permit microbial adaptation [25]. Using the patented method, flow disturbances were not observed during a field study at Traverse City, MI, where the H_2O_2 concentration was gradually increased [1], but oxygen gas was evolved faster than it could be used biologically or solubilized [26].

Addition of air to waste material in land treatment, in composting, and to the vadose zone in bioventing to increase oxygen availability is more successful because it avoids the mass transfer problems; also, gases are capable of greater diffusion through a formation than are liquids. The tilling process in land treatment promotes oxygenation, oil-soil interaction, and disruption of aggregates. Recent laboratory research indicates that disruption of aggregates before application of waste material to soil improves biodegradation [27].

Alternate electron acceptors have also been used in bioremediation; nitrate addition is a method of supplying both a mineral nutrient and an electron acceptor. After 120 days of treatment with nitrate, 7.5 tons of hydrocarbons were degraded; the decrease was observed in ground water samples and in contaminant concentration and distribution patterns in the subsurface [28].

The addition of a carbon source is imperative when the degradative process is cometabolic. The predominant contaminants which can be degraded through a cometabolic process are the chlorinated aliphatics, such as trichloroethene (TCE). TCE is metabolized when either methane is supplied as a primary carbon source [29,30] or when phenol or toluene is the carbon source [31,32]. In a field demonstration at Moffett Naval Air Station, methane addition was required for biodegradation of dissolved chlorinated ethenes introduced into an injection well [33]. Two meters down gradient, the concentration of TCE was diminished by 20–30% and vinyl chloride was reduced by 90–95%; when stimulation ceased, 95% of the injected chlorinated ethene mass was detected in observation wells. The cometabolic degradation of chlorinated aliphatics is now being tested as a Superfund Innovative Technology Evaluation (SITE) emerging technology [34]; practical application and process efficacy remain to be demonstrated.

Chemical considerations

What contaminant properties influence the decision making process? In addition to biodegradability, waste characteristics that determine the suitability of bioremediation as a treatment technology include concentration, bioavailability, and the toxicity of the parent compound or its metabolites. One must ensure that biodegradation of the parent compound does not result in the evolution of a more toxic, less biodegradable metabolite. Vinyl chloride is produced from the anaerobic degradation of TCE; fortuitously, vinyl chloride can be degraded in an aerobic, cometabolic process [3]. Likewise, if a chemical is present at an elevated, toxic concentration or if another toxicant is present in the waste material, the site may be unsuited to bioremediation. Preliminary biodegradation testing should always precede field trials of bioremediation so that the adverse effects of toxicants or elevated concentrations can be detected. Laboratory experiments using soil microcosms demonstrated the adverse effect of high concentrations of tertiary butylphenyl diphenyl phosphate (BPDP). Addition of 0.56 mg/l of BPDP resulted in the greatest mineralization; addition of 5.6 mg/l and 56 mg/l of BPDP inhibited mineralization [35]. Conversely, low concentrations of contaminants might preclude biodegradation, implying that a threshold concentration exists for biodegradation. Initial 2,4-dichlorophenoxyacetate (2,4-D) concentrations of 22 mg/l and 220 mg/l added to stream water samples resulted in >66% mineralization of the C-2 carbon in six days; 2.2 μ g/l and 22 ng/l of 2.4-D resulted in <10% mineralization in eight days [36].

Process considerations

Which bioremediation method is appropriate? After determining that biological treatment of a waste material is possible, the appropriate technology depends on the solubility, volatility, and sorptive ability of the contaminant, the location and extent of contamination, the hydrogeology of the site, and the goal of the remediation project, i.e., source remediation or plume control.

A soluble chemical lends itself to *in situ* bioremediation; a volatile chemical is most suited to bioventing; a highly sorbed chemical might best be treated with land farming, in a liquid-solids contactor reactor, or by soil washing followed with biological treatment of the wash. If a site is contaminated with volatile organic compounds (VOCs), any treatment involving excavation or venting will lead to air emissions and another instance of cross-media contamination.

