Soil heating and optimized nutrient addition for accelerating bioremediation in cold climates

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ABSTRACT. Low temperatures severely limit the rate of bioremediation in cold climates. The goal of this study was to investigate the effect of temperature on bioremediation rates and to optimize the nutrient supply in order to accelerate bioremediation. Samples of diesel-contaminated soil from two Alaskan sites were incubated in the laboratory at different temperatures (1, 6, or 20°C) with varying nutrient levels (0, 100, 300, or 800 mg nitrogen per kg dry soil). As expected, biodegradation was fastest at higher temperatures. However, after approximately two months of treatment, the cumulative amount of CO_2 produced at a temperature of 6°C could reach 75% of the amount produced at 20°C. Based on this data, researchers concluded that a temperature of 6°C was sufficient to achieve efficient treatment if all other potentially limiting factors — such as oxygen, water content, and nutrient supply — were addressed. A cost-effective process based on passive solar soil heating is proposed to raise the soil temperature and thereby improve the bioremediation rate. Furthermore, in field applications, an even distribution of nutrients cannot be taken for granted, especially when granulized fertilizer is applied; this method often results in suboptimal local concentrations (too high or too low). Particularly in cold climates with a short growth season, it is important to utilize this short time-window efficiently. The second part of the study explored improving the method of fertilizer application (granular versus liquid application, with and without irrigation) to achieve a homogeneous nutrient distribution. It was found that irrigation can substantially improve the distribution of nitrogen, thereby increasing bioremediation rates and shortening treatment times by as much as 50%.

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Introduction

Bioremediation has proven to be an efficient method for degrading a variety of organic pollutants in soil, and it is commonly used worldwide for this purpose. Bioremediation on site provides a viable treatment option, particularly in remote rural locations where it is not economically feasible to haul the excavated contaminated soil to a specialized treatment facility.

Although it has been shown that cold-adapted hydrocarbon-degrading micro-organisms exist in Arctic (Rike and others 2003, Whyte and others 2001) and Antarctic (Aislabie and others 2000) environments, bioremediation faces a major challenge in cold climates: low temperatures significantly slow the decontamination process. Previous studies have shown that increasing temperatures lead to increasing biodegradation rates (Ferguson and others 2003b; Mohn and Stewart 2000), even though biodegradation may still proceed slowly at sub-zero temperatures, according to Rike and others (2003), who noted that the measured partial pressure of oxygen in the contaminated soil was lower than predicted from modeling abiotic processes, which may be explained by microbial oxygen consumption. Walworth and others (2001) noted that rates increased up to a temperature of approximately 20–30°C and decreased at higher temperatures. These findings indicate that biodegradation of organic pollutants in Arctic soils can require exceedingly long treatment times. If the spill is situated in an environmentally sensitive area, these timeframes can be unacceptable.

Consequently, it is necessary to develop strategies for an improvement of bioremediation performance in cold climates. Soil heating and insulation can be used to accelerate biodegradation rates, as demonstrated in stockpile systems with electric heating and insulation at a site in Prudhoe Bay, Alaska (Filler and others 2001). However, electric heating is a major cost factor for such processes. Especially in remote locations, electric power produced by generators is very costly and only available in limited quantities. From an economic standpoint, a process that utilizes only a minimal amount of electric energy would vastly improve the use of bioremediation in such areas.

One major aspect of this work focused on determining how temperature affects bioremediation of diesel-contaminated sites. A subtask was to develop a design concept for soil heating appropriate for Arctic locations. Passive solar heating is proposed as a cost-efficient method for achieving a moderate temperature

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increase, one sufficient to enable effective contaminant biodegradation.

The second main focus of this study concerned optimized nutrient addition. Providing nitrogen and phosphorus compounds is important to bioremediation even in moderate climates; in cold climates, where the growing season is much shorter, optimizing applications of these compounds is essential.

Walworth and others (2003) noted a synergistic interaction of soil heating and nutrient supply, and several studies, including one by Swannell and others (1995), reported on the use of different fertilizer types with a focus on nutrient release and prevention of 'washout' (for example, for shoreline applications), but the literature search for this study revealed no studies on the effect of fertilizer application methods on nutrient distribution and soil bioremediation rates in soil. The present work presents data to close this gap.

Materials and methods

Sampling sites and sample characteristics

Diesel-contaminated soil was obtained from two Alaska Department of Transportation and Public Utilities (DOT) maintenance stations. The first site, located in Tok, at the junction of the Alaska Highway and the Tok cutoff road to the Glenn Highway (63.3°N, 143.0°W), represents a moderate sub-Arctic climate with an average low temperature in January of –36°C, an average high temperature in July of 22°C, more than five months with average temperatures above 5°C, and a mean annual precipitation of approximately 220 mm. The second site, at Sag River (68.8°N, 148.9°W) on mile 306 of the Dalton Highway north of the Brooks Range, is characterized by an Arctic climate, where average temperatures are only for approximately 2–3 months above 5°C and never reach 20°C. The Sag River sample results from material excavated in the course of a tank removal. Samples from both sites were taken in the fall of 2002 and stored in air-tight containers at 4°C until use. Both materials consisted of a sand-gravel mixture. After removing stones larger than 20 mm, the grain-size distribution was very similar for both samples, with approximately 20% by weight finer than 1 mm and 30% coarser than 10 mm.

The initial water content for the Sag River sample was 7.8 mass percent of water per wet sample weight; that for Tok was originally 3.8%. Since this water content is suboptimal, water was added up to a water content of 8.4% for the first set of experiments and 7.1% for the second set of experiments (initial water content before irrigation). In experiments without irrigation, the water content was maintained at the initial level.

The soil pH values remained between pH 6 and pH 8 for all samples. Therefore no pH adjustment was required.

Degradation experiment

For each site, samples were sieved to eliminate stones and mixed for homogenization. Due to low contamination levels, the samples were generally spiked with 2.2 g diesel kg^{-1} soil; a small number of samples from the Sag River site were spiked with 4.4 g diesel kg^{-1} soil in order to investigate whether the substrate concentration was rate-limiting. Measured diesel concentrations at the beginning of the experiment after spiking with 2.2 g diesel kg^{-1} soil were 1180 and 1220 mg kg^{-1} for the Sag River and Tok samples, respectively. Parallel identical experiments were performed for the Tok and Sag River samples. Due to space limitations, only few data are shown for Tok, while most data reported here are for Sag River, since this site features an Arctic climate. Additional data for the Tok site from the first set of experiments are reported in a prior publication (Niemeyer and Schiewer 2003).

Since prior characterization of the soil showed low levels of exchangeable nitrogen (nitrate, nitrite, and ammonia) after extraction with KCl, fertilizer was added. For each site, two sets of experiments were conducted, in which the method of nutrient application differed. For each set of experiments, 1 kg of soil (dry wt) was placed in a 2 L sealed container and incubated at controlled temperatures (1, 6, and 20°C).

In the first set, the nitrogen concentrations were varied (0, 100, 200, and 800 mg kg^{-1} dry soil) using Schultz Greenhouse Professional™ (20+20+20). This fertilizer contained 20% nitrogen (4% ammonia-N, 6% nitrate-N, and 10% urea-N), 20% phosphate (as P_2O_5), and 20% potassium (as K_2O). Correspondingly, an initial concentration of 100 mg N kg^{-1} dry soil consisted of 20 mg ammonia-N kg^{-1} soil, 30 mg nitrate-N kg^{-1} soil, and 50 mg urea-N kg^{-1} soil. The fertilizer was applied as a powder and mixed well with the sample to achieve a homogeneous nutrient distribution.

In the second set of experiments, different methods of fertilizer application were compared, using a fertilizer concentration of 100 mg N kg^{-1} dry soil based on results of the first set of experiments. Commercial granular fertilizer (20+20+10) manufactured by UAP Northwest, Burlington, was applied without prior mixing with the soil. This fertilizer contained 20% total nitrogen (14.1% ammonia-N and 5.9% nitrate-N), 20% phosphate (as P_2O_5), and 10% potassium (as K_2O). Correspondingly, an initial concentration of 100 mg N kg^{-1} dry soil consisted of 70.5 mg ammonia-N kg^{-1} soil and 29.5 mg nitrate-N kg^{-1} soil. The fertilizer was applied either as granules distributed in the top centimeter of the soil or dissolved in water (50 g of fertilizer L^{-1}), of which 10 ml were equally distributed on the soil surface, to simulate fertilization by irrigation. In either case the amount of fertilizer applied was 0.5 g kg^{-1} dry soil.

A further factor investigated in the second set was the effect of subsequent irrigation. For this purpose, the set-up for half of the tests was modified. A grid was placed 5 cm above the bottom of the container and covered with a thin layer of geotextile, and then soil was added to the container. This set-up allowed water applied by irrigation to drain into a reservoir without saturating the soil. For removing water from the base of the container, a plastic tube was installed across the soil layer to the bottom of the

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container; water was withdrawn using a syringe connected to the plastic tube and reused for irrigation. Two irrigation events took place within a two-week period while the jars were opened for respiration measurements.

Determination of respiration rate, diesel range organics, and nutrient concentrations

In order to determine the amount of carbon dioxide produced — which is a product of hydrocarbon degradation and indicates the microbial activity — a beaker with CO_2-absorbing NaOH solution was included in the incubation containers. The beakers were periodically removed and replaced. Opening the jars for this purpose also served to replenish oxygen and maintain aerobic conditions. The CO_2 removed by the NaOH solution via formation of carbonate was precipitated as barium carbonate by adding barium chloride. The amount of unused NaOH was determined by titration with HCl solution of a known molarity. The amount of CO_2 absorbed, and thereby the respiration rate, can be calculated from the HCl consumption (Page and others 1982).