If the contamination is located in the vadose zone, the methods of choice are bioventing and land treatment. If the contamination is a light, non-aqueous phase liquid (NAPL) located in the capillary fringe, bioventing and land treatment may be the best choices. *In situ* bioremediation of soluble constituents in the vadose zone might be possible if the water table can be elevated, as was performed at an aviation gasoline spill site in Traverse City, MI [1]. Unfortunately, *in situ* biorestoration is less effective for the treatment of NAPLs present in pools or trapped as residuals in soil pores [37,38]. The dense NAPLs pose extraordinary problems with source detection and delineation; extraction and remediation are especially problematic. Vacuum enhanced pumping (nonbiological) has shown promise for increasing the amount of dense NAPL removed from a shallow aquifer [39]. Recovery and above ground treatment of the contaminant in a bioreactor might be feasible, as might excavation and land treatment, but depth of the contaminant source will limit the utility of this option.

Subsurface hydraulic conductivity is of paramount importance for *in situ* bioremediation. Injection and transport of nutrients is difficult in aquifers with hydraulic conductivity lower than 10^{-4} cm/s [40]. Lower permeability aquifers may be amenable to *in situ* biorestoration, but the time and cost of a project will increase if the site is heavily contaminated [41]. The types of aquifer media and associated soil chemistry are also important for bioventing and *in situ* treatment. A clayey formation will inhibit vapor extraction and therefore, bioventing. Reduced iron (Fe²⁺) can precipitate from solution as ferric oxide when oxygen is added to the subsurface. High concentrations of dissolved iron and manganese, coupled with the presence of iron-fixing bacteria have led to the plugging of injection wells [16]. In a field experiment at a site contaminated with jet fuel, excessive oxygen gas production was partially attributed to interaction of hydrogen peroxide with soil minerals [41]. Aquifer heterogeneity complicates all remediation processes, including *in situ* treatment and bioventing.

Cost considerations

How costly are the bioremediation technologies? The cost of a bioremediation project varies with the methodologies used and on the location, quantity, and characteristics of the wastes present. If ground water is impacted, an average of \$100 is spent for each gallon of gasoline spilled, according to a 1988 study by the American Petroleum Institute [43].

Bioremediation of contaminated soils is inexpensive compared to some of the non-biological methods. Conventional land treatment costs range from \$50 to $80/yd^3$ [5]. For 6,000 yd³ (4,400 m³) of a soil contaminated with diesel fuel, land treatment in an unlined system requiring no excavation costs about $33/yd^3$; land treatment with excavation and a liner costs about $74/yd^3$ [6]. Composting fees average $100/yd^3$ [5]. Treatment costs in a liquid-solids contactor reactor range from $100-150/yd^3$ [5]. If contaminated off gas treatment can

be eliminated, bioventing could cost only $125-160/yd^3$. The costs of other hazardous waste remediation methodologies can be substantially greater than bioremediation. Two estimates for disposal of soil contaminated with fuel in a sanitary landfill are $300-500/yd^3$ [44] and $200-300/yd^3$ [45]; excavation can be 100% effective in removing the source of contamination, although volatile organic compounds (VOCs) will be released in the process. Incineration is capable of removing 99.99% of gasoline components found in soil [45]. Incineration using a mobile, transportable rotary kiln which is capable of reaching 1200-1800°F (650-1000°C) can cost from \$100-\$500/ton (\$223-\$1115/ vd^3) [46]; if less than 20,000 vd^3 of soil are treated, costs increase considerably [45]. Incineration using a new technology such as a plasma arc torch which can achieve temperatures greater than 10,000°F (5500°C) costs \$800-\$2000/ ton (\$1784-\$4460/yd³) [46]. These estimates do not include the costs of excavation and transportation, which can be considerable. Excavation coupled with a standard incineration method can cost from $1000-2000/yd^3$ [44]. Enhanced volatilization of VOCs (non-biological) is most effective when 15.000-18.000 tons of soil are treated; its cost is \$245-\$320/yd³ [45]. Soil washing is theoretically capable of removing 99% of VOCs, but it is less efficient if the soil contains much silt and clay; the cost ranges from 150-200/vd³ [45].