To confirm that the measured respiration was linked to hydrocarbon degradation, selected samples were analyzed for diesel range organics (DRO) by Northern Testing Laboratory (Anchorage, Alaska) using GC/FID standard method AK102 after storage in tightly sealed containers at –20°C.

Nutrient concentrations were determined in triplicate with a spectrophotometer (Hach model DREL/2010). Samples were processed by extracting 10 g wet soil with 100 mL 1 M KCl solution (Page and others 1982), then centrifuging the extract. The extracted supernatant was analyzed for nitrate, nitrite, and ammonia.

Results and discussion

Effect of temperature on respiration

Figure 1 shows the cumulative amount of CO_2 produced up to any given time for the Sag River sample; these samples were incubated at different temperatures and with different initial diesel concentrations for a nutrient concentration of 100 mg N kg^{-1} dry soil (fertilizer powder homogeneously distributed, first set of experiments). It is apparent that the initial respiration rate (that is, the slope of the curves) is considerably higher at 20°C than at 6° or 1°C. After a period of 11 days, the CO_2 production at 20°C (2300 mg CO_2 kg^{-1} dry soil for an initial concentration of 2.2 g diesel kg^{-1} dry soil) is about five times as high as the CO_2 production at 6°C (480 mg CO_2 kg^{-1} dry soil for 2.2 g diesel kg^{-1}). However, after a period of 58 days, for an initial concentration of 2.2 g diesel kg^{-1} dry soil, the CO_2 production at 6°C (2600 mg CO_2 kg^{-1} dry soil) reaches 76% of that observed at 20°C (3400 mg CO_2 kg^{-1} dry soil). This data indicates that over time the samples incubated at 6°C 'catch up' to those incubated at 20°C. Over a timeframe of months, a temperature of 6°C is sufficient to achieve successful biodegradation (see further discussion below). At a temperature of 1°C, however, cumulative respiration after 58 days was less than half of that for 20°C. Detailed respiration data for the Tok and Sag River sites (including two different samples from Sag River) at varying nutrient concentrations and temperatures are reported in Niemeyer and Schiewer (2003).

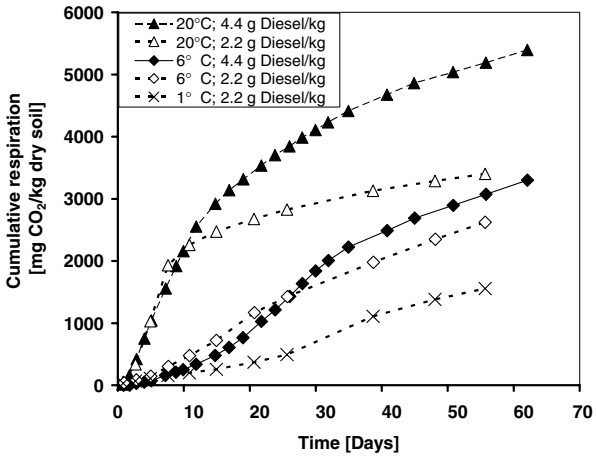


Fig. 1. Effect of temperature (1, 6, and 20°C) and substrate concentration (diesel spikes 2.2 and 4.4 g kg^{-1} dry soil) on cumulative respiration for Sag River soil sample at a nutrient concentration of 100 mg N kg^{-1} dry soil with homogeneous nutrient distribution for a water content of 7.8%.

Effect of substrate concentration on respiration

Note that in Figure 1, for the samples with the lower diesel concentration (2.2 g kg^{-1}), the respiration rate (slope of curve) at 20°C declines drastically after about 10 days, whereas the rate at 6°C still increases and ultimately surpasses that at 20°C. One explanation for this contrast can be found by comparing the data obtained for the two diesel concentrations at 20°C. For the first 10 days, the rates are about equal for high (4.4 g kg^{-1}) and low (2.2 g kg^{-1}) diesel concentrations. At this point, the rate is not affected by the substrate concentration. At the lower concentration, the rate becomes substrate-limited around day 10, and the curves diverge. One interpretation is that the easily degradable fractions are already depleted after 10 days, and the remaining degradation progresses more slowly. At a concentration of 4.4 g kg^{-1}, the rate is not yet limited by the available substrate and therefore remains higher for a longer time period. At 6°C the curves for the two concentrations do not diverge markedly, since substrate limitations do not play as significant a role as temperature limitations. It is, however, possible that substrate limitations would occur at 6°C after time periods of more than two months, when the majority of the substrate, particularly the easily degradable fractions, have been degraded.

Temperature can affect mineralization rates via several mechanisms. While on the one hand microbial growth and metabolic rates can be slowed down at low temperatures, physiochemical processes may also contribute to lower rates. Reduced mobility and bioavailability of petroleum hydrocarbons (for example, due to solidification of some

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Table 1. Comparison of diesel removal (mg of diesel per kg of dry soil) and cumulative CO_2 production (mg of CO_2 per kg of dry soil) after 56 days for samples from Sag River and Tok, with a fertilizer concentration of 300 mg N kg^{-1} dry soil.

	DRO Initial (mg g^{-1})	DRO final (mg g^{-1})	DRO removed (mg g^{-1})	DRO removed (%)	CO_2 produced (mg g^{-1})	CO_2 produced per DRO removed
Sag 20°C	1180	120	1060	90	4050	3.8
Sag 6°C	1180	310	870	74	2870	3.3
Tok 20°C	1220	78	1142	93	2600	2.3
Tok 6°C	1220	390	830	68	1400	1.7

diesel components) can indirectly contribute to lower microbial mineralization of hydrocarbons.

Comparison of DRO degradation and CO_2 production

The DRO were determined initially and after 56 days for the samples with 300 mg N kg^{-1} dry soil and 2.2 g diesel kg^{-1}. After 56 days, the DRO concentration at 20°C was 120 mg kg^{-1} dry soil, 10% of the initial contamination, whereas at 6°C it was 310 mg kg^{-1} dry soil, or 26% of the initial value of 1180 mg kg^{-1} dry soil for Sag River sample. Similar percentages of reduction were achieved for the Tok sample, as shown in Table 1. Even at 6°C, it was possible to achieve the threshold limit for DRO in contaminated soil (500 mg kg^{-1} in most areas in Alaska) within two months for both Sag River and Tok.

The measured initial diesel concentration was considerably lower than expected based on the spike amount. This reduction might be due to evaporation of the more volatile fraction of diesel, and/or by limitations of the DRO-estimation methods, such as incomplete extraction. It is possible that the actual amount of diesel degraded could be higher than that measured in this study. While non-homogeneous diesel distribution in the soil may reduce the accuracy of DRO determination, the systematically lower recoveries observed in this study cannot be explained by this factor.

Based on the stoichiometry of hydrocarbon degradation ($2\ C_{16}H_{34} + 49\ O_2 \rightarrow 32\ CO_2 + 34\ H_2O$), approximately 3.1 mg CO_2 are produced for the mineralization of 1 mg diesel, assuming that all of the carbon source is mineralized and not used for building up new biomass. The CO_2 produced after 56 days for 300 mg N kg^{-1} is shown in Table 1. Based on these data, a diesel consumption of 1306 mg at 20°C and of 926 mg at 6°C can be expected for Sag River. This matches the magnitude of the measured DRO degradation, which was 1060 mg at 20°C and 870 mg at 6°C. The observed ratio of CO_2 produced per diesel degraded (last column of Table 1) fluctuates around the theoretical value. Compared to Sag River, a lower CO_2/diesel ratio was observed for Tok, indicating that a lower fraction of the diesel was converted to CO_2. Possibly a higher amount of the carbon source was used for the synthesis of new biomass in the Tok samples. The percentage of substrate used for synthesis of new biomass may vary depending on the local microbial community and environmental conditions. The comparison of diesel degradation and CO_2 production confirmed that other degradation processes did not yield significant quantities of CO_2, since the CO_2/diesel ratios are close to the expected value. Considering that the soil was gravel with virtually no organic content, it can be assumed that diesel was the main substrate and therefore the main source of carbon for CO_2 production. These comparisons confirm that CO_2 production is a good indicator of diesel degradation, as performed in this study. Respiration rates can be measured more easily and reliably than the diesel concentration itself.

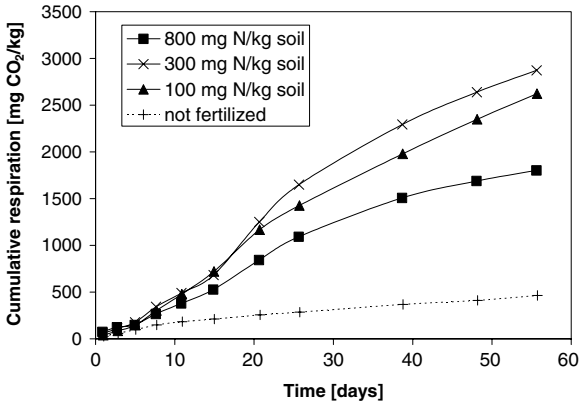


Fig. 2. Effect of fertilizer concentration (homogeneously distributed) on cumulative respiration for the Sag River sample at 6°C with a water content of 7.8%.