Bioremediation of fuel contaminated ground water is cost-effective, but the process is often unable to achieve the low effluent concentrations achieved by some other methods. An *in situ* bioremediation scheme can cost \$8 to \$15 per pound of contaminant [5] or $66-123/vd^3$ of material treated [45]. When hydrogen peroxide is included in an in situ treatment project, the average increase in cost is \$50/gal (\$6/lb or \$13.2/kg) of contaminant [47]. Effluent concentrations at the mg/l level are possible, but it is difficult to realize an effluent concentration at the $\mu g/l$ level [45]. Above-ground biological treatment of contaminated ground water cost estimates range from \$30-\$40/yd³ treated or \$4-\$6/lb of contaminant degraded [45]. A bioreactor capable of treating a waste stream of 25 gpm (95 l/min) can cost \$60,000 to \$80,000; an additional \$15,000 to \$25,000 may be required for bench-scale development [48]. In comparison, air stripping of VOCs in ground water can cost \$50,000-100,000 for a system or 5-25/1000 gal ($1.3-6.5/m^3$) treated, remove 99-99.5% of the VOCs, and either release VOCs to the atmosphere or require a vapor phase treatment, which doubles the cost of the method; the effluent concentration achieved is not less than 5 μ g/l [45]. Granular activated carbon adsorption can achieve effluent concentrations less than 5 g/l and can cost \$300,000-\$400,000 to install; operation and maintenance can range from \$25,000-\$30,000/y [45]. Pump-and-treat for plume management is a proven method; a 10 gpm extraction system with a column aerator, but no vapor phase treatment, used for 100 years would cost \$285/gal for a 1000 gallon spill or \$28.50/gal for a 10,000 gallon spill [47].

Concluding remarks

The diversity of bioremediation technologies ensures that one or more of them may be suitable for at least part of a remediation scheme. With our increased understanding of subsurface processes, we are shifting from the treatment of effects, i.e., treatment of contaminated ground water, to the treatment of contaminant sources. If one considers the failure of previously applied treatments, such as pump-and-treat, to remove the source of contamination [38] and recent regulatory requirements, such as the ban on land disposal of hazardous waste, it is almost certain that bioremediation technologies will gain prominence. In fiscal year (FY) 1984, only one Superfund Record of Decision (ROD) utilized bioremediation technologies as a method of source control; the number increased to seven in both FY 1988 and 1989. As shown in Fig. 1, bioremediation technologies were used in only 7.5% of 93 treatment methods specified at Superfund sites in FY 1989 [49]. The physical or chemical methods are more widely used, possibly because biological treatments are more sitespecific and often require more knowledge about a site than is necessary when using other remediation methods. In 1987, the first year of the SITE program, one biological treatment method was under investigation out of a total of eight methods evaluated; in 1988, there were no bioremediation methods included in the seven technologies studied; in 1989, the number had risen to 5 out of 16 [34]. The overall cost-effectiveness of bioremediation and its potential to eliminate contaminants without causing cross-media pollution indicate a continuing need for knowledge and research. Indeed, an expanding requirement for bioremediation technologies can be seen looming in the future.

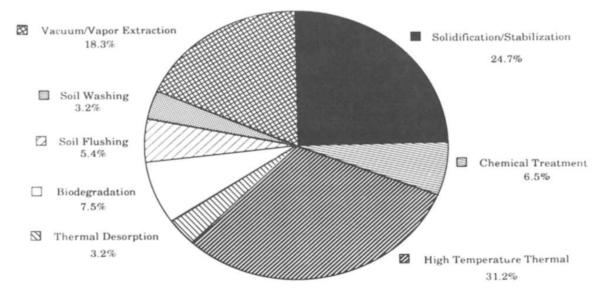


Fig. 1. Technologies used for remediation of Superfund sites in Fiscal Year 1989.

Acknowledgment

The authors thank Dr. Hans Stroo of ReTeC, Kent, WA and Ms. Elaine Higgins of Groundwater Services, Inc., Houston, TX for their assistance in gathering the cost estimates. Although the research described in this article was supported by EPA through Assistance Agreement No. CR-812808 to Rice University, it has not been subjected to Agency review and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

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