Effect of nutrient concentration on respiration

Figure 2 shows cumulative respiration at different nutrient concentrations for the Sag River sample at 6°C for homogeneous nutrient distribution (first set of experiments). The fertilizer was applied as a powder and mixed well with the soil to achieve a homogeneous distribution. It appears that concentrations of 300 and 100 mg N kg^{-1} soil are most favorable. At 800 mg N kg^{-1}, the respiration was already lower than for 100 or 300 mg N kg^{-1}. This drop in respiration can be ascribed to reduced water availability at high solute concentrations. Similar results were found for samples incubated at 20°C (Niemeyer and Schiewer 2003).

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Other researchers have similarly noted reduced biodegradation rates at too high nutrient concentrations. Ferguson and others (2003a) found that nutrient dosages according to the Redfield ratio of C:N:P = 106:16:1, which is based on complete incorporation of the carbon source into biomass, could lead to severe inhibition of hydrocarbon mineralization due to a nutrient overdose. Lower nutrient dosages were sufficient for hydrocarbon degradation, since a large part of the carbon is not incorporated into new biomass but mineralized. The same authors determined a nutrient dosage of 508–1570 mg N kg^{-1} of soil-water as optimal, relating the nutrient dosage to the water content (rather than to the dry soil weight) to better account for osmotic potentials.

Walworth and others (2003) noted that excess nutrients can inhibit degradation. Bacteria in coarse-grained soils such as sands and gravel are particularly sensitive to osmotic shock, since the water-holding capacity in such soils is generally low. To prevent negative effects on microbial activity for nitrogen concentrations exceeding 2500 mg N kg^{-1} soil-water, where osmotic shock can occur, slow-release fertilizers may be used. Higher rates were observed for cod bone-meal (a slow-release fertilizer) compared to the readily soluble di-ammonium phosphate, each dosed at 7000 mg N kg^{-1} soil-water (Walworth and others 2003). In the present study, nutrient concentrations of 800 mg N kg^{-1} dry soil, for which lower respiration was observed, correspond to 10,256 mg N kg^{-1} soil-water for the Sag River soil with a water content of 7.8% and 9524 mg N kg^{-1} soil-water for the Tok soil with a water content of 8.4% (after adjustment). These values are clearly above the level of 2500 mg N kg^{-1} at which osmotic limitations were noted by Walworth (2003), supporting the statement that reduced water availability may have been the cause for lower respiration at 800 mg N kg^{-1} dry soil observed here. However, for the next lower fertilization level of 300 mg N kg^{-1} dry soil, high respiration rates were achieved even though the fertilizer content per soil-water was 3846 mg N kg^{-1} soil-water for Sag River and 3571 mg N kg^{-1} soil-water for Tok, which is already in the range where osmotic shock effects were observed by Walworth (2003).

In the unfertilized sample, respiration was very low. These results highlight the necessity of providing nutrients, especially for a soil such as gravel with low initial nitrogen and phosphorus concentrations. Based on these results, all further experiments were conducted with a nutrient concentration of 100 mg N kg^{-1} dry soil.

Effect of nutrient application method on respiration rate

In field applications, a homogeneous nutrient distribution cannot be guaranteed, and it is impossible to achieve an optimal concentration throughout the soil pile; local concentrations differ from the average concentrations, leading to suboptimal (either too high or too low) concentrations. In the second set of experiments, different nutrient application methods were compared. To simulate field conditions, fertilizer (100 mg N kg^{-1}) was either applied as granules or as a concentrated liquid only to the surface of the sample, without vertical mixing (second set of experiments).

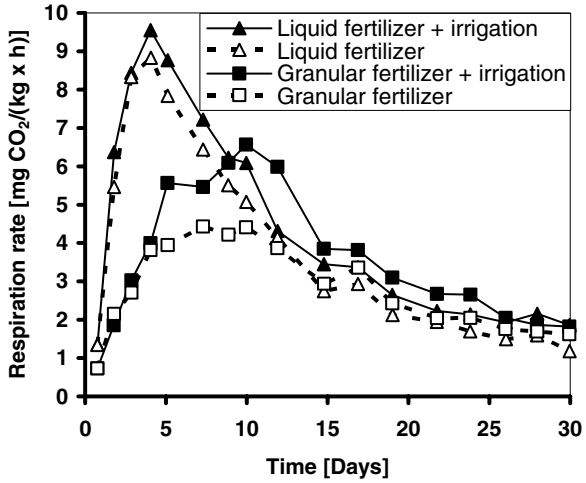


Fig. 3. Effect of fertilizer application mode (liquid or granular fertilizer, with or without irrigation) on respiration rates of the Sag River soil sample at 20°C for 100 mg N kg^{-1} dry soil. The water content without irrigation was 7.8% and with irrigation 8.9 or 8.6%, respectively, for liquid and granular fertilizer.

Figure 3 shows the respiration rate, in mg CO_2 kg^{-1} soil, as a function of time. Liquid fertilizer clearly leads to higher rates for the first 10 days, suggesting that liquid application yields a better nutrient distribution. Particularly for granular application, but also to a certain extent for liquid application, irrigation increases respiration rates, again likely due to better nutrient distribution. However, after about two weeks the respiration rates were similar for all application methods.

Effect of nutrient application method on nutrient distribution

To confirm that the increased respiration rates obtained by irrigation were linked to a more homogeneous nutrient distribution, the concentrations of nitrate and ammonia were determined in the upper and lower soil layers. Nutrient concentrations for several experiments with liquid fertilizer application (partially shown in Fig. 3), were averaged to obtain a representative comparison between upper- and lower-layer nutrient levels. For this purpose, results from experiments for both sites (Sag River and Tok) at both temperatures (6°C and 20°C) were used, such that each value represented in Figure 4 is the average for four experiments. Figure 4 shows the average concentrations (different temperatures, different sampling locations, triplicate samples) for liquid application with (twice) or without irrigation (second set of experiments). Soil-water contents are listed in Table 2. In either case, the concentration in the bottom soil layer was lower, showing that even after 18 days a vertical concentration gradient still existed. However, with irrigation, the concentrations

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Table 2. Comparison of the water content of the different samples before and after irrigation (measured 18 days after the start of the experiment, as the average of all four experiments under the same conditions, that is, liquid and granular application each at 6°C and 20°C).

Water content (%)	Before irrigation	After irrigation	Water content change
Sag River upper layer	7.8	8.1	+0.3
Sag River lower layer	7.8	10.0	+2.2
Tok upper layer	7.1	9.1	+2.0
Tok lower layer	7.1	10.9	+3.8

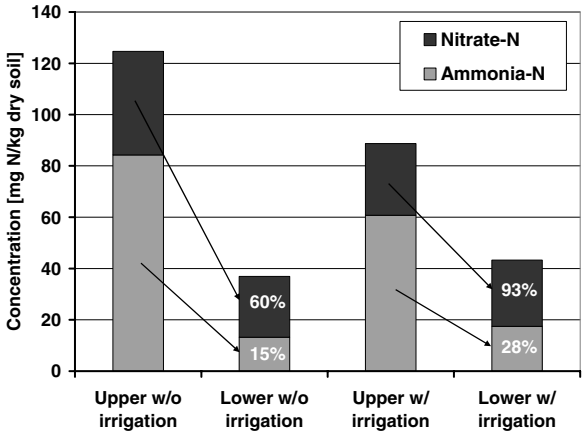


Fig. 4. Comparison of average (Tok and Sag River, each at 6 and 20°C) nitrate and ammonia concentrations in the upper and lower soil layers with or without irrigation after 18 days, for 100 mg N kg^{-1} applied as liquid fertilizer. Without or before irrigation, the average water content was 7.5%. After irrigation, the average water contents in the upper and lower layer were 8.8% and 10.0%, respectively.

in the lower layer were more similar to those in the upper layer. Nitrate in the lower layer was 93% of the upper-layer concentration with irrigation, but only 60% without irrigation. Ammonia was not as well distributed; the concentration in the lower layer was 28% with and 15% without irrigation, compared to the upper layer. Generally ammonia is less mobile than nitrate and sorbs more strongly to soil due to its positive charge.

Effect of nutrient application method and temperature on respiration

Figure 5 shows the cumulative CO_2 production for granular fertilizer application at different temperatures, with or without irrigation (second set of experiments) compared to homogeneous nutrient distribution (first set of experiments). At either temperature, irrigation increases CO_2 production. However, this effect is more noticeable at 20°C than at 6°C. Since metabolic activity and microbial growth are higher at 20°C than at 6°C, a higher nutrient usage rate can be expected at higher temperatures, leading to nutrient limitation if sufficient nutrients are not readily available. This may be the

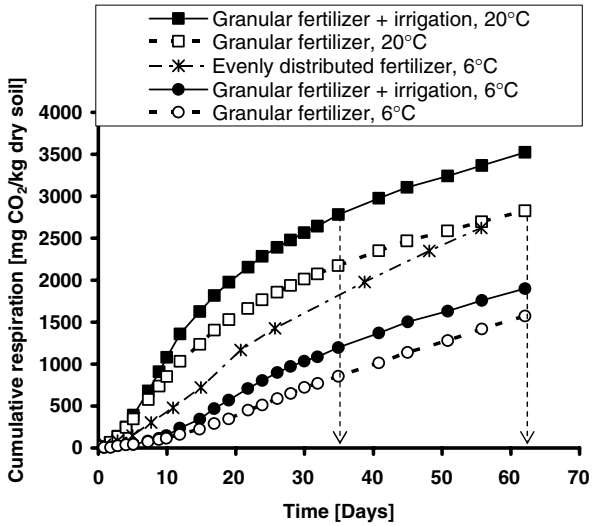


Fig. 5. Effect of temperature and irrigation on cumulative respiration of Sag River sample for 100 mg N kg^{-1} dry soil. The water content without irrigation was 7.8%, and with irrigation 8.6% or 9.1%, respectively, for 20°C and 6°C.

case here for the 20°C samples without irrigation. Other researchers have similarly found that less fertilization is required at lower temperatures.

At 20°C, irrigation can speed up biodegradation by as much as four weeks. The 62-day period required to reach a final CO_2 production of 2830 mg kg^{-1} without irrigation is halved by irrigation: after only 35 days 2780 mg CO_2 kg^{-1} dry soil are produced.

For comparison, one data series for 6°C is included with fertilizer homogeneously distributed as a powder. At the end of two months, samples with homogeneous fertilizer distribution at 6°C and samples at 20°C with granular application without irrigation show the same amount of CO_2 produced. These results suggest that, over a two-month timeframe, fertilizer distribution can be as important as temperature.

Comparison of Sag River and Tok samples with respect to effectiveness of irrigation

While irrigation was very successful in increasing biodegradation for the Sag River sample, irrigation actually had a slight negative effect on degradation in the Tok sample, as shown in Figure 6 (second set of experiments). These results can be explained by differences in the water content of the two samples. Average soil-water contents (for liquid and granular fertilizer application each at 6°C and 20°C) are listed in Table 2. While the water content for Sag River was favorable, the Tok sample had too high a water content after irrigation, a factor that negatively impacted the degradation rates. Table 2 shows that the water content in the Tok sample increased more in both layers — but particularly in the lower layer — than in the Sag River sample, even though the irrigation procedure and amount were the same for both samples. Porosity and sand grain structure are likely the cause for this. In spite of both being sand/gravel mixtures with similar grain-size distribution,

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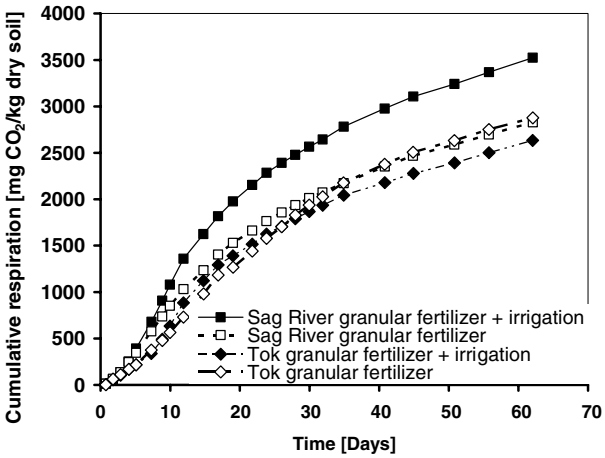


Fig. 6. Comparison of cumulative respiration for Tok and Sag River samples under different fertilization regimes at 20°C for 100 mg N kg^{-1} dry soil applied as granular fertilizer with or without irrigation. For Sag River, the water content was 7.8% without and 8.6% with irrigation; for Tok, the water content was 7.1% without and 10.0% with irrigation.

the Tok soil appeared to have a lower porosity, and became more easily water-saturated, which may have limited the oxygen mass transfer. While the Sag River soil consisted of crushed gravel with irregular shapes, the Tok natural gravel had more spherical grains. Clearly, it is important to take the physical soil characteristics and water content into consideration when deciding whether or not to use irrigation for improved nutrient distribution.

Implications for field applications

Since the experimental results show that it is not necessary to maintain temperatures as high as 20°C to achieve successful bioremediation, it is possible that a cost-efficient system for solar heat could be used to produce the lower temperatures sufficient for bioremediation in Arctic and sub-Arctic sites. A concept and design for treatment in a soil cell have been developed, but not yet tested. Key elements of the design appear in Figure 7, including a 'greenhouse' for soil heating, a ventilation system for oxygen supply and warm air intake, and an irrigation and leachate collection system to control moisture and provide nutrient solution.

According to this concept, the soil pile is located in a 'greenhouse' that captures solar radiation. A cost-effective greenhouse cover would be HDPE sheets supported by a simple structure (wood or ABS plastic pipe frame) to provide spacing between greenhouse cover and insulation, as well as between insulation and irrigation pipes.

Perforated pipes at the bottom of the pile serve for collection of leachate and for aeration. The collection pipes are at a gradient such that all leachate is collected in a storage tank. From this tank, the collected water may be reused for irrigation if necessary. For that purpose a sump pump pumps the water to a distribution system located just above the soil.

A thermostat-controlled vacuum pump connected to the same perforated pipes as serving for leachate collection (as common in bioremediation pile design) draws air from inside the greenhouse into the pile. Due to the greenhouse cover of the pile, leachate amounts are low, so that the presence of leachate will not impede air flow. The aeration system is activated when the air temperature inside the greenhouse is higher than the soil temperature. When the soil temperature exceeds the air temperature (as expected in the fall) only the minimum air circulation necessary to supply oxygen for remediation is provided.

The pile is covered with insulation in order to reduce heat loss when air temperatures are low. However, due to the ventilation system, the insulation layer does not impede heating of the soil when air temperatures within the greenhouse are higher. While insulation and greenhouse significantly reduce air convection, neither of these layers is sealed completely airtight. Therefore, it is nevertheless possible to draw fresh air from outside into

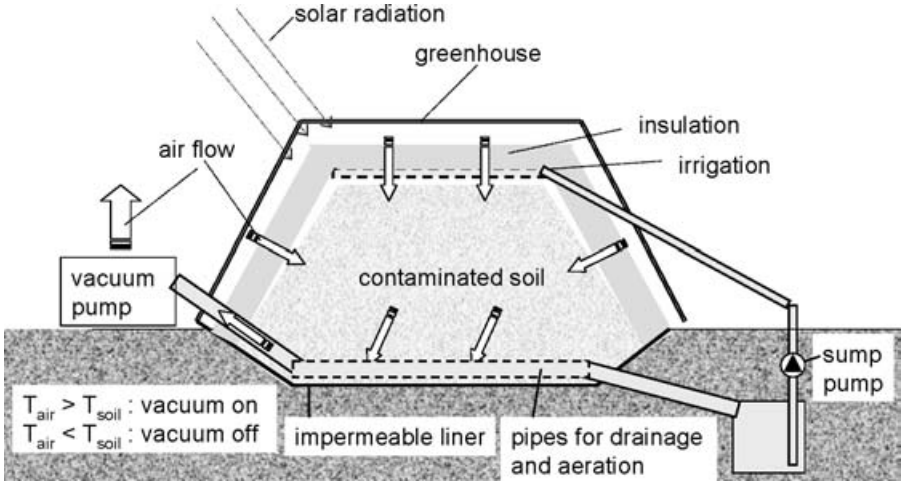


Fig. 7. Conceptual design of soil pile. Solar heat and insulation contribute to maintain higher soil temperatures.

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the greenhouse and warm air from the greenhouse into the biopile when the forced aeration system is activated. The design is intended to extend the treatment time period such that remediation can be achieved in one or two seasons.

At Sag River, average temperatures are only above 5°C for approximately 2–3 months, and never reach 20°C (ARC LTER 2004). Therefore soil heating by means of a greenhouse is expected to be very beneficial for bioremediation at this site. On the other hand, in Tok, where average temperatures of 5°C or higher are present for more than 5 months, soil heating may not be required. A factor favoring the feasibility of solar heating for Arctic locations is the high amount of solar radiation available in the Arctic summer, with periods of 24 hours per day of daylight.

Conclusions

Diesel degradation experiments at different temperatures showed that while respiration rates increase with temperature, at a temperature of 6°C, DRO concentrations can be reduced from initially 1180 mg kg^{-1} to 313 mg kg^{-1} within two months. This value is below the Alaska Arctic zone clean-up level of 500 mg kg^{-1} for cases where contamination is due to a diesel spill with low levels of benzene, toluene, ethylbenzene, and xylene (Department of Environmental Conservation 2000). Since degradation at 20°C slowed after 10 days due to rate-limiting substrate concentrations for a diesel concentration of 2.2 g kg^{-1} of dry soil, the cumulative respiration at 6°C eventually 'caught up' after two months, reaching 75% of the CO_2 production achieved at 20°C.

Rather than using expensive electric heat to raise temperatures for accelerating degradation, the authors propose passive heating by means of an 'intelligent greenhouse,' where air is circulated through an enclosed soil pile in the greenhouse when the air temperature is higher than the soil temperature.

Although conditions in field applications are often less optimal than in the laboratory (mass-transfer limitations, weathered contaminants), leading to longer treatment times than observed in laboratory experiments, it is likely that remediation can be achieved in one or two growing seasons, depending on local conditions. Prior investigations should still be performed for the specific contaminated site.

In experiments at different nutrient concentrations with homogeneous fertilizer distribution, the best performance was achieved for 300 and 100 mg N kg^{-1} dry soil. However, the rates were lower when the fertilizer was not evenly distributed but applied only at the top of the microcosm, simulating heterogeneous distribution as it occurs in field applications. Respiration rates for granular fertilizer were lower than for liquid fertilizer, which can be explained by less homogeneous nutrient distribution for granular application. For the Sag River sample, irrigation improved nutrient distribution, leading to higher respiration and significantly shortening treatment times. For the Tok sample, on the other hand, irrigation decreased respiration rates. This was explained by an increase of the water content beyond the optimum in soil with lower porosity. Therefore the benefit of better nutrient distribution and the potential negative impact of too high water contents have to be weighed when evaluating the use of irrigation.

